

# POTENTIAL OF TiO<sub>2</sub>: PVP COMPOSITE FILM AS SENSING ELECTRODE FOR EGFET pH SENSOR

Nurbaya Zainal<sup>a,b</sup>, Nur Syahirah Kamarozaman<sup>a,b</sup>, Zurita Zulkifli<sup>a,b</sup>, Sukreen Hana Herman<sup>a,b,c\*</sup>

<sup>a</sup>Integrated Sensors Research Group, College of Engineering, University Teknologi MARA, 40450 Shah Alam, Selangor, Malaysia

<sup>b</sup>NANO-ElecTronic Centre, College of Electrical Engineering, School of Engineering, Universiti Teknologi MARA (UiTM), 40450 Shah Alam, Selangor, Malaysia

<sup>c</sup>Microwave Research Institute, Universiti Teknologi MARA, 40450 Shah Alam, Selangor, Malaysia

## Article history

Received

02 October 2023

Received in revised form

01 March 2024

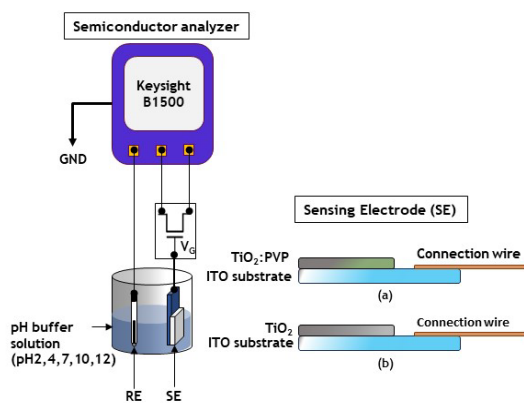
Accepted

03 March 2024

Published online

30 November 2024

\*Corresponding author  
hana1617@uitm.edu.my



## Abstract

A variety of uses for the commercial pH sensor have been established; one of them is figuring out the pH of plant soil. The accuracy of the commercial pH sensor and the long-term reliability of the device, however, appear to be questionable. In this paper, we demonstrate highly sensitive extended-gate field effect transistor (EGFET) pH sensors that have been examined in pH buffer solutions. The sol-gel spin coating process was employed for fabricating TiO<sub>2</sub> films and TiO<sub>2</sub>:PVP composite films on an indium-doped tin oxide (ITO)-coated glass substrate to form two distinct sensing electrodes for the EGFET. The pH detection performance was investigated using a variety of pH buffer solutions ranging from pH2 to pH12. In contrast to TiO<sub>2</sub> films (49.69 mV/pH), the TiO<sub>2</sub>:PVP composite films showed a higher sensitivity of 63.2 mV/pH. To evaluate the stability capabilities of the EGFET pH sensor, hysteresis and drift studies were conducted. It was found that the composite TiO<sub>2</sub>:PVP films were able to achieve a distinguished low hysteresis value of 10 mV and a low drift rate of 11 mV/h compared to TiO<sub>2</sub> films and previous reported studies. Further analysis of repeatability measurement was carried out three times for the composite films, resulting in a constant deviation of 0.002 that corresponded to low pH values.

**Keywords:** Sensing electrodes, composite film, pH sensor, sensitivity, stability

© 2024 Penerbit UTM Press. All rights reserved

## 1.0 INTRODUCTION

The establishment of electrochemical pH sensor performance has attracted numerous studies to replace the conventional pH glass sensor, and the extended-gate field effect transistor (EGFET) is one of the recent pH sensors. EGFET is widely known for its fast pH detection response and high reliability. Due to this factor, it has been used in a wide range of areas, such as waste water treatment, food quality monitoring, the medical industry, and many more [1]–[4]. This sensor is built with only two electrodes: a sensing electrode (SE) and a reference electrode (RE). The structure of the EGFET is represented by the isolation of the sensing electrode from the gate of the metal oxide semiconductor field effect transistor (MOSFET), while the connection can be completed by conductive wire like copper or aluminium wire.

The utilisation of MOSFET contributes to the complexity and cost reduction in the fabrication of EGFET sensors, which is beneficial for semiconductor manufacturing processes. It also offers flexibilities in variety material selection and types of method deposition as well. Recently, many nanomaterials have been tremendously studied; for instance, metal oxides such as titanium dioxide (TiO<sub>2</sub>) have the ability to produce highly sensitive EGFET pH sensors. It has a high degree of precision and a high sensitivity to H<sup>+</sup> ions, which is a factor in material selection [5]–[7]. There are already numerous published methods for producing TiO<sub>2</sub>-based thin films [8]–[10]. Due to its affordability, straightforwardness of use, and low-temperature thin film fabrication procedure, sol-gel spin coating was chosen among all other methods for depositing TiO<sub>2</sub>-based thin films.

Yang *et al.* produced a nano-flower-structured TiO<sub>2</sub> sensing membrane prepared by hydrothermal with a 46 mV/pH

sensitivity value and a great linear response of 0.9989 [11]. In another study, TiO<sub>2</sub> nanowires were produced using an E-beam evaporator, and the pH sensitivity was lower than expected, which is 32.65 mV/pH [12]. This might be due to the thick layer of TiO<sub>2</sub> films that existed beneath the nanowire structures that created sheet resistance at the interfacial layer between the metal surface and the sensing membrane layer. Furthermore, it could cause a discrepancy in charge transfer and affect the response time, especially for basic solutions. To overcome this, Chen *et al.* proposed functional superhydrophilicity TiO<sub>2</sub> films with pore networks to improve the time response; however, the films are facing other difficulties, such as sensing stability, that can have an impact on the real-time monitoring system [13]. Other than the sensitiveness towards OH<sup>-</sup> and H<sup>+</sup> ions, fast response and stability also play roles in a high-performance EGFET pH sensor. Considering this drawback, researchers are focusing on employing TiO<sub>2</sub> nanocomposite-based films as the pH sensing membrane to improve the sensing behaviour of the EGFET sensor.

Polyvinyl pyrrolidone (PVP), also commonly known as polyvidone or povidone, is a water-soluble polymer that has beneficial material properties such as flexibility, biodegradability, non-toxicity, and environmental stability. Furthermore, it has a low diffusion loss, which makes it the perfect polymer for composite materials that involve a substantial charge transport mechanism [14]. Due to these reasons, PVP has been developed as a sensing electrode in various ion detection applications due to its remarkable material properties [15], [16]. Recently, Kamarozaman *et al.* suggested that EGFET sensors with conducting polymers to be mixed with TiO<sub>2</sub> revealed very sensitive performance, as the sensor reached over Nernstian limits, i.e., 66.1 mV/pH [8]. Another work has been done using TiO<sub>2</sub>:PPI nanocomposite, which was initiated by highly stable SE by taking into account its low drift rate [17]. The combination of organic and inorganic composites can be aptly considered to create unique properties such as high-selectivity EGFET pH sensors, which favour biosensor applications as well [2], [18]. Table 1 depicts the pH sensitivity value of TiO<sub>2</sub>-based thin films.

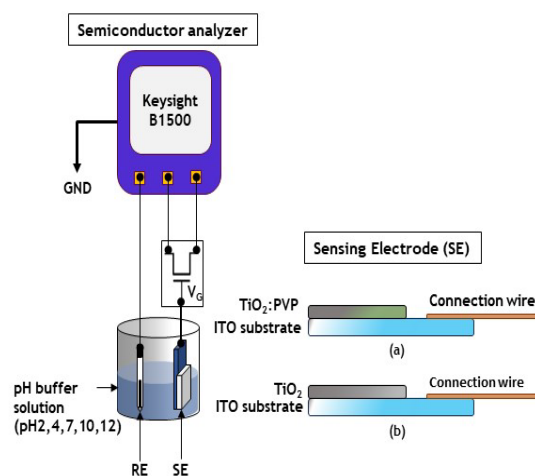
**Table 1** Previous studies on TiO<sub>2</sub> films-based pH sensitivity

No.	Sensing Electrode	Sensitivity Value (mV/pH)	Reference
1	TiO <sub>2</sub> Nano-flower	46	[11]
2	TiO <sub>2</sub> nanowires	32.65	[12]
3	TiO <sub>2</sub> :PANI	66.1	[8]
4	TiO <sub>2</sub> :PPI	57	[17]

To date, TiO<sub>2</sub>-based nanocomposite films for EGFET pH sensor applications have not yet reached comprehensive electrochemical measurements. In this study, the sensing electrode was prepared utilizing composition of TiO<sub>2</sub> and PVP (TiO<sub>2</sub>:PVP) film. Meanwhile, TiO<sub>2</sub> film was deposited as another sample of SE for comparison purpose. Furthermore, to investigate the reliability and long-term stability of the sensing electrode in pH sensing response, the measurements were repeated multiple times in accordance with the pH2 to pH12 range.

## 2.0 METHODOLOGY

Two sensing electrodes (i) TiO<sub>2</sub> film and ii) TiO<sub>2</sub>:PVP films were fabricated on indium-doped tin oxide (ITO) coated glass substrates. Before the deposition process, the substrates were cleaned with methanol and deionized water and subsequently dried with an air blower to remove any residual moisture. The preparation of TiO<sub>2</sub> solution consists of several materials: titanium isopropoxide, Ti[OCH(CH<sub>3</sub>)<sub>2</sub>]<sub>4</sub>, Triton X-100, glacial acetic acid, and ethanol absolute as a precursor, surfactant, stabiliser, and solvent, respectively. The preparation of polyvinylpyrrolidone (PVP) solution was separated in another beaker. First, 0.1 wt% PVP powder was dissolved in a mixture of ethanol and deionized (DI) water. Then, a mixture of TiO<sub>2</sub> and PVP solutions was set at a ratio of 2:1 and stirred for 1 hour at room temperature. Prior to the material deposition process, Kapton tape was used to cover half of the ITO-conductive surface. The purpose is to create a connection between SE and the gate of the FET device, as illustratively shown in Figure 1. In the same figure, the sample structures of (a) TiO<sub>2</sub> and (b) TiO<sub>2</sub>:PVP SE can be referred to as well. The thin film deposition process was set at 3000 rpm spin speed for 60 seconds. The films were dried in a hot oven at 150 °C for 10 minutes. Then, only TiO<sub>2</sub> was annealed at 400°C for 10 minutes and finally formed the TiO<sub>2</sub>/ITO sensing electrode. Meanwhile, TiO<sub>2</sub>:PVP/ITO was prepared as a sensing electrode without annealing treatment procedure.



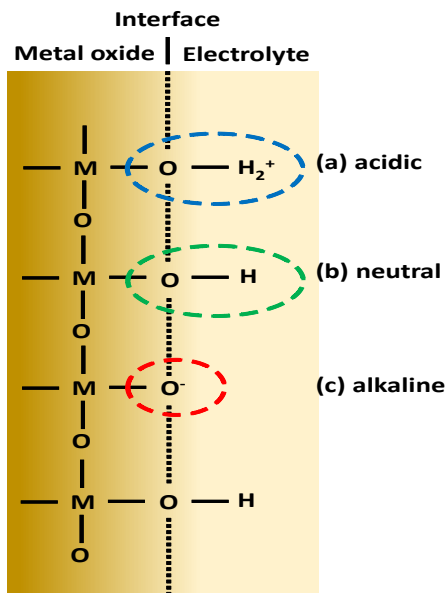
**Figure 1** Electrochemical EGFET pH sensor measurement setup with sensing electrode configuration, (a) TiO<sub>2</sub>:PVP composite film, and (b) TiO<sub>2</sub> films

The pH EGFET electrochemical measurement set-up is depicted in Figure 1, where SE and RE are connected to a commercial MOSFET (CD4007UBE) and a Semiconducting Devices Analyzer Keysight B15000A, respectively. A conductive Cu wire was used to connect SE and the gate of the MOSFET device. To maintain a complete potential loop throughout the measurements, a commercial reference electrode (RE) (RE-1B Ag/AgCl, Metrohm) was employed in this study. In a pH buffer solution, these electrodes were carefully placed opposite each

other and immersed for a minute prior to measurement. Various pH values were utilised in this study, i.e., pH2, pH4, pH7, pH10, and pH12.

### 3.0 RESULTS AND DISCUSSION

Using a semiconductor device analyser, transfer characteristics ( $I_{DS}-V_{REF}$ ) for both SEs, the single film of  $TiO_2$  and the composite films of  $TiO_2:PVP$  in the linear region is shown in Figure 3. At x-axis, reference voltage ( $V_{REF}$ ) was set in range from 0 V to 3 V. The pH sensing measurement was focused within pH2 to pH12. As a result of the pH increasing (decreased hydrogen ions concentration), all transfer curves for the samples exhibited right shifts. This result formed an interaction between the hydroxyl groups on SE's oxide surface and an electrolyte solution that contains  $H^+$  and  $OH^-$  ions. Figure 2 essentially depicts the immersion process interaction between the metal oxide and electrolyte. According to the types of pH buffer solution, it can be categorised into three reactions: neutral, acidic, and alkaline.



**Figure 2.** Illustration of metal oxide surface reaction at different pH conditions

Since there is a significant concentration of  $H^+$  ions in most acidic solutions, the hydroxyl ( $-OH$ ) group on the metal oxide surface picks up an  $H^+$  ion and creates a positive charge surface. As a result, the protonation process is taking place. The chance for the metal oxide surface to lose the  $H^+$  ion from the hydroxyl group and undergo deprotonation is greater in an alkaline solution due to the larger concentration of  $OH^-$  [19].

The most crucial requirement for a pH sensor is that it has good sensitivity. Sensitivity is a measurement in which the sensing material could serve as the sensitive layer and its capacity to interact with hydrogen ions in the solution is assessed. According to the site-binding theory, the surface potential between the electrolyte interface and the sensing electrode is influenced by the number of binding sites on the

sensing surface. Hence, the difference in surface potential is described in Equation (1):

$$2.303(pH_{PZC} - pH) = \frac{q\psi}{kT} + \sinh^{-1} \left[ \frac{q\psi}{kT} \frac{1}{\beta} \right] \quad (1)$$

Where  $pH_{PZC}$  is the value of pH buffer solution at zero charge point,  $q$  is the electron charge,  $k$  is Boltzmann's constant,  $T$  is the temperature,  $\psi$  is a linear response to the pH, and  $\beta$  is the sensitivity parameter of the gate insulators that is reliant upon the hydroxyl groups' surface density. At room temperature (300 K), the maximum pH sensitivity of  $\sim 59.2$  mV/pH at the gate sensing electrode surface is obtained when the sensitivity parameter ( $\beta$ ) approaches unity, and it is known as the Nernst limit [20], [21], [22].

The The constant current of  $100 \mu A$  from drain current ( $I_{DS}$ ) was used to extract the  $V_{REF}$  value from the transfer curve plot, as indicated in Figure 3 (a). Also, this constant current has been applied by other studies that utilised the same CD4007UBE as a commercial MOSFET [11], [23], [24]. Hence, the sensitivity and linearity values of the samples were determined with regard to each pH value. In Figure 3(b), the sensitivity value for  $TiO_2$  film was quite low, i.e.,  $49.69$  mV/pH. Meanwhile, the sensitivity value of the  $TiO_2:PVP$  composite film was found to be the highest in this study, which is  $63.17$  mV/pH. Hence, the  $TiO_2:PVP$  composite film showed a super-Nernstian pH response, where the sensitivity value exceeded the Nernstian limit. The super-Nernstian response could be associated with a more efficient proton-exchange process due to the polymeric and metal oxide material composites that cause the sensing electrodes to experience distinct oxidation states, which raises sensitivity and far more accurate pH change detection [8], [23], [25].

Furthermore, Furthermore, this is in line with the previous research review, which discovered that a metal oxide and conductive material combination is advantageous for pH sensing since it creates an ion transfer channel [6]. The sensitivity value found in the current study was comparable to earlier studies utilising a similar combination of the  $TiO_2$ -conducting polymer, for instance,  $TiO_2:PANI$  [8]. Veira *et al.* performed pH sensor measurement with fabricated layer-by-layer PPI/ $TiO_2$  and obtained a slightly low sensitivity value of  $57$  mV/pH. Additionally, research on the pH sensitivity of single  $TiO_2$  films made using different fabrication techniques has been done elsewhere and exhibited sensitivity values ranging from  $36$  to  $52$  mV/pH [5], [7], [13], [26].

The output characteristics ( $I_{DS}-V_{DS}$ ) in the saturation regime are shown in Figure 4. This regime is formed by the current flow from drain to source in the channel, and at a fixed gate voltage,  $I_{DS}$  becomes almost constant. Here, the saturated drain current was taken at  $V_{DS} = 2$  V, and the sensitivity was determined by a similar procedure using slope and linear regression of the  $I_{DS}$ -pH level graph. This revealed that the sensitivity for  $TiO_2$  films and  $TiO_2:PVP$  composite films was  $1.0$  and  $1.27$  ( $\mu A$ )<sup>1/2</sup>/pH, respectively. From this, it is confirmed that the prepared  $TiO_2:PVP$  composite film possessed highly sensitive criteria as SE for an EGFET pH sensor.

The capacity of a sensor to be used under repeated measurement more than once is known as repeatability measurement. In this work, only  $I_{DS}-V_{REF}$  transfer characteristics in the linear region measurement were repeated with the same

setting three times. Table 2 depicts the  $V_{REF}$  for each pH value at readings 1, 2, and 3, and the pH sensitivity and linearity of the EGFET sensor were determined for each reading accordingly. Here, almost no changes in  $V_{REF}$  values were observed within the measurement readings as the standard deviation of 0.002 remained the same for each pH2 to pH10; however, a slightly large deviation of 0.007 occurred at the

highest basic solution used in this study (pH12). Additionally, the sensitivity values for Reading 1, Reading 2, and Reading 3 were 63.8 mV/pH, 63.1 mV/pH, and 62.6 mV/pH, respectively. As a result, the average sensitivity and linearity value of the  $TiO_2$ :PVP EGFET pH sensor obtained with the deviation factor were  $63.1 \pm 0.60$  mV/pH and  $0.9902 \pm 0.002$ .

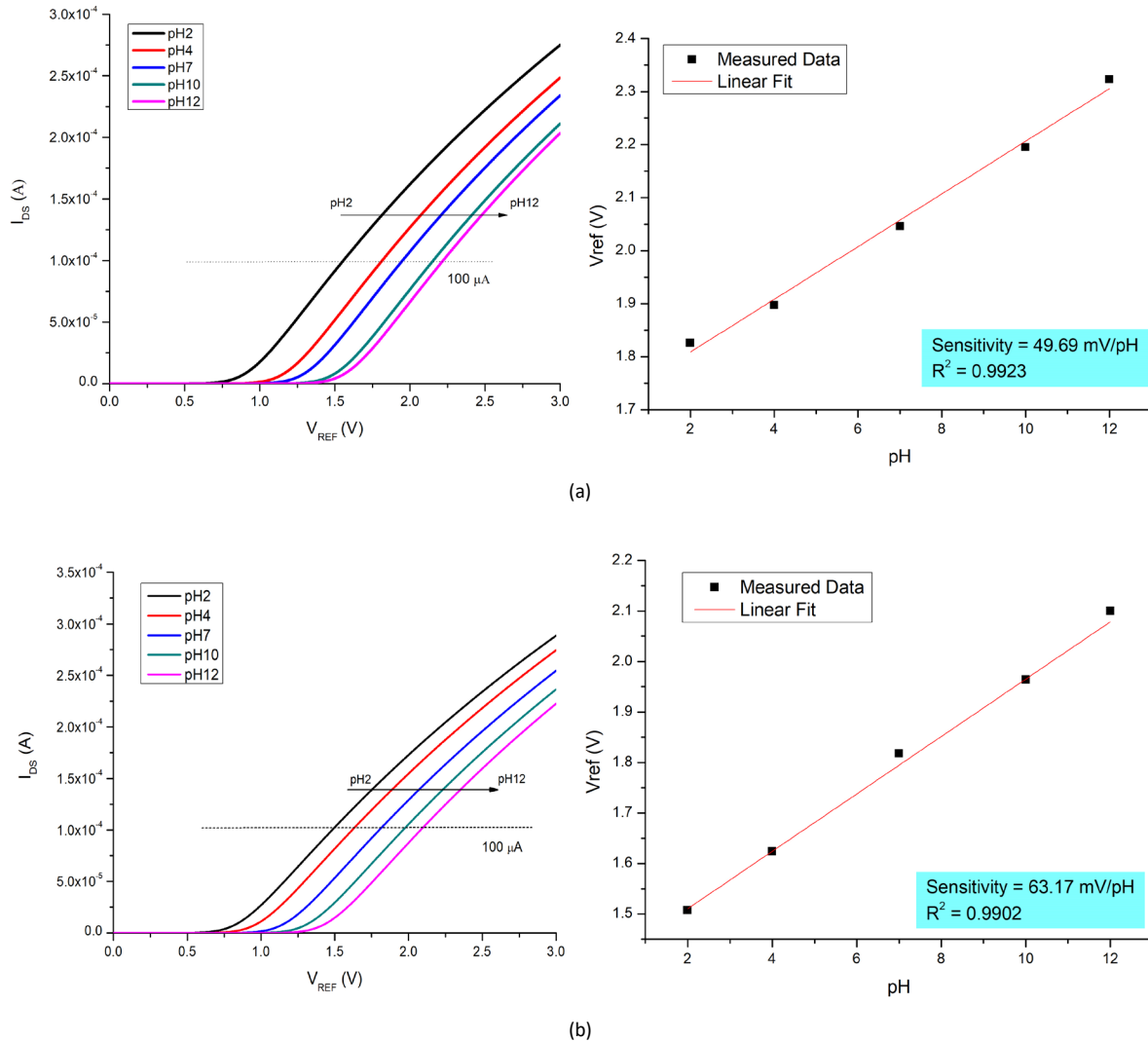
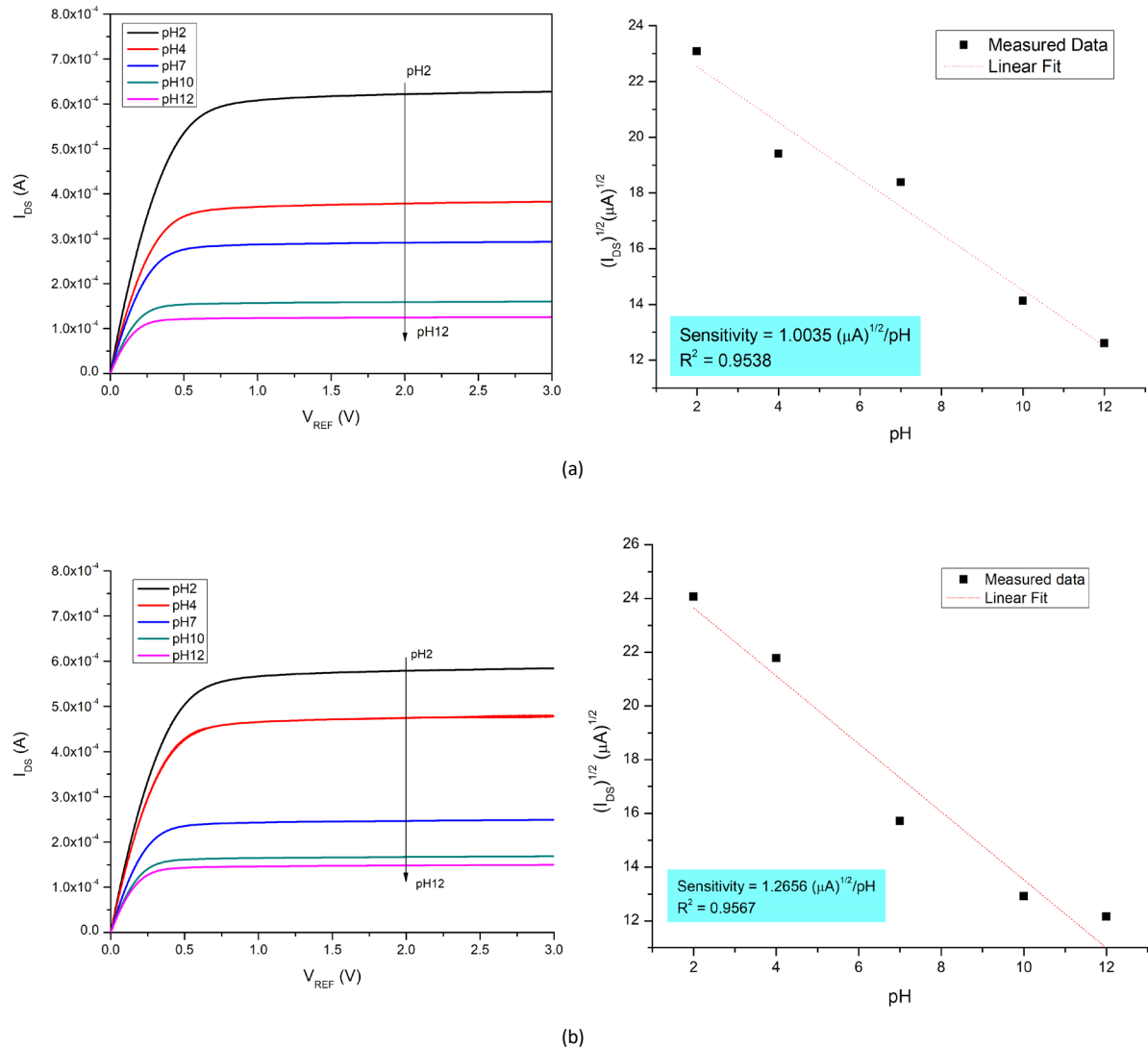


Figure 3 Plot of  $I_{DS}$ - $V_{REF}$  transfer characteristic in linear region (left) and  $V_{ref}$ -pH value (right), (a)  $TiO_2$  film and (b)  $TiO_2$ :PVP composite film.

Table 2 Repeatability measurement of  $TiO_2$ :PVP composite film

pH	* $V_{REF}$ (V)			Average (V)	Standard Deviation
	01	02	03		
2	1.507	1.507	1.507	1.507	0.000
4	1.620	1.622	1.623	1.624	0.002
7	1.819	1.818	1.816	1.818	0.002
10	1.906	1.904	1.903	1.904	0.002
12	2.209	2.202	2.196	2.202	0.007

\* $V_{REF}$  value at 100  $\mu A$  of  $I_{DS}$



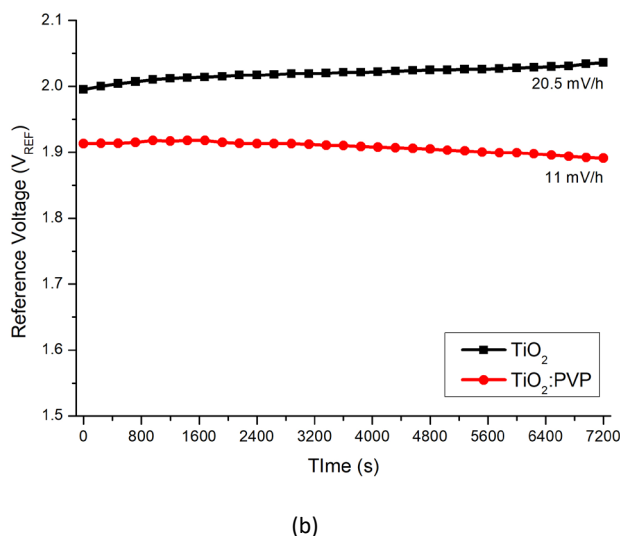
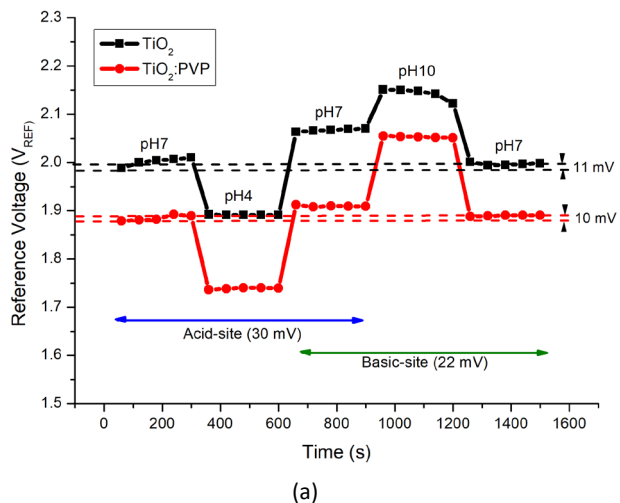
**Figure 4** Plot of  $I_{DS}$ - $V_{REF}$  output characteristic in saturation region (left) and  $(I_{DS})^{1/2}$ -pH value (right), (a) TiO<sub>2</sub> films and (b) TiO<sub>2</sub>:PVP composite film

Further investigation of the electrochemical pH sensor characterization is the hysteresis. This measurement was set for 25 minutes of alternate cycles of various pH buffer solutions, *i.e.*, pH pH7→10→7→4→7 arrangement loops as shown in Figure 5(a). Most previous studies highlighted pH4, pH7, and pH10 loops to be conducted for hysteresis measurement [8], [23], [27]. To obtain the hysteresis value of the sensing layer, the difference in reference voltage between the initial,  $t_0$ , and final,  $t_f$ , described as  $\Delta V_{REF} = V_{REF}(t_0) - V_{REF}(t_f)$ , was determined. The TiO<sub>2</sub> and TiO<sub>2</sub>:PVP composite films demonstrated almost similar hysteresis voltages, *i.e.*, 11 mV and 10 mV, respectively. In Figure 4(a), a 1 mV difference for both electrodes resulted from hysteresis measurement. Nevertheless, degradation of the sensing performance of TiO<sub>2</sub> films can be seen on the basic side compared to TiO<sub>2</sub>:PVP composite films. Yao *et al.* reported alleviation of hysteresis value when the contrast arrangement was demonstrated, pH7→10→7→4→7 [26]. Similar observations were obtained in other studies as well [22]. Besides that, the difference in hysteresis value between acid-site (pH 7→4→7) and basic-site

(pH 7→10→7) was also highlighted in this study. As a result, the hysteresis characteristic of TiO<sub>2</sub>:PVP composite film at the acid site demonstrated a slightly greater value (30 mV) than at the basic site (22 mV). This finding is in contrast to the most previous studies, which stated there were significant highly hysteresis characteristics at the basic site compared to the acid site [7], [13], [26]. This could be a result of the interaction between the electrolyte's H<sup>+</sup> and/or OH<sup>-</sup> ions and the surface sites that are delayed in responding underneath the surface of the membrane [19]. Also, because of irregularities in the membrane, hysteresis becomes affected by the chemical interaction of ions in the electrolyte with delayed response surface sites.

The drift, or change in the measured value over a period of time in a fixed pH buffer solution, essentially characterises the electrochemical sensor's long-term stability. The drift rate ( $\Delta V_{REF}/h$ ) is proportional to the changes in  $V_{REF}$  in accordance with the given time. The drift measurement was taken precisely for pH7 buffer solutions in 2 hours, and the value of  $V_{REF}$  vs. time was plotted in Figure 5(b). The drift rate of 20.5 mV/h was

obtained using TiO<sub>2</sub> films. Meanwhile, a smaller drift rate of 11 mV/h was found with TiO<sub>2</sub>:PVP films; therefore, the composite films improved the device's stability, and this proved that the surface of TiO<sub>2</sub>:PVP films showed better ion interaction in the given immersion period. The summarized sensing performance of the TiO<sub>2</sub> and TiO<sub>2</sub>:PVP films are tabulated in Table 3.



**Figure 5** Stability performance of TiO<sub>2</sub> film (square, black) and TiO<sub>2</sub>:PVP composite film (circle, red), (a) hysteresis characteristic in pH7→pH4→pH7→pH10→pH7 arrangement and (b) drift measurement for 2 hours in pH7 buffer solution.

**Table 3** Sensing performance of TiO<sub>2</sub> films and TiO<sub>2</sub>:PVP composite films.

Sensing Electrode	Sensitivity (mV/pH)	Linearity	Hysteresis (mV)	Drift Rate (mV/h)
TiO <sub>2</sub>	49.69	0.9923	11	20.5
TiO <sub>2</sub> :PVP	63.17	0.9903	10	11

## 4.0 CONCLUSION

In conclusion, we successfully fabricated a sensing electrode of TiO<sub>2</sub> film and TiO<sub>2</sub>:PVP composite film for an EGFET pH sensor as a sensing electrode. Compared to TiO<sub>2</sub> film, the composite film was shown to be highly sensitive to a wide range of pH values (pH2, pH4, pH7, pH10, and pH12), and repeatability measurements showed that it made a good pH-sensing electrode. The TiO<sub>2</sub>:PVP:PVPsing electrode experienced a super-Nernstian response, a pH sensitivity of 63.2 mV/pH, and a linearity almost ~1 achieved in this study. The composite films managed to serve as an accurate and stable sensing electrode, proven by a lower drift rate of 11 mV/h and a small hysteresis value of 10 mV as well. This demonstrated the capability of the composite film to serve as a sensing electrode for the EGFET pH sensor. To perform for a longer period, additional analysis will be done.

## Acknowledgement

The Fundamental Research Grant Scheme of the Ministry of Higher Education of Malaysia (Project Code: FRGS/1/2021/TK0/UITM/02/50) contributed a partial grant for this study. The authors credit NANO-Electronic Centre (NET), Universiti Teknologi MARA (UiTM) for their technical assistance.

## Conflicts of Interest

The author(s) declare(s) that there is no conflict of interest regarding the publication of this paper

## References

- [1] Villamizar, R. A., Maroto, A., Rius, F. X., Inza, I., and Figueras, M. J. 2008. Fast detection of Salmonella Infantis with carbon nanotube field effect transistors. *Biosensors and Bioelectronics*. 24(2):279–283. DOI: 10.1016/j.bios.2008.03.046
- [2] Purwidyantri A. Lai, H. C., Tsai, S. H., Luo, J. D., Chiou, C. C., Tian, Y. C., Cheng, C. H. Lin, Y. T., and Lai, C. S. 2015. Sensing performance of fibronectin-functionalized Au-EGFET on the detection of S. epidermidis biofilm and 16S rRNA of infection-related bacteria in peritoneal dialysis. *Sensors and Actuators, B: Chemical*. 217: 92–99. DOI: 10.1016/j.snb.2014.11.017
- [3] Pandey, A., Gurbuz, Y., Ozguz, V., Niazi, J. H., and Qureshi, A. 2017. Graphene-interfaced electrical biosensor for label-free and sensitive detection of foodborne pathogenic E. coli O157:H7. *Biosensors and Bioelectronics*. 91(2017):225–231. DOI: 10.1016/j.bios.2016.12.041
- [4] Könemund, L., Neumann, L., Hirschberg, F., Biedendieck, R., Jahn, D., Johannes, H. H., and Kowalsky, W. 2022. Functionalization of an Extended-gate field-effect transistor (EGFET) for Bacteria Detection. *Scientific Reports*. 12(1): 1–10. DOI: 10.1038/s41598-022-08272-3
- [5] Khizir, H. A., and Abbas, T. A. H. 2022. Hydrothermal synthesis of TiO<sub>2</sub> nanorods as sensing membrane for extended-gate field-effect transistor (EGFET) pH sensing applications. *Sensors and Actuators A: Physical*. 333(2022): 113231-40. DOI: 10.1016/j.sna.2021.113231
- [6] Manjakkal, L., Szwagierczak, D., and Dahiya, R. 2020, May. Metal oxides based electrochemical pH sensors: Current progress and future perspectives. *Progress in Materials Science*. 109:100635-66. DOI: 10.1016/j.pmatsci.2019.100635
- [7] Zulkefle, M. A., Herman, S. H., Abdul Rahman, R., Yusof, K. A., Rosli, A. B., Abdullah, W. F. H., and Zulkifli, Z. 2021. Evaluation on the EGFET pH sensing performance of sol-gel spin coated titanium dioxide thin

- film. *Jurnal Teknologi*. 83(4): 119-125. DOI: 10.11113/jurnalteknologi.v83.16313
- [8] Kamarozaman, N. S., Zainal, N., Rosli, A. B., Zulkefle, M. A. Nik Him, N. R., Abdullah, W. F. H., Herman, S. H., and Zulkifli, Z. 2022. Highly Sensitive and Selective Sol-Gel Spin-Coated Composite TiO<sub>2</sub>-PANI Thin Films for EGFET-pH Sensor. *Gels*. 8(11): 690. DOI: <https://doi.org/10.3390/gels8110690>
- [9] Venkatachalaiah, C., Venkataraman, U., and Sellappan, R. 2020. PANI/TiO<sub>2</sub> nanocomposite-based chemiresistive gas sensor for the detection of E. Coli bacteria. *IET Nanobiotechnology*. 14(9): 761–765. DOI: 10.1049/iet-nbt.2020.0046
- [10] Zhao, R., Xu, M., Wang, J., and Chen, G. 2010. A pH sensor based on the TiO<sub>2</sub> nanotube array modified Ti electrode. *Electrochimica Acta*. 55 (2010): 5647–5651. DOI: 10.1016/j.electacta.2010.04.102
- [11] Yang, C. C., Chen, K. Y., and Su, Y. K. 2019. TiO<sub>2</sub> nano flowers based EGFET sensor for pH sensing. *Coatings*. 9(4): 251-257. DOI: 10.3390/coatings9040251
- [12] Li, J., Chang, S., Chang, S., and Tsai, T. 2014. Sensitivity of EGFET pH Sensors with TiO<sub>2</sub> Nanowires. *ECS Solid State Letter*. 3(10): 126–129. DOI: 10.1149/2.0091410ssl
- [13] Chen, C., Zhang, Y., Gao, H., Xu, K., and Zhang, X. 2022. Fabrication of Functional Super-Hydrophilic TiO<sub>2</sub> Thin Film for pH Detection. *Chemosensors*. 10(5): 182-196. DOI: 10.3390/chemosensors10050182
- [14] Sreekanth, K., Siddaiah, T., Gopal, N. O., Jyothi, N. K., Kumar, K. V., and Ramu, C. 2021, August. Thermal, Structural, Optical and Electrical Conductivity studies of pure and Mn<sup>2+</sup> doped PVP films. *South African Journal of Chemical Engineering*. 36(2021): 8–16. DOI: 10.1016/j.sajce.2020.09.003.
- [15] Tang, I. H., Sundari, R., Lintang, H. O., and Yuliaty, L. 2016, October. Polyvinylpyrrolidone as a new fluorescent sensor for nitrate ion. *Malaysian Journal of Analytical Sciences*. 20(2): 288–295. DOI: 10.17576/mjas-2016-2002-09.
- [16] Rosli, A. B., Hashim, S. B., Rahman, R. A., Herman, S. H., Abdullah, W. F. H., and Zulkifli Z. 2022, November. Correlation of ZnO Surface Morphology and Sensing Performance of EGFET Nitrate Sensor. *Journal of Mechanical Engineering*. 11(Special Issue 1):65–80. DOI: 10.24191/jmeche.v11i1.23586.
- [17] Vieira, N. C. S., Figueiredo, A., Faceto, A. D., De Queiroz, A. A. A., Zucolotto, V., and Guimarães, F. E. G. 2012, July. Dendrimers/TiO<sub>2</sub> nanoparticles layer-by-layer films as extended gate FET for pH detection. *Sensors and Actuators, B: Chemical*. 169(2012): 397–400. DOI: 10.1016/j.snb.2012.01.003
- [18] Palit, S., Singh, K., Lou, B. S., Her, J. L., Pang, S. T., and Pan, T. M. 2020, January. Ultrasensitive dopamine detection of indium-zinc oxide on PET flexible based extended-gate field-effect transistor. *Sensors and Actuators, B: Chemical*. 310(2020): 127850-127858. DOI: 10.1016/j.snb.2020.127850
- [19] Al-Hardan, N. H., Abdul Hamid, M. A. Ahmed, N. M., Jalar, A. Shamsudin, R., Othman, N. K., Keng, L. K., Chiu, W., Al-Rawi, H. N. 2016. High sensitivity pH sensor based on porous silicon (PSi) extended gate field-effect transistor. *Sensors (Switzerland)*. 16(6): 839-850. DOI: 10.3390/s16060839.
- [20] Dwivedi, P., Singh, R., and Singh, Y. C. 2020, August. Crossing the Nernst Limit (59 mV/pH) of Sensitivity Through Tunneling Transistor Based Biosensor. *IEEE Sensor Journal*. 21(3): 3233–3240. DOI: 10.1109/JSEN.2020.3025975.
- [21] Oh, J. Y., Jang, H. J., Cho, W. J., and Islam, M. S. 2019, April. Highly sensitive electrolyte-insulator-semiconductor pH sensors enabled by silicon nanowires with Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> sensing membrane. *Sensors and Actuators, B: Chemical*. 171–172(2019): 238–243. DOI: 10.1016/j.snb.2012.03.052.
- [22] Yusof, K. A., Rahman, R. A., Zulkefle, M. A., Herman, S. H., and Abdullah, W. F. H. 2016, July. EGFET pH Sensor Performance Dependence on Sputtered TiO<sub>2</sub> Sensing Membrane Deposition Temperature. *Journal of Sensors*. 2016: 431-439. DOI: <https://doi.org/10.1155/2016/7594531>
- [23] Das, A., Ko, D. H., Chen, C. H., Chang, L. B., Lai, C. S., Chu, F. C. Chow, L., Lin, R. M. 2014, December. Highly sensitive palladium oxide thin film extended gate FETs as pH sensor. *Sensors and Actuators, B: Chemical*. 205(2014): 199–205. DOI: 10.1016/j.snb.2014.08.057.
- [24] Sharma, P., Singh, R., Sharma, R., Mukhiya, R., Awasthi, K., and Kumar, M. 2021, September, Palladium-oxide extended gate field effect transistor as pH sensor. *Materials Letters: X*. 12(2021): 100102. DOI: 10.1016/j.mblux.2021.100102.
- [25] Shaibani, P. M., Jiang, K., Haghighat, G., Hassanpourfard, M., Etayash, H., Naicker, S. The Detection of Escherichia coli (E. coli) with the pH Sensitive Hydrogel Nanofiber-Light Addressable Potentiometric Sensor (NF-LAPS). *Sensors and Actuators, B: Chemical*. 226(2016): 176–183. DOI: 10.1016/j.snb.2015.11.135.
- [26] Yao, P. C., Chiang, J. L., and Lee, M. C. 2014. Application of sol-gel TiO<sub>2</sub> film for an extended-gate H<sup>+</sup> ion-sensitive field-effect transistor. *Solid State Sciences*. 28 (2014):47-54. DOI: 10.1016/j.solidstatesciences.2013.12.011
- [27] Rosli, A. B. B., Hamid, N. H. B., Zulkefle, M. A. B., Shariffudin, S. S. B., Abdullah W. F. H. B., and Herman S. H. 2023, October. Post-deposition heat treatment effect on pH sensing behavior of chemical bath deposited nanostructured zinc oxide. *International Journal of Hydrogen Energy*. 48(4): 1636-1648. DOI: 10.1016/j.ijhydene.2022.10.036.