

Determination of glucose in human urine by cyclic voltammetry method using gold electrode

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Abstract. This study has been the determination of glucose in human urine by cyclic voltammetry method using gold electrode. The gold electrode was prepared using gold wire with purity 99.99%, size 1.0 mm by length and wide respectively, connected with silver wire using silver conductive paint. The effect of electrolyte, pH and glucose concentration has been determined to produce the optimum method. The research showed the KNO_3 is a good electrolyte for determination of glucose in human urine using gold electrode. The effect of glucose concentration have the coefficient correlation is $R^2 = 0.994$. The results of the recovery using addition method showed at range 95-105%. As a conclusion is a gold electrode is a good electrode for electrochemical sensors to the determination of glucose in human urine.

1. Introduction

Innovation to improve the quality of medical diagnostic equipment is very necessary to produce tools at affordable prices, easy to mass-produced, easy to operate, have good resistance and environmentally friendly. The demand for medical diagnostic tools for early detection of various diseases such as diabetes and kidney caused by increased uric acid and urea levels is increasing. The increase in demand is due to the increasing number of patients both nationally and internationally. Prevention is better than treatment, the detection of glucose, uric acid and urea is needed to prevent the occurrence of the disease in the community.

Diabetes mellitus (DM) is a condition of disruption of metabolism in the body due to the inability of the body to make or supply the hormone insulin resulting in increased blood glucose levels, exceeding the normal value. If left unchecked, diabetes will be the leading cause of end-stage renal disease, heart attack, stroke, disease, and other complications. Based on WHO data, Indonesia currently ranks fourth as a country with a total diabetes mellitus reaching about 8 million people and estimated the number exceeds 21 million in the year 2025.

Quantitative analysis of glucose is essential in clinical chemistry [1]. Glucose is usually only analyzed by methods involving specific enzymes for glucose so it is not easily disturbed by other sugars that may be present. Various enzyme immobilization techniques may be used, including adsorption on the solid buffer. This is due to the simple sol-gel process conditions and the possibility to design as needed. Sol-gel flexibility permits the formation of sensors as monoliths and thin layers that can be paired with optical fibers or deposited on the electrodes, as well as nanoparticles.



The weakness of the enzyme-based sensors is that the tool has a usage limit and requiring storage conditions. Therefore, research to create a current-based sensor for analysis or nonenzymatic glucose sensors is necessary [2]. Many researchers have been development of sensor glucose non enzyme using various electrode is graphene [3], N,N-Dibromo Dimethylhydantoin [4], platinum nanoparticle [5], nickel-cobalt [6], nickel modified indium [7], nickel [8], Ag and NiO-Ag [9], Pt-Au [1], gold [10,11,12].

In this study has been done cyclic voltammetry was used to evaluate the electrocatalytic activity of the sensor against the oxidation of nonenzymatic glucose in various electrolyte solutions. Non-enzyme glucose sensors are highly resistant to poisoning by chloride ions and are able to overcome interference from uric acid and ascorbic acid up to concentrations of 0.05 mM and 0.5 mM, respectively. In this paper, the results of research on the electrochemical response of Au electrode in various electrolytes are KNO_3 , NaOH, KOH, buffer phosphate solution pH 4, 7 and 10. This study also conducted a study of the effect of glucose concentration on the peak oxidation and reduction currents. This study has produced data on the sensitivity and selectivity of Au electrodes for glucose analysis. Au metal can be used as a working electrode in the manufacture of nonenzyme glucose sensors.

2. Experimental Section

2.1. Material and Instrumentation

The instrument used in this research is a set of analysis tools cyclic voltammetry PGSTAT 100 N 100 V/250 mA (Metrohm Autolab) equipped with Nova Software and pH meter (Hanna). The materials used in this study were glucose (Merck), KNO_3 (Merck), KOH (Merck), buffer phosphate solution pH 4, 7 and 10, NaOH (Merck), KH_2PO_4 (Merck), distillate water and human urine. Human urine was collected from student.

2.2. Preparation of Gold Electrode

Au metal with purity 99.99% (Sigma Aldrich) cut with length and width 1 cm x 0.5 cm. Au metal is connected to the silver wire with silver conductive paint. Silver wire is inserted in the perforated glass. Silver conductive paint is covered with epoxy so that it comes into contact with only Au metal solution. Figure 1 shown is the design of gold electrode for cyclic voltammetry of glucose analysis.

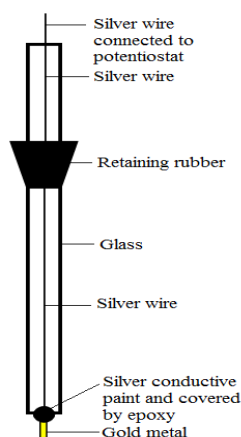


Figure 1. Design of gold electrode for glucose analysis using cyclic voltammetry

2.3. Cyclic voltammetry of Gold in Glucose in KOH, NaOH, and Buffer solution pH

Electrolysis cell and Au electrode are cleaned with distillate water and dried. As much as 5 mL of glucose 1M and 5 mL of 0.1 M KNO_3 solution were included in the electrolysis cell. The platinum electrode is the counter electrode, Au as the working electrode and Ag/AgCl as the reference electrode connected to the

potentiostat. The steps were repeated using different electrolyte solutions is KOH, NaOH and buffer phosphate solution pH 4, 7 and 10.

3. Result and Discussion

3.1. Cyclic Voltammetry Glukosa using Au electrode

Figure 2 shows the results of analysis with cyclic voltammetry glucose at Au electrode in KNO_3 solution (without glucose) and Figure 1B cyclic voltammogram glucose solution 0.001 M with electrolyte KNO_3 0.1 M. Figure 2 shows that the reversible voltammogram is that starts from a potential of -1.0 volts moving towards a positive direction then moves towards a more negative potential and back to potential -1.0 volts. In the peak voltammogram of the electrolyte solution showed that at a positive potential there is a peak of the electrolyte at a potential of 0.5 V. The peak for glucose and electrolyte at the same potential of 0.5 V and there is an anodic peak rise in current. The peak is the oxidation of glucose. The reaction of glucose oxidation on the surface of the Au electrode is shown in Figure 3. Figure 3 shows that glucose is oxidized on the surface of the Au electrode to gluconolactone. Glucose also undergoes a reduction shown by the appearance of a cathodic peak current at a potential of 0.35 V.

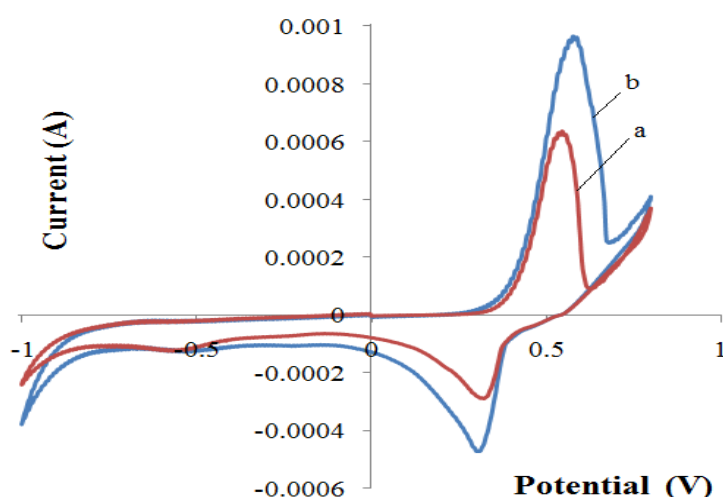


Figure 2. Cyclic voltammogram of (a) 0.1 M KNO₃ (without glucose), (b) 0.1 M KNO₃ + glucose 0.001 M glucose at scan rate 100 mVs⁻¹

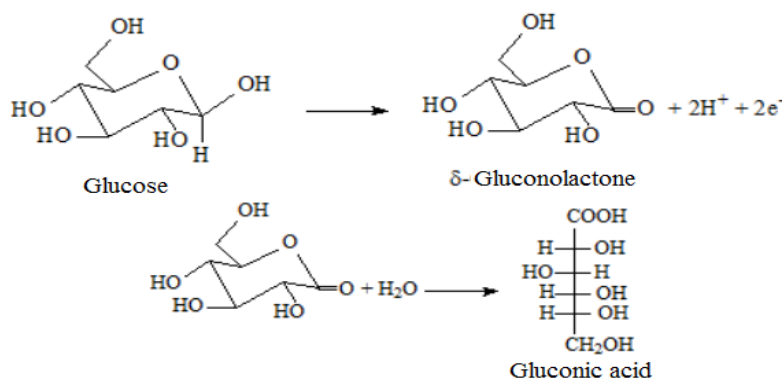


Figure 3. Redox reaction of glucose in gold electrode surface [11]

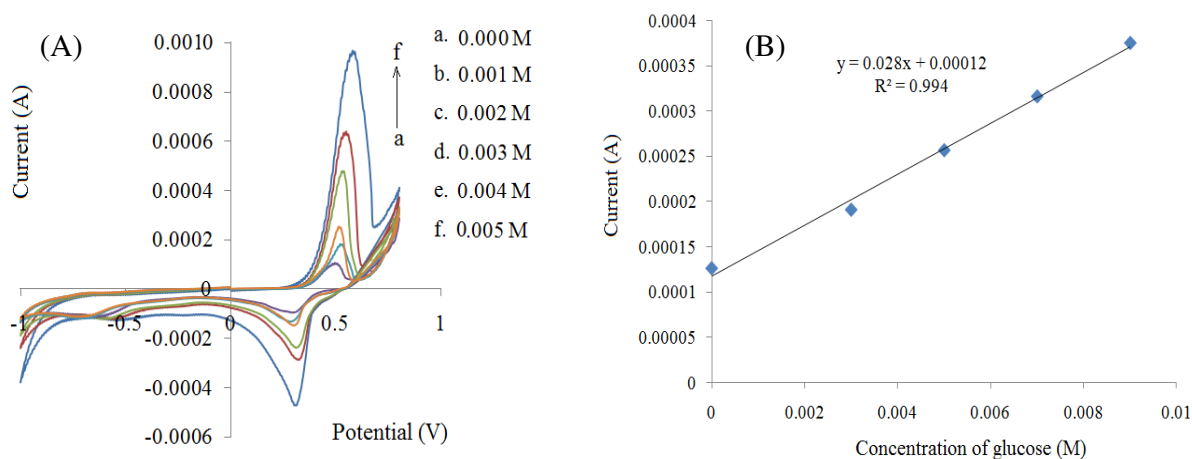


Figure 4. Cyclic voltammogram of various concentration of glucose at Au electrode in 0.1 M KNO₃(without glucose) (a) and (b-f) is 0.1 M KNO₃ + (0.001-0.005 M), at scan rate 100 mVs⁻¹

The method to ensure that the anodic current peak at 0.5 V potential is the oxidation of glucose, then cyclic voltammetry glucose with different concentrations is performed. Figure 2 shows that the greater the glucose concentration, the peak potential of oxidation and the peak potential of glucose reduction, the greater. The results of this study in accordance with the applicable formula for cyclic voltammetry are $i_p = 2.686 \times 10^5 n^{3/2} A c D^{1/2} v^{1/2}$.

Where i_p is the peak current A: electrode area, c: concentration, v: scan rate and D: diffusion coefficient. The above formula has a constant value that is, n, A, D and v so that it can be simplified to $i_p = kc$, the peak current is directly proportional to the concentration. Figure 4B shows the calibration curve of the effect of glucose concentration (M) on the anodic peak current. The anodic peak current rise is directly proportional to glucose concentration. Kumary et al. (2013) have research and that the linearity of the anodic peak current of glucose is directly proportional to the concentration [4]. Figure 3 shows that the correlation coefficient or $R^2 = 0.993$. KNO₃ is the best electrolyte for glucose analysis with gold electrodes.

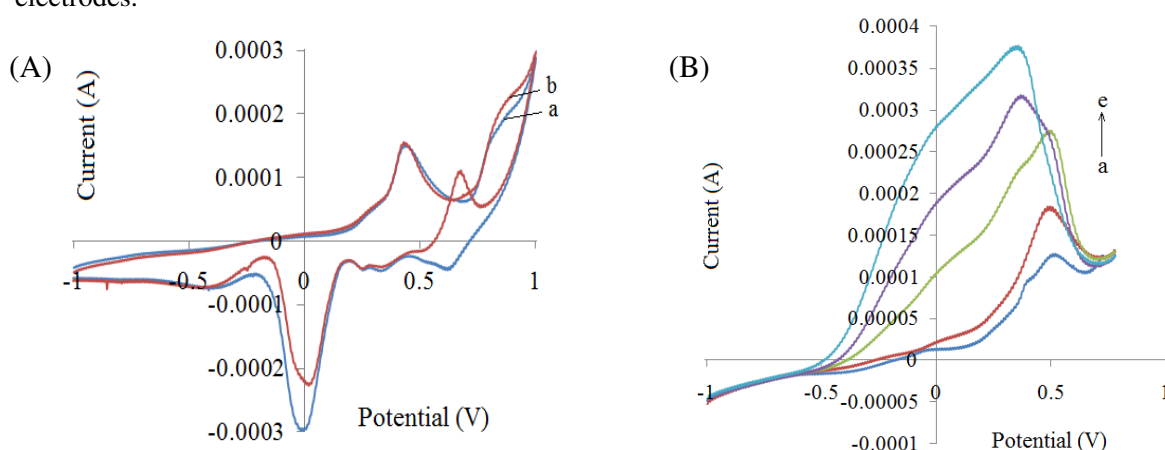


Figure 5. Cyclic voltammogram of (A) (a) 0.1 M NaOH (without glucose), (b) 0.1 M NaOH + glucose 0.001 M glucose. (B) Cyclic voltammogram of various concentration of glucose at Au electrode in 0.1 M NaOH solution in glucose (a-e) 0.003-0.009 M, at scan rate 100 mVs⁻¹

Figure 5 shows a cyclic voltammogram of glucose with NaOH electrolyte. Figure 5A shows the cyclic voltammogram comparison between the electrolyte NaOH and glucose. The peak of glucose oxidation is difficult to identify because of the nearly same voltammogram form. Figure 5B shows a cyclic voltammogram with various variations of glucose concentration. Figure 5 B shown is the greater the concentration of glucose, the higher the peak oxidation current. Glucose oxidation peaks occur at a potential of 0.4 V. Figure 6 shows cyclic voltammogram glucose with KOH electrolyte. The resulting cyclic voltammogram did not differ significantly between the NaOH and KOH electrolytes. The significant difference between the NaOH and KOH electrolytes is between Figure 5B and Figure 6B. Figure 5B shows a cyclic voltammogram having an oxidation peak and has no reduction peak. Figure 6B shows a cyclic voltammogram having an oxidation peak and a reduction peak.

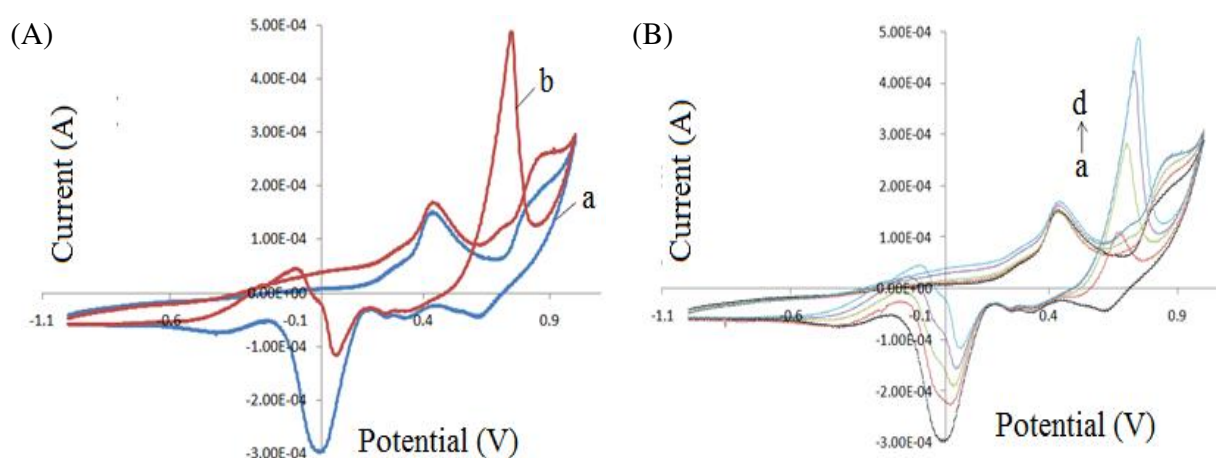


Figure 6. Cyclic voltammogram of (A) (a) 0.1 M KOH (without glucose), (b) 0.1 M NaOH + glucose 0.001 M glucose, (B) Cyclic voltammogram of various concentration of glucose at Au electrode in 0.1 M NaOH solution in glucose (a-e) 0.001-0.004 M, at scan rate 100 mVs^{-1}

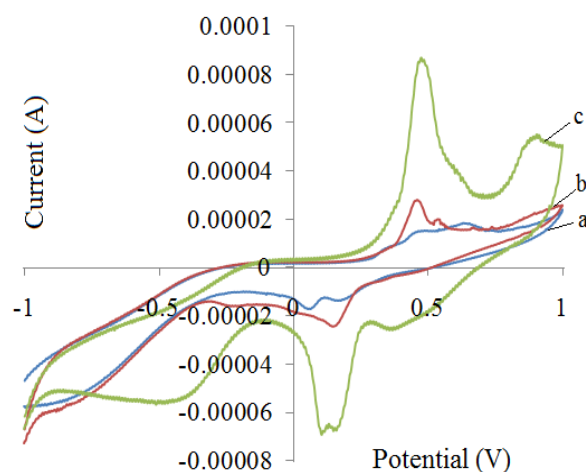


Figure 7. Cyclic voltammogram of 0.001 M glucose in buffer phosphate solution with various pH solutions (a) 4 (b) 7 (c) 10

Determination of effect pH on glucose analysis with Au electrode was performed at pH 4, 7, and 10. The tests were performed to determine the effect of pH on glucose response. Cyclic voltammogram comparison pH 4, 7, and 10 can be seen in Figure 7. Effect of pH on anodic and cathodic response can be seen at pH 7 and pH 10. Figure 7 (a) shown the buffer phosphate pH 4 shows a low anodic peak. The Au metal at the acidic solution has corrosion. Cyclic voltammogram at buffer phosphate pH 7 shows the anodic peak rise and at phosphate buffer pH 10 shows the highest anodic peak. The buffer phosphate pH 10 is an electrolyte suitable for glucose analysis with Au electrode.

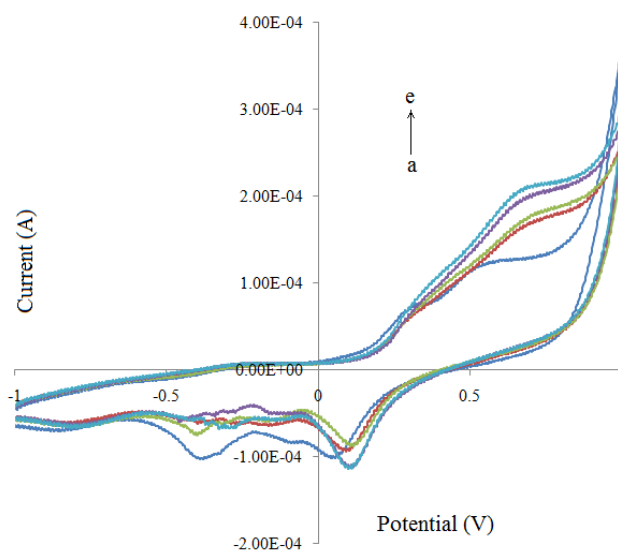


Figure 8. Cyclic voltammogram of (a) 0.1 M NaOH + human urine (without glucose), (b) 0.1 M NaOH + human urine + glucose 0.003 M, (c) 0.1 M NaOH + human urine + glucose 0.005 M (d) 0.1 M NaOH + human urine + glucose 0.007 M (e) 0.1 M NaOH + human urine + glucose 0.009 M, at scan rate 100 mVs⁻¹

Figure 8 shows a cyclic voltammogram by addition method to determine the concentration of glucose in the urine. Glucose concentration in human urine was determined using anodic peak with a glucose concentration of 0.003-0.009 M in 0.1 M NaOH electrolyte and scan rate 100 mVs⁻¹. The cyclic voltammogram of the analysis is presented in Figure 8. Table 1 shows the results of recovery of determination of glucose in human urine using cyclic voltammetry at Au electrode by the KNO₃ electrolyte. Determination of glucose in human urine by cyclic voltammetry at Au electrode by KNO₃ electrolyte has very good recovery value that is in the range 95-105%.

Table 1. Recovery of determination of glucose in human urine using cyclic voltammetry at Au Electrode by KNO₃ electrolyte

Urine Sample	Glucose added (mM)	Glucose founded (mM)	Recovery (%)
Sample 1	3.00	2.99	99.67
Sample 2	5.00	5.12	102.40
Sample 3	7.00	6.79	97.00
Sample 4	9.00	8.78	97.56

4. Conclusion

KNO₃ is a good electrolyte for determination of glucose in human urine using gold electrode. The effect of glucose concentration has the coefficient correlation is $R^2 = 0.994$. The results of the recovery showed the recovery at range 95-105% with addition method. As a conclusion a gold electrode is a good electrode for electrochemical sensors to the determination of glucose in human urine.

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