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Effect of Polymer Types on Metal Oxide Substrates as an EGFET Sensor-Based Nitrate Sensing Layers

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ABSTRACT

The effect of polymer-based sensing electrodes on the detection of nitrate was presented in this paper. Polyvinyl butyral (PVB) and Polyvinylpyrrolidone (PVP) were employed as the nitrate sensing layers of the Extended Gate Field Effect Transistor (EGFET), which was fabricated on different metal oxide substrates, namely Tantalum Pentoxide (Ta_2O_5) and Indium Titanium oxide (ITO). The sensing electrode was connected to the gate of CD4007 commercial MOSFET. The spin coating technique was used to deposit the polymer concentration at 3000 rpm for 30 seconds for each sample. The samples were characterized to obtain the linearity and sensitivity of the sensing electrode from three different nitrate concentrations (0, 50 and 100 ppm). Ta_2O_5 shows a higher sensitivity for the metal oxide substrate than the ITO, with 7.2 mV/dec for ITO and 14.5 mV/dec for Ta_2O_5. The study has proved that the deposition of polymers on the sensing electrode improved sensitivity and linearity. The PVB on ITO and Ta_2O_5 shows the sensitivity and linearity of 40.9 mV/dec and 0.9994 and 36.9 mV/dec and 0.3832, respectively. It shows that the nitrate sensing performance of the ITO sensing electrode improved almost six times when PVB polymer was used as a composite layer. While the use of PVB polymer on top of the Ta_2O_5 sensing layer improved the sensitivity values by approximately three times than sensitivity value obtained using bare Ta_2O_5. In contrast, the sensitivity for PVP demonstrates 24.4 mV/dec and 36.3 mV/dec, with an increment of 68% and 404% on Ta_2O_5 and ITO, respectively. The linearity shows 0.8713 for Ta_2O_5 and 0.5723 for ITO. Thus, PVP is better on Ta_2O_5, and PVB is better on ITO substrate. Both conditions are significant in improving the sensitivity of the sensing electrode.

Keywords: *Polyvinyl Butyral, Polyvinylpyrrolidone, Nitrate Sensing, Spin Coating, EGFET Nitrate Sensor*

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INTRODUCTION

The most common neutral element in the atmosphere and soil is nitrogen. Nitrogen can be found in forms such as nitrate (NO_3^-) and ammonium (NH_4^+) [1]. NO_3^- are commonly known and found in soil and fertilizer, but in the commercialized food industry, nitrate ions have been widely used as the preservative [2]. The excessive use of fertilization in the agricultural industry has recently increased surface contamination and groundwater. The properties of nitrate that are soluble in water make nitrate can be easily transported to another surface and subsurface, then flows to rivers and lakes or moved down to groundwater. This exposure to groundwater, especially in agricultural areas, has become a significant public health issue and causes environmental pollution [3].

Because of the worrying impact of nitrate on the industry, agriculture, environment and ecological systems, a highly sensitive, selective, and accurate method to monitor nitrate ion concentration is needed. Many methods have been developed for nitrate sensing in liquid samples, such as electrochemical, spectrophotometry, chemiluminescence, and chromatography [1, 3-6]. Among the mentioned methods, the electrochemical method is considered more efficient due to the good high usability, including portable instruments, more straightforward setup, ease of sample preparation and higher sensitivity or selectivity [1, 7-9].

Various materials detect nitrates, such as conducting polymer, copper, biosensor, carbon nanotubes, graphene, graphene oxide and silver nanoparticles [10–13]. The use of polymers for sensing applications has increased in recent years due to the characteristics of the polymer, such as low cost, easy processibility, and improved sensing performance. Thus, the use of polymer can further amplify the detection response [14]. Polyvinyl butyral (PVB) and Polyvinylpyrrolidone (PVP) are a polymer that was employed as nitrate sensing layers. PVP is used as a passivator or a capping agent for semiconductors, and PVB is usually used as a resin, being stronger in binding and providing flexibility and toughness. [15, 16].

In general, an electrochemical sensor can translate ion analysis into an existing signal, potential difference and impedance. This sensor is commonly used due to its simple operation, good sensitivity to nitrate ions in water, easy miniaturization, and low power consumption. Electrochemical sensors usually can be divided into two groups for nitrate detection: potentiometric sensors [17, 18] and voltammetric or amperometric sensors [19, 20], depending on the details of the experimental design. The electrostatic potentials [volt (V)] are measured for the potentiometric method and do not involve current in the measurement. While for voltametric or amperometric methods, current measurement is involved, and the potential difference between the two electrodes was used as a variable input during measurement.

One example of an electrochemical sensor is the ion-sensitive field-effect transistor (ISFET) sensor. The ISFET sensor with SiO_2 insulator was employed as the sensing electrode, as reported by Bergveld in 1970 [21]. The basic configuration of ISFET is derived from a metal-oxide-semiconductor field-effect transistor (MOSFET), in which the metal gate is substituted by an ion-sensitive membrane with direct contact with a buffer solution [22]. However, the ISFET suffers several disadvantages, including device instability and poor sensitivity [23]. However, these disadvantages can be overcome by the extended-gate field-effect transistor (EGFET) structure.

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EGFET was proposed by Spiegel et al. [24] in 1983. EGFET structure isolated the FET from the chemical environment. A chemically sensitive membrane (or sensing electrode) was deposited at the end of the signal line extending from the FET gate electrode. It offers several advantages, such as light insensitivity, simpler packaging, and flexibility for the shape of the extended gate area [23–25]. EGFET has been applied in an extensive range of applications to detect any substance, especially as biosensors. EGFET is also favourable because it can be characterized using the simple connection and flexibility of the extended gate design [26].

According to previous research on EGFET, most researchers focus on sensing electrode materials [27-29]. Since this part is believed can improve the EGFET sensing performance. Among the materials used as sensing material, metal oxide received extensive attention as an EGFET sensing electrode. Tantalum pentoxide (Ta_2O_5) shows excellent performance as an EGFET pH sensor due to several advantages, such as mechanical strength, stability, and semiconducting nature in bulk form. Other than that, Ta_2O_5 thin films are stable at high temperatures [30, 31]. However, using Ta_2O_5 as an EGFET nitrate sensor is still lacking.

Therefore, in this research, the sensitivity of the electrochemical sensor toward nitrate was studied based on the EGFET configuration with polymer-metal oxide-based sensing electrodes. The result of PVB and PVP fabricated on Ta_2O_5 and ITO were compared. The method used to fabricate the sensing electrode is spin coating.

EXPERIMENTAL

Preparation of Polyvinylpyrrolidone (PVP) and Polyvinyl Butyral Solution (PVB)

Figure 1 shows the preparation step of the PVP and PVB solution. PVP and PVB were prepared by mixing 2.0 g of powder in 20 mL of mixed ethanol and deionized (DI) water with a ratio of 8:2. The prepared solution was stirred for 1 hour at 350 rpm. Then, the prepared solution was dropped on top of the substrate using a spin coater with three steps. Lastly, the prepared sample was dried on a hot plate at 100 ºC for 10 minutes. Table 1 shows the complete parameters for preparing the solution.

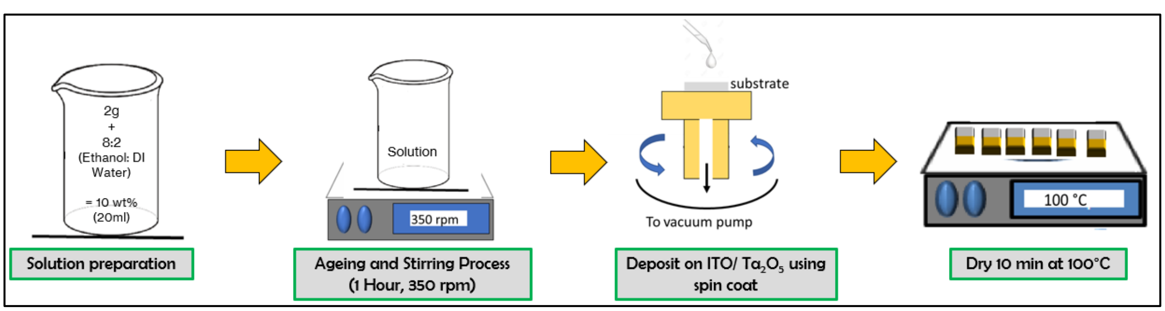

Figure 1: Preparation step of polyvinylpyrrolidone (PVP) and polyvinyl butyral (PVB) solution

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Table 1: Materials and Parameters for Solution Preparation

Materials	Mass (g)	Ethanol (ml)	DI Water (ml)	Ageing and Stirring Process	Deposition Process	Drying Process
PVP PVB	2	16	4	Time: 1 hour 350 rpm	Step 1: 500 rpm (10 sec) Step 2: 3000 rpm (30 sec) Step 3: 3000 rpm (30 sec)	Heat: 100 ºC Time: 10 min

Sample Preparation and Sensing Structure

Figures 2 (a) and (b) show the cross-sectional schematic diagram of PVB and PVP on ITO and PVB and PVP on top of Ta_2O_5 thin film. Before the experiment, the ITO substrate was ultrasonically cleaned using Hwashin Technology Powersonic 405 ultrasonic cleaner. The cleaning solution used was methanol, and DI water and argon gas were used to blow-dry the cleaned substrate. Ta_2O_5 thin film was deposited using Radio Frequency (RF) magnetron sputtering with RF power 200 Watt, working pressure 5 mTorr, room temperature for substrate heating temperature, and 1 hour deposition time.

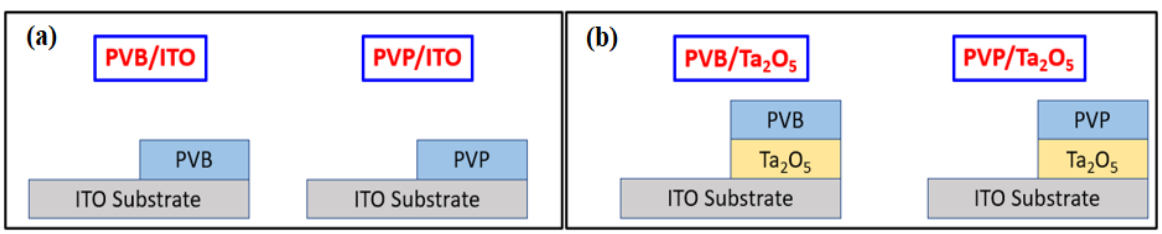

Figure 2: Cross-sectional view of (a) PVB and PVP on ITO substrate and (b) PVB and PVP on Ta_2O_5 thin film

EGFET Measurement Setup

Figure 3 shows the experimental EGFET measurement setup. The reference electrode (RE) was connected to a Semiconductor Device Analyzer (SDA) Keysight B1500A. In contrast, the sensing electrode (SE) was connected to the gate of a commercialized MOSFET CD4007 as the extended gate sensing electrode. The transfer and output characteristics were obtained from this measurement setup. Figure 4 shows the internal schematic circuit of CD4007 MOSFET. The CD4007 consists of 3 pairs inverter with pin 14 as P-type metal oxide semiconductor (PMOS) and pin 7 N-type metal oxide semiconductor (NMOS). This inverter shares a common pin gate (6, 3 and 10). Pins 6, 7 and 8 were used for this measurement. Other pins (3, 4, 5, 10, 9 and 12) also can be used for measurement setup.

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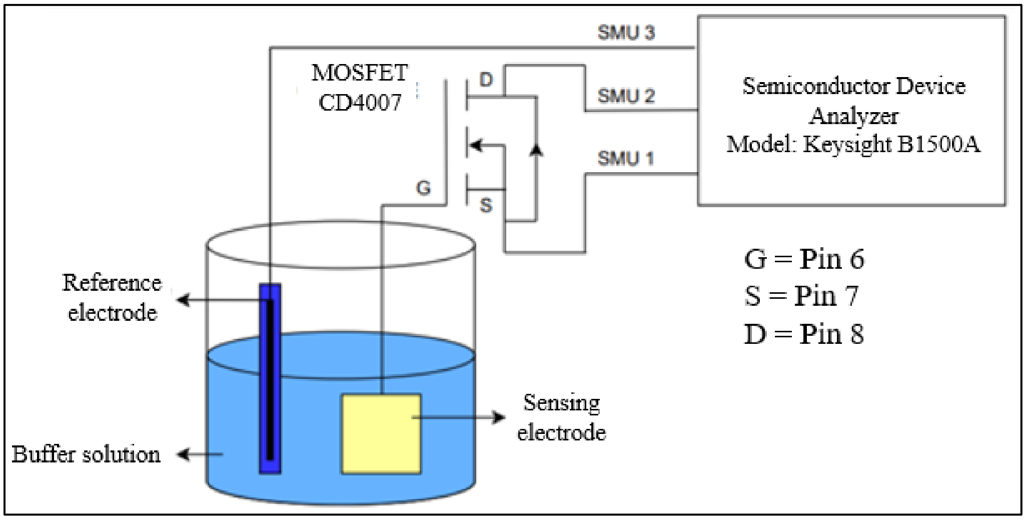

Figure 3: EGFET measurement setup

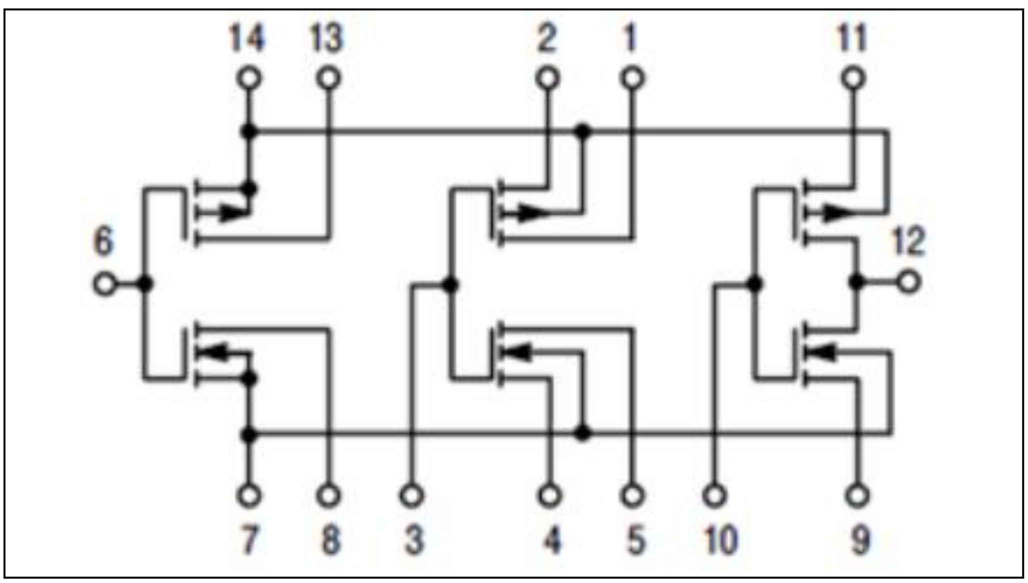

Figure 4: Schematic circuit CD4007 MOSFET

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RESULTS AND DISCUSSION

Figure 5 shows the reference voltage (V_{ref}) graph versus nitrate for bare ITO and Ta_2O_5 plotted at different nitrate concentrations (0, 50 and 100 ppm). The sensitivity and linearity were around 7.2 mV/dec and 0.6569 for bare ITO, while sensitivity and linearity for bare Ta_2O_5 were around 14.5 mV/dec and 0.498, respectively. The figure shows that the sensitivity on Ta_2O_5 was slightly higher than ITO, but for the linearity, ITO was greater. This result shows that both ITO and Ta_2O_5 can be used to detect nitrate ions. However, both materials show low detection values due to the higher applied potential needed for bare electrodes, which limits the detection of a bare electrode [32]. Therefore, using a polymeric layer is required to improve the sensing performance. In order to observe the effect of the polymeric layer on nitrate sensing, performance PVB and PVP were used in this work.

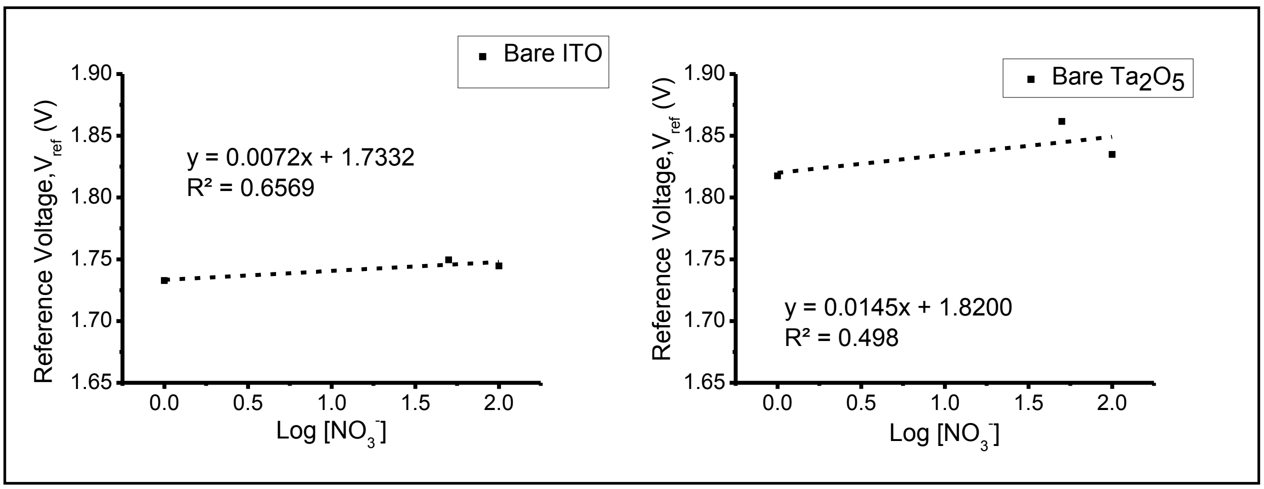

Figure 5: Reference voltage versus nitrate of bare ITO and Ta_2O_5

Figure 6 shows the reference voltage versus nitrate graph plotted for PVB deposited on ITO and Ta_2O_5 thin film. The sensitivity of PVB/ITO and PVB/Ta_2O_5 sensing electrodes was recorded correspondingly at 40.9 mV/dec and 36.9 mV/dec. The graph shows that using PVB polymer improves the sensitivity value compared with bare ITO and Ta_2O_5, as shown in Figure 5. It also can be observed that the linearity values of PVB/ITO are increased from 0.6569 to 0.9994, which is approaching 1. However, the linearity obtained for PVB/Ta_2O_5 is slightly decreased from 0.498 to 0.3832 compared to the linearity value of bare Ta_2O_5.

Further study on polymer effect towards nitrate sensing performance has been done using PVP on ITO and Ta_2O_5 and plotted as shown in Figure 7. From the results, the sensitivity values of PVP/ITO and PVP/Ta_2O_5 are 36.3 and 24.4 mV/dec, while their linearity values are 0.5723 and 0.8713, respectively. Comparing the bare ITO and Ta_2O_5 sensing performance, as shown in Figure 5, shows that the use of PVP

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improved the sensitivity values from 7.2 to 36.3 mV/dec and 14.5 to 24.4 mV/dec, respectively. However, the sensing performance obtained from the sensing electrode using PVB is better than PVP. This might be due to the C=O sites for PVB having much stronger interactions to sense the nitrate ions compare to C=O sites for PVP.

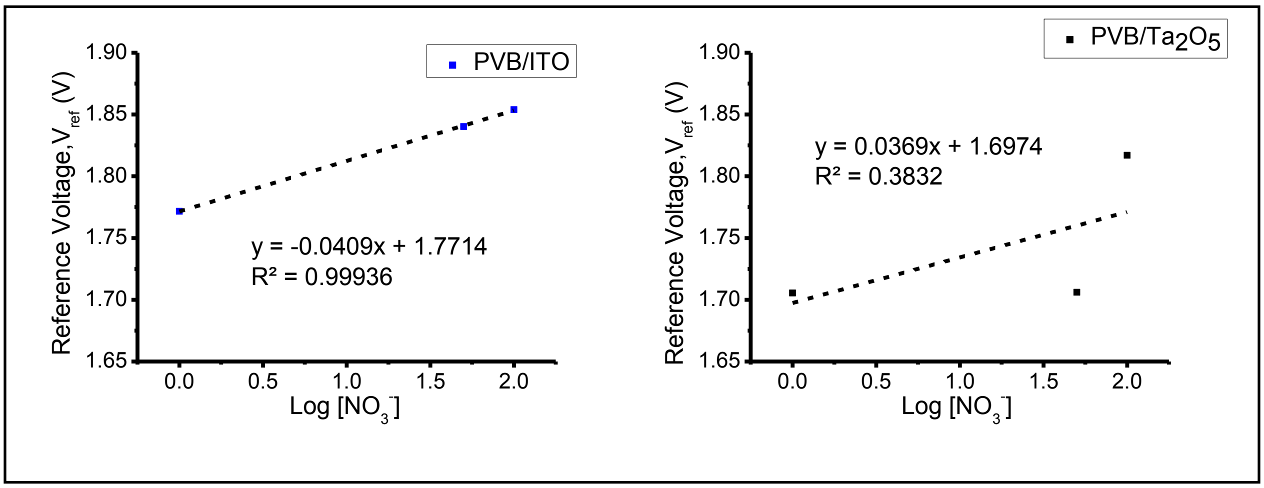

Figure 6: Reference voltage versus nitrate of PVB on top of ITO and Ta_2O_5

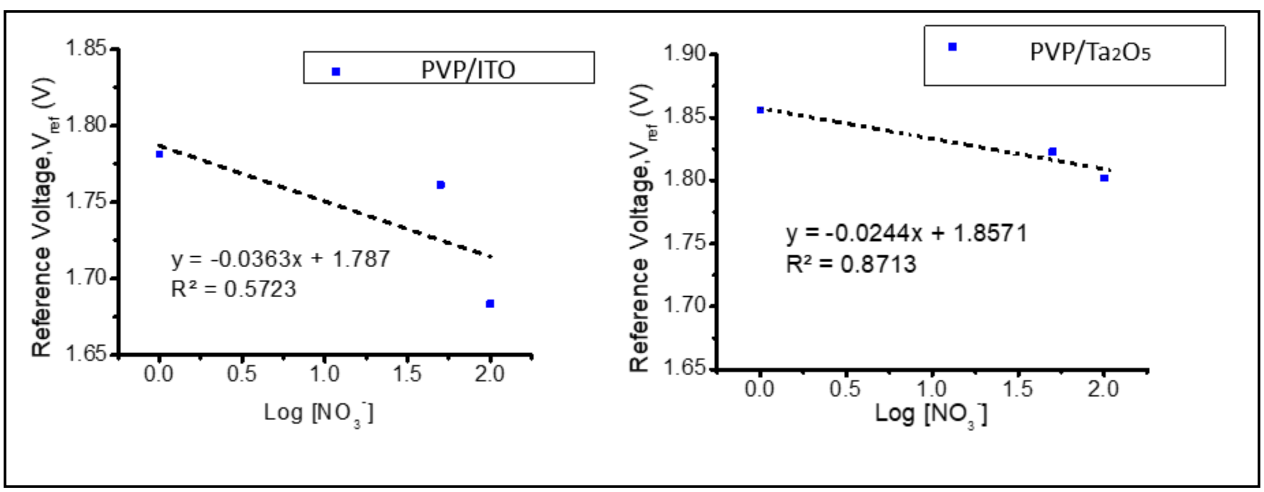

Figure 7: Reference voltage versus nitrate of PVP on top of ITO and Ta_2O_5

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Table 2: Summarize the sensitivity and linearity of PVB and PVP on top of ITO and Ta_2O_5

	Bare ITO	Bare Ta_2O_5	PVB/ITO	PVB/ Ta_2O_5	PVP/ITO	PVP/ Ta_2O_5
Sensitivity (mV/dec)	7.2	14.5	40.9	36.9	36.3	24.4
Linearity (R^2)	0.6569	0.4980	0.9994	0.3212	0.5723	0.8713

The summary of sensitivity and linearity for PVB and PVP on ITO and Ta_2O_5 thin film was tabulated in Table 2. PVB/ITO shows the highest sensitivity and linearity compared to other samples. It also indicates that PVB and PVP have improved the sensitivity of the sensing electrode as the sensitivity value increased more than half compared to the value on bare ITO and Ta_2O_5. The properties of PVB with the formula ($C_8H_{14}O_2$) contain an oxygen-containing functional group. They can be an excellent immobilization material for nano-functional components with fixing on sensor interface [33] are some factors contribute to the higher sensitivity. Besides that, the C=O sites were found to have much stronger interactions with nitrate ions than the N-C sites, suggesting that the C=O sites were more favourable to sensing the nitrate ions, as reported by [16].

CONCLUSION

PVB and PVP as nitrate sensing have been successfully fabricated on top of ITO and Ta_2O_5 using the spin coating method. From the result, the deposition of polymers on the sensing electrode improved sensitivity and linearity compared to bare ITO and Ta_2O_5. The nitrate sensing performance of the ITO sensing electrode improved almost six times when PVB polymer was used as a composite layer. While the use of PVB polymer on top of the Ta_2O_5 sensing layer improved the sensitivity values by approximately three times than sensitivity value obtained using bare Ta_2O_5. In comparison, the sensitivity for PVP increased with an increment of 68% and 404% on Ta_2O_5 and ITO, respectively. In conclusion, PVP performs better on Ta_2O_5, and PVB performs better on ITO substrate. Both conditions have a substantial impact on the sensing electrode's sensitivity.

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AUTHOR'S CONTRIBUTION

Shaiful Bakhtiar wrote and revised the article. Shaiful Bakhtiar and Aimi Bazilah carried out the experiment. Sukreen Hana and Wan Fazlida conceptualized the central research idea, provided the theoretical framework, and supervised research progress. Sukreen Hana and Zurita anchored the review and revisions and approved the article submission.

CONFLICT OF INTEREST STATEMENT

The authors agree that this research was conducted without any self-benefits, commercial or financial conflicts and declare the absence of conflicting interests with the funders.

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