Imprinted zeolite modified carbon paste electrode as a potentiometric sensor for uric acid

Miratul Khasanah, Alfa Akustia Widati, and Sarita Aulia Fitri

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Preface: 5th International Conference and Workshop on Basic and Applied Sciences.

Foreword from Chairman of 5th ICOWOBAS 2015

Ladies and Gentleman,

The 5th International Conference and Workshops on the Basic and Applied Sciences (ICOWOBAS) is organized of existing collaborations between Airlangga University, Universiti Teknologi Malaysia and Salahaddin-Hawler University (Erbil, Iraqi Kurdistan) in order to promote the development of sciences and their prospect of application in industry and medical devices. The program of this activity are the scientific program involves the presentation of the paper and poster in the area of chemistry, biology, physics, mathematics and their applications. It also conducts the workshop program was presented the current issues in optical instrumentation. Thus invited many participants as academic researchers, scientists, industrial professionals, government officers, students and other participants. The meeting intends to bring together researcher, scientists and scholars to exchange and share their experiences, new ideas, research novelties in related fields and discuss the practical challenges and the solutions adopted.

The AIP proceedings hold the full papers presented at the 5th ICOWOBAS. The conference took place in Surabaya (Indonesia) at the Garuda Mukti Room, Kampus C Universitas Airlangga, October 16th - 17th, 2015, and the workshop was conducted at the Faculty of Science and Technology, Airlangga University, October 15th, 2015.

The conference included: Prof. Dr. Retna Apsari (Universitas Airlangga, Indonesia) as invited speaker. In total, we received 152 abstracts for oral and 29 posters, and 46 full paper selected in AIP proceeding. As the acceptance rates illustrates the competition is stiff, and the accepted submission reflected high rates of reviewer enthusiasm. By design, these papers have been through peer-review process and they are almost accepted.

ICOWOBAS is a lot of work. We could not have done it without help from many people. We would especially like to thank: Scientific board of ICOWOBAS, for inviting us to chair the meeting; The Rector of Airlangga University, for supporting us to conduct the conference; our colleagues in the Faculty of Science and Technology, Airlangga University, for their support in the conference; the local committee, for organizing and handling the conference; the many reviewers, for providing professional reviews; our sponsor: Vitalong C and DGHE through Airlangga University funding.

Surabaya, February 2016

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Imprinted Zeolite Modified Carbon Paste Electrode as a Potentiometric Sensor for Uric Acid

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Abstract. Imprinted zeolite modified carbon paste electrode (carbon paste-IZ) has been developed and applied to determine uric acid by potentiometry. The imprinted zeolite (IZ) was synthesized by the mole ratio of uric acid/Si of 0.0306. The modified electrode was manufactured by mass ratio of carbon, IZ and solid paraffin was 40:25:35. The modified electrode had shown the measurement range of 10^{-5} M to 10^{-2} M with Nernst factor of 28.6 mV/decade, the detection limit of 5.86×10^{-6} M and the accuracy of 95.3 – 105.0%. Response time of the electrode for uric acid 10^{-5} M - 10^{-2} M was 25 - 44 s. The developed electrode showed the high selectivity toward uric acid in the urea matrix. Life time of the carbon paste-IZ electrode was 10 weeks.

Keywords: *carbon paste, imprinted zeolite, modified electrode, potentiometry, uric acid*

1   Introduction

The high levels of uric acid in blood can causes of several health problems such as cardiovascular, high blood pressure (hypertension), and gout [1]. Normally, the level of uric acid in the blood is a range from 2.4 to 5.7 mg/dL in women and 3.4 to 7.0 mg / dL in men [2]. Electrometry is one of the methods have been reported for its determination. Most of attention has been focused on creating the high selectivity method to determine uric acid concentration in the blood because of coexisting of other compounds whose structure is similar with uric acid [3,4]. The selectivity and sensitivity of the electrode can be improved by modifying the electrode. There are some studies on utilizing zeolite as the electrode modifier material due to the unique characteristics of zeolite, has adsorbent properties, and high thermal stability [5].

On the basis of our previous report [6], we prepared imprinting zeolite-modified glassy carbon as sensor for analysis of uric acid by voltammetry. The developed sensor showed a high selectivity toward uric acid. The presence of ascorbic acid, creatine, and creatinine with an equal concentration with uric acid did not interfere on the uric acid analysis. Comparing the spectrophotometry analysis as a commonly method, our method shows the similar results when it used to analyze uric acid levels in serum. However, the use of glassy carbon as a supporting material of sensor unable to produce the composite electrodes, so the zeolite must be added in analysis process every time.

In this work, we developed a potentiometric method by imprinting zeolite (IZ) as an alternative material to modify the carbon paste electrode in attempt to produce a selective sensor for uric acid. The IZ was synthesized by mixing of TEOS as a silica source, TBOT as titania source, TPAH as a structure directing agent and uric acid as a template [7]. The trapped uric acid in the zeolite framework was extracted and produced imprinted in the zeolite framework. Furthermore, synthesized imprinted zeolite was mixed with carbon powder and molten paraffin in the manufacture of sensor. Carbon is an inert material, has a large surface area and high conductivity [8]. Conformity between the pore size of synthesized zeolite and molecular size of uric acid could increase the adsorption capacity of zeolite, which can provide high sensitivity for uric acid determination. Zeolite has rigid structure, so that in the aqueous media can maintain the shape and size of its pore (selective) [9]. The performance of developed sensor for uric acid analysis has been studied in this research.

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2   Experimental

2.1   Material and instrument

Chemicals used in this study were uric acid (Fluka, 98%), tetraethyl orthosilicate (Merck, 99%), tetrabutyl orthotitanate (Merck, 98%), tetrapropyl ammonium hydroxide (Merck, 40%), sodium hydroxide (Merck, 99%), acetic acid glacial (Merck, 100%), sodium acetate trihydrate (Merck, 99%), sodium dihydrogenphosphate dihydrate (Merck, 98%), sodium hydrogenphosphate dihydrate (Merck, 99,5%), 2-propanol (Merck, 99,8%), urea (Merck, 98%), silver wire (100%), solid paraffin, and carbon powder. All solutions were prepared with distilled water.

The instruments used were potentiometer Cyberscan 510, reference electrode Ag/AgCl, hotplate Lab Tech, magnetic stirrer, micropipette tube, pH meter Seven Easy Mettler-Teledo GmbH, sentrifuge Hittech EBA 20, oven NAPCO Vacuum Oven Model 5851, polypropilene bottle, and glassware.

2.2   Synthesis of Zeolite

Zeolite was synthesized by mixing TEOS into TBOT and stirred for 30 min at room temperature. TPAH was then added dropwise to the mixture, and stirred for 15 h, so that the mixture has a mole ratio of TEOS, TiO_2, TPAH and H_2O was 1:0.017:0.24:21.2. The mixture was aged at 80 °C for 4 days. In order to create the template uric acid molecule, uric acid was added to the mixture with molar ratio of uric acid/Si was 0.036. Subsequently, the mixture was aged for 3 h [7]. The uric acid was then extracted from the zeolite framework using warm water and dried at 80°C overnight.

2.3   Fabrication of carbon paste – IZ sensor

The sensor was manufactured by inserting a wire of silver to the micropipette tube. Solid paraffin was filled to the micropipette as much as 3/4 tube. Furthermore, the empty part of the tube was filled by a paste from the mixture of solid paraffin, carbon powder, and IZ with a mass ratio of 40:25:35 assisted by heating.

2.4   Optimization of pH

Optimization of pH of uric acid solution was performed to determine the effect of the pH on the electrode potential measurement. Optimization was done by measuring the potential of uric acid solution 10^{-3} M with pH 4, 5, 6, 7, and 8 by using carbon paste–IZ sensor.

2.5   The potentiometric performance of the carbon paste-IZ sensor

A series of uric acid standard solution with concentration of 10^{-8} to 10^{-2} M were analyzed by potentiometry using carbon paste - IZ sensor. The measured potentials were used to create a calibration curve and determine the performance of the sensor including measurement range, linearity, Nernst factor, detection limit, and accuracy.

Measurement range can be seen from the range of concentrations that exposes a linear curve and a slope value which obey the nernstian. The slope of the curve represents the magnitude Nernst factor. The detection limit was determined from the intersection of linear and non linear regression line on the curve of log [$C_{uric\ acid}$] versus potential. The accuracy was determined by measuring the uric acid solutions which are in the measurement range. Furthermore, the potential of uric acid is substituted into the equation 1.

$$Acc = \frac{C_{sp}}{K_s} x100\% \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \ (1)$$

Acc is the accuracy, C_{sp} is the concentration of uric acid standard solution analyzed, and K_s is the measured uric acid concentration.

2.6 Selectivity of the sensor

The selectivity of the sensor was studied by the addition of urea (compound which is always coexist with uric acid in serum sample) on the uric acid analysis. Determination of the coefficient of selectivity was done by measuring the potential of uric acid solution 5×10^{-5} M. Furthermore, the measurement of the potential of uric acid solution 5×10^{-5} M was added a solution of 1×10^{-3} M , 2×10^{-3} and 15×10^{-3} M urea, respectively. The potentials obtained were substituted into the equation 2.

$$K_{ij} = \frac{a_i(10^{(E2-E1)/s} - 1)}{a_j^{n/x}} \dots\dots\dots\dots\dots\dots\dots\dots\dots(2)$$

a^i is the activity of the uric acid, a_j is activity of urea , E1 is a potential of uric acid, E2 is potential of mixture of uric acid and urea, and s is the slope of the calibration curve of the uric acid [10].

2.7 Response and life time of the sensor

The response time of sensor is determined by analyzing concentration of uric acid within the range of measurement. The electrode potential of each solution was determined using carbon paste-IZ sensor for 5 minutes. The life time of sensor was studied by analyzing uric acid of 10^{-8} M to 10^{-2} M by potentiometry using carbon paste –IZ sensor in every week to know the performance of measurement range and the value of the slope of the calibration curve beyond the limits allowed Nernst factor.

3 Result and Discussion

3.1 Optimization of pH

Optimization of uric acid pH of the solution was conducted to determine the most stable pH range for analysis (Fig. 1). In this study, the optimization of pH was done at 10^{-3} M solution of uric acid with a pH range of 4 to 8. The pH measurement is done at 10^{-3} M solution of uric acid on the assumption that the solution of uric acid with other concentrations showed the identical response.
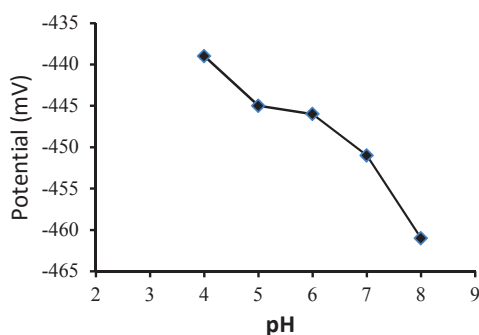

Figure 1 The effect of pH uric acid solution on electrode potential

Figure 1 represented that with an increase of pH through the decreasing of potential. It is noticed that pH 5 and 6 generated relatively constant potential. It can be explained that uric acid has a pKa 5.4 [11], therefore uric acid is present in the molecular form as itself in an acidic solution [12, 13]. Based on this result, the acidity of the uric acid was conditioned at pH 5 with the addition of acetate buffer. At this pH, the developed sensor recognizes uric acid in the form of molecules.

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3.2 Range of Measurement and Nernst Factor

Concentration of measurement range is the range that still exhibits a linear curve. An electrode has to be good if it has a wide measuring range. The sensor measurement range was studied using uric acid solution of pH 5. From the data was obtained the measuring range was 10^{-5} - 10^{-2} M and the Nernst factor was 28.6 mV/decade. The curve showed the correlation between log concentration of uric acid (log[C]) and measured electrode potential (Fig. 2).
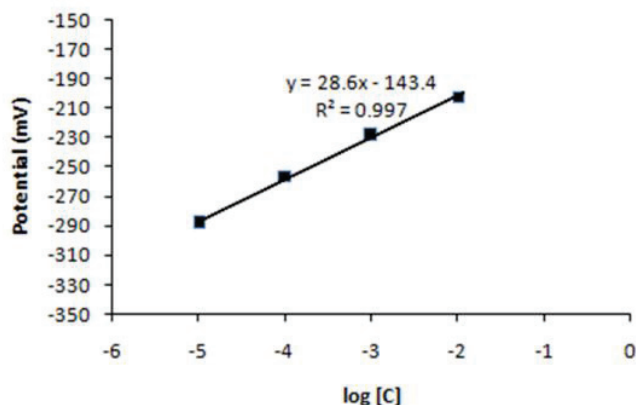

Figure 2 Calibration curve of uric acid determined by potentiometry

Because of uric acid is a divalent molecule (Fig 3), so that the Nernst factor obtained is approaching (59.2 /2) mV/decade, equivalent to 29.6 mV/decade (± 1-2 mV).
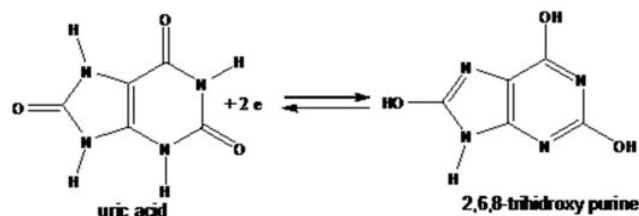

Figure 3 Redox reaction of uric acid

3.3 Limit of Detection and Accuracy

The detection limit was determined by the intersection of linear and non-linear curve of the curve relationship between log [C] and the potential electrodes. In this research, obtained equation linear curve of $Y = 28.6x - 143.4$, whereas non-linear equation of $Y = 1.5^{X^2} + 15.5x - 253$ (Fig 4). The intersection of both equations resulted detection limit of 5.86 x 10^{-6} M. This value is about 2 times lower than using spectrophotometry method [1]. It can be interpreted that analysis of uric acid using the developed sensor requires smaller volume of sample than needed in the spectrophotometric method as commonly used method in the medical field.
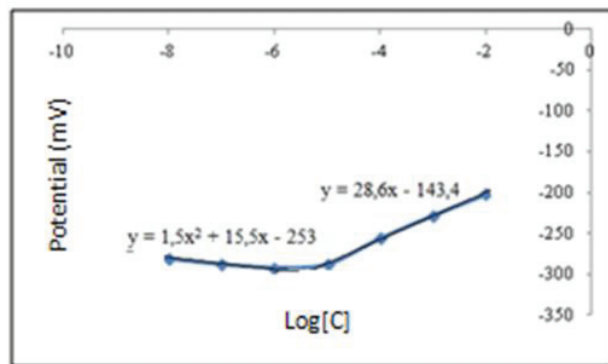
Figure 4 The curve of relationship of log [C] and potential on the determination of detection limit

The accuracy of the calculations the uric acid concentration of 10^{-5} M, 10^{-4} M, 10^{-3} M, and 10^{-2} M were 95.3; 98; 101.6; and 105% (n=3), respectively. The accuracy values are very good and statistically acceptable [14]. The developed sensor recommended as a new alternative sensor for uric acid analysis by potentiometry.

3.4   Selectivity

One of the major problems on the analysis of uric acid in the serum samples electrometrically is the presence of other compounds which are usually found together with uric acid in serum sample. In this study, to determine the selectivity of the developed sensor, the interference of urea on analysis uric acid by potentiometry has been studied.

TABLE 1. Selectivity coefficient of electrode toward uric acid 5×10^{-5} M in the urea matrix

Concentration of urea (M)	Coefficient of selectivity ($K_{i,j}$)
1×10^{-3}	-1.3400
2×10^{-3}	-0.9512
15×10^{-3}	-0.5956

Table 1 showed that value Kij of the third urea concentration is less than 1, it means that the carbon paste electrodes -IZ more selective towards molecules of uric acid than urea molecules. Thus the presence of urea did not interfere to the analysis of uric acid by potentiometry using carbon paste-IZ sensor.

3.5   Response and life time of the sensor

The response time can be used to determine the sensitivity of the sensor. A sensor is said to be more sensitive when the time required to respond to the analyte is the less. The response time of the sensor to determine uric acid 10^{-5}, 10^{-4}, 10^{-3} and 10^{-2} M were 44, 39, 34, and 25 s, respectively. The higher the concentration of uric acid, the faster the response of sensor. The higher concentration, the shorter time of uric acid to achieve equilibrium on the electrode surface because of the mobility of molecule becomes faster.

Life time of the sensor stated the time of the electrode is still has good performance as a sensor in potentiometric measurement which in study by a good Nernst factor for the concentration range of 10^{-5} - 10^{-2} M. The result showed that the sensor had a good Nernst factor for 10 weeks with 127 times of measurement. The life time of the electrode depends on the mechanical properties of an electrode material, such as the flexibility of the material, pH, and the solubility of the material. Flexibility of the material causing the analyte is easier to diffuse into the template. The use of carbon paste-IZ sensor in more acidic and alkaline solution will affect the life time of the sensor, because the zeolite material used is resistant to acidic solution, while carbon is a material that soluble in alkaline.

4   Conclusion

The carbon paste-IZ showed good performance as sensor for uric acid analysis by potentiometry. The uric acid analysis using the sensor has a high sensitivity, accuracy, and low detection limit. The limit of detection obtained was low enough for the purpose of uric acid determination in serum samples. Analysis of uric acid using the developed sensor was not interfered by urea. The sensor response time was less than one minute, while the life time was more than 10 weeks. Potentiometry method using the developed sensor is suggested to be used as an alternative method to determine of uric acid in the medical field.

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