THE INFLUENCE OF ASCORBIC ACID, CREATINE, AND CREATININE ON THE URIC ACID ANALYSIS BY POTENTIOMETRY USING ACARBON PASTE MODIFIED IMPRINTING ZEOL

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THE INFLUENCE OF ASCORBIC ACID, CREATINE, AND CREATININE ON THE URIC ACID ANALYSIS BY POTENTIOMETRY USING A CARBON PASTE MODIFIED IMPRINTING ZEOLITE ELECTRODE

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ABSTRACT

The high level of uric acid in the body is often associated with some diseases such as hyperuricemia, hypertension, renal and cardiovascular disease. Therefore early detection of the levels of uric acid is required. Spectrophotometry is the method commonly used in the medical field to determine the uric acid level. Its detection limit refers to a low uric acid level in a serum sample as the complexity of the serum matrices decreases its selectivity. The development of an imprinted zeolite based sensor to analyze uric acid by potentiometry is conducted. The carbon paste-IZ electrode shows a range of measurement of 10^{-6} M - 10^{-4} M, a Nernst factor of 28.2 mV decade^{-1}, and response time of 34 s - 44 s. The developed electrode has a high selectivity towards uric acid. Ascorbic acid, creatine, or creatinine do not interfere with uric acid analysis using the electrode suggested as the value of the selectivity coefficient (K_{ij}) is less than 1. The method recovery equals (113.18±14.79) % (n = 3). The developed method is compared to the spectrophotometric one and shows an accuracy of (94.47±14.64) % (n = 7).

Keywords: uric acid, carbon paste-IZ, selectivity, potentiometry.

INTRODUCTION

Uric acid is the end product of purine metabolism. The normal level of uric acid in the blood is in the range of 2.4 mg/dL to 5.7 mg/dL for women and 3.4 mg/dL to 7.0 mg/dL for men [1]. Uric acid has been used as a biomarker diagnosing health problems because of its ability to cause some dangerous diseases. Thus, the levels of uric acid in the body should always be controlled. Spectrophotometry using a chemical reagent or the enzymatic method are the common current methods used in field of medicine to analyze uric acid content. The method used to determine uric acid levels must be selective because uric acid is found together with other interfering compounds [2]. Creatine and creatinine are also potentially interfering uric acid analysis because of the similarity of their functional groups and structure. These compounds presence hamper obtaining accurate results in respect to uric acid levels.

Various electrochemical methods are developed to solve the problem of uric acid analysis. Voltammetry application presents a definite interest because it requires less chemicals and time compared to the colorimetric and enzymatic methods [3, 4]. However, the major problem encountered by this method is the interference of ascorbic acid and urine which can be oxidized at the potentials close to that of the uric acid [5].

A previous study [6] have advances a method of uric acid content determination by potentiometry using ZnO nano wires electrode immobilized by the enzyme uricase. Its measurement range refers to 1.0×10^{-6} M – 6.5×10^{-4} M which makes it very suitable for uric acid determination in the blood serum. Glucose, ascorbic acid and urea do not interfere in case of this method application. Another study reports the potentiometric use of a ZnO nanoflakes-based sensor immobilized by

the enzyme uricase [7]. The results show that the sensor has a measuring range of $5.0x10^{-7}$ M – $1.5x10^{-3}$ M and a detection limit of $5.0x10^{-7}$ M. The sensor function is not interfered by the presence of ascorbic acid, glucose and urea. A potentiometric method using carbon paste electrodes modified by imprinting zeolite (carbon paste-IZ) is also developed to measure uric acid levels [8]. Its application is not affected by the presence of urea. The zeolite has a rigid structure and it can maintain the shape and size of its pores (i. e. its selectivity) [9] in an aqueous medium. The conformity of the zeolite print size and shape and those of the uric acid molecule increases zeolite's adsorption capacity which in turn provides high sensitivity in respect to uric acid determination.

The present work reports a study on the influence of ascorbic acid, creatinine and urea on uric acid analysis by potentiometry using a carbon paste electrode modified by an imprinting zeolite. The preparation of latter is already described [8]. The lof ascorbic acid, creatinine and creatine is followed by introducing each compound of a varying concentration to the uric acid standard solution.

EXPERIMENTAL

Material and methods

Uric acid (Fluka), creatine and creatinine (Sigma-Aldrich), ascorbic acid, acetic acid, sodium acetate, and sodium hydroxide trihydrate, carbon powder, solid paraffin, tetraethyl orthosilicate (TEOS), tetrabuthyl orthotitanate (TBOT), tetrapropyl ammonium hydroxide (TPAH) (all products of Merck) were used. They were of an analytical grade. Distilled water was used as a solvent. The stock solution of 10^{-2} M uric acid was prepared by dissolving 0.1680 g uric acid in about 10 mL 50 % (w/w) aqueous solution of sodium hydroxide (a 100 mL volumetric flask was used for the preparation of the latter). The standard 10^{-8} M - 10^{-3} M solutions of uric acid were prepared by diluting appropriate uric acid working solutions, and their pH were adjusted through the addition of acetate buffer of pH 5. Urine from patients of a local pathological clinic was used as a sample.

Fabrication of carbon paste – IZ electrode

Carbon paste electrode was manufactured by mixing carbon powder, imprinting zeolite and paraffin in a mass ratio of 40:25:35 [8]. The process was assisted by heating. The zeolite was synthesized by mixing TEOS, TBOT and TPAH in mole ratios reported in a previous study [10]. The uric acid was then extracted from the zeolite framework using warm water to produce imprinting zeolite. The electrode investigated was prepared by inserting a silver (Ag) wire in a micropipette tube filled to its 3/4 by solid paraffin and subsequent introduction of by the paste already described.

Determination of the measurement range, the Nernst factor and the response time of the electrode

Measurement range was determined by measuring the electrode potential in a uric acid solution of a concentration ranging from 10^{-8} M to 10^{-3} M and the subsequent presentation of the dependence of the logarithm of uric acid concentration (log [uric acid]) on the electrode potential (E). The measurement range referred to the concentration interval where the relation pointed above was linear, while the slope of that line was equal to the Nernst factor value. The response time of electrode was determined by the time required to obtain a potential value referring to uric acid presence.

Selectivity of the electrode

The selectivity of the electrode, expressed by the selectivity coefficient (K_{ij}), was studied through the addition of ascorbic acid, creatine and creatinine, compounds which always coexist with uric acid in urine or in a serum sample. Uric acid used was 10^{-4} M, while the ascorbic acid, the creatine and the creatinine were added in varying concentrations. The K_{ij} value was calculated by Matched Potential Method (MPM) [11].

Method recovery and comparative test

The recovery of the method was followed by measuring the potential in a urine sample, a uric acid standard solution, and urine samples spiked by a uric acid standard solution. The urine samples were taken from the urine of adults collected over 24 hours. The potential of the carbon paste-IZ electrode was read and its value was substituted into the linear regression equation describing the standard curve aiming to obtain the concentration of

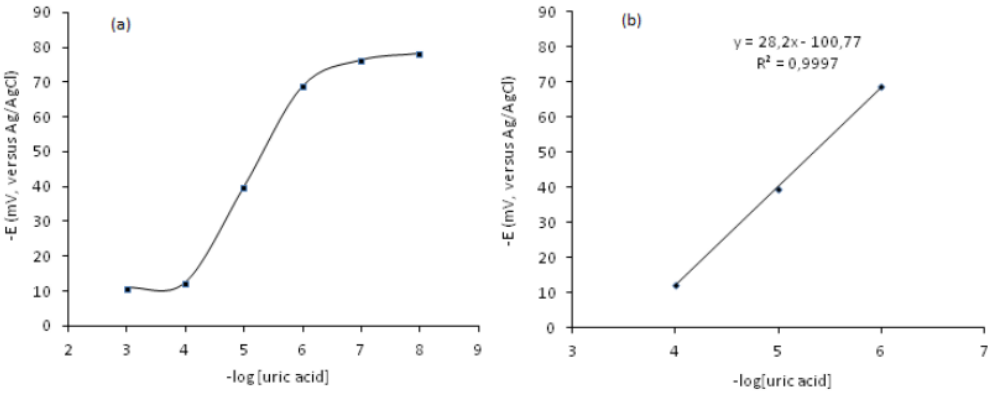

Fig. 1. Curve of –E versus –log [uric acid] (a) and calibration curve (b).

each solution. The recovery (R) value was calculated by substituting the concentration of each solution into Eq. 1:

$$R = \frac{c_{ss} - c_{sp}}{c_{std}} x100\% \quad (1)$$

where R was the recovery, C_{ss} was the concentration of the spiked sample, C_{sp} was the concentration of the urine sample, while C_{std} was the concentration of the standard solution.

The method described was compared to the UV-Vis spectrophotometrical one to determine its accuracy. The latter was calculated by referring the concentration determined potentiometrically (C_{dev}) to that obtained spectrophotometrically (C_{std}) (Eq. 2):

$$A = \frac{c_{dev}}{c_{std}} x100\% \quad (2)$$

RESULTS AND DISCUSSION

Performance of the electrode

The performance of the electrode assembled is studied by measuring its potential in solutions of uric acid of a concentration ranging from 10^{-8} M to 10^{-3} M using Ag/AgCl as a reference electrode. The relationship between –log [uric acid] and –E (potential) is presented in Fig. 1a.

The measurement range found refers to the concentration interval of 10^{-6} M - 10^{-4} M (Fig. 1b), where the linearity is characterized by r value of 0.9995. The response time is found equal to 34 s - 44 s, while the Nernst factor estimated is equal to 28.2 mV decade^{-1}. In view of the fact that uric acid is a divalent molecule [12, 13], the Nernst factor is expected to be equal to (29.6±2) mV decade^{-1}. As previously reported the method has a limit of detection of $5.86x10^{-6}$ M. precision of 98.6 to 98.8 % (n=3), and a life time of 10 weeks [8].

Selectivity of the electrode

Selectivity is the ability of electrodes to measure an analyte selectively wherein the analyte coexist with other components in a sample. One of the most important characteristics in this respect electrode response to primary compound (i) in presence of other compounds (j), which is measured in terms of the potentiometric selectivity coefficient (K_{ij}). The estimation of the coefficient of selectivity is required due to the presence of uric acid in the body together with other molecules of functional groups similar to those of uric acid, such as urea, creatine, creatinine, and ascorbic acid [14]. The selectivity coefficient is determined in this research by the Matched Potential Method (MPM) [11]. If the selectivity coefficient value is less than 1, the analysis of uric acid is not hampered by the presence of the interfering compounds.

Creatine, creatinine, and ascorbic acid are the compounds used to study the effect of the matrix in this investigation. The concentration of uric acid used coincides with that in the body which is 10^{-4} M. The

Table 1. Coefficient of selectivity (K_{ij}) of carbon paste-IZ electrode obtained in the course of analysis of uric acid in presence of ascorbic acid, creatine, and creatinine as interfering compounds.

Interfering compound (M)	K_{ij}
Ascorbic acid	
$5.0x10^{-6}$	0.1769
$1.0x10^{-5}$	0.3847
$1.0x10^{-4}$	0.7273
$5.0x10^{-4}$	0.2597
Creatine	
$1.0x10^{-5}$	0.2002
$3.0x10^{-5}$	0.1004
$1.0x10^{-4}$	0.0870
$3.0x10^{-4}$	0.1749
Creatinine	
$1.0x10^{-5}$	0.3267
$3.0x10^{-5}$	0.2093
$1.0x10^{-4}$	0.1023
$3.0x10^{-4}$	0.2059

*) *Concentration of uric acid used was 10^{-4} M*

concentrations of creatine and creatinine used are $1x10^{-5}$ M, $3x10^{-5}$ M, $1x10^{-4}$ M and $3x10^{-4}$ M, while those of ascorbic acid refer to $5x10^{-6}$ M, $1x10^{-5}$ M, $1x10^{-4}$ M, and $5x10^{-4}$ M. The choice of the concentrations values pointed above is determined by these substances low, normal and high levels in real urine and serum samples. The value of the selectivity coefficient (K_{ij}) for each concentration is shown in Table 1.

It is seen that the selectivity coefficient is less than 1 for each concentration of the compounds suspected to interfere with the analysis of uric acid. This means that creatine, creatinine, as well as ascorbic acid do not hamper uric acid analysis by potentiometry using the electrode assembled. It is so because the electrode recognizes specific molecules, namely uric acid.

Table 2. Values of the electrode potential obtained in the course of analysis of a urine sample.

Solution	E (mV, vs Ag/AgCl)
Urine	23.90
Urine + uric acid 10^{-6} M	23.91
Uric acid 10^{-6} M	-62.70
Urine + uric acid 10^{-5} M	24.01
Uric acid 10^{-5} M	-37.20
Urine + uric acid 10^{-4} M	25.00
Uric acid 10^{-4} M	-7.60

Comparative test and recovery of the method

The recovery of the method applied is identified by measuring the electrode potential in a uric acid standard solution, a urine sample, and a urine sample spiked by a uric acid standard solution. The concentration of the uric acid standard solution used is 10^{-6} M - 10^{-4} M. The electrode potential values obtained are listed in Table 2.

The data referring to the juxtaposition of the method considered and the spectrophotometrical analysis usually applied are summarized in Table 3. It is evident that 3 the recovery of the method advanced reaches 96.18 - 123.04 % in case of spiking uric acid solutions of three different concentrations to a urine sample. The results

Table 3. Data illustrating the potentiometric method recovery and its juxtaposition to spectrophotometry.

Sample	Concentration (M) Potentiometry	Concentration (M) Spectrophotometry *)	Recovery (%) Potentiometry	Accuracy (%) Toward spectrophotometry
Urine	$1.89x10^{-3}$	$2.39x10^{-3}$		79.00
Urine + uric acid 10^{-6} M	$1.89x10^{-3}$	$1.94x10^{-3}$	96.18	97.28
Uric acid 10^{-6} M	$1.61x10^{-6}$	$1.92x10^{-6}$		83.51
Urine + uric acid 10^{-5} M	$1.91x10^{-3}$	2.30x10-3	120.34	82.80
Uric acid 10^{-5} M	$1.29x10^{-5}$	$1.31x10^{-5}$		98.42
Urine + uric acid 10^{-4} M	$2.08x10^{-3}$	$2.10x10^{-3}$	123.04	98.41
Uric acid 10^{-4} M	$1.44x10^{-4}$	$1.18x10^{-4}$		121.89

*) *data from clinical laboratory*

referring to the comparative testing of potentiometry and spectrophotometry in case of 7 sample solutions show accuracy of 79.00 % - 121.89 %. The range of accuracy required by the Association of Official Analytical Chemist (AOAC) for the concentration range of 10^{-6} M - 10^{-4} M is 80-110 % [15].

CONCLUSIONS

A carbon paste electrode modified by an imprinting zeolite is prepared. It has a high selectivity towards uric acid. The presence of ascorbic acid, creatine, or creatinine does not affect uric acid analysis by potentiometry using the electrode assembled. The recovery of the potentiometry method amounts to (113.18±14.79) % (n=3). The comparison of the method to the spectrophotometrical analysis usually applied shows an accuracy of (94.47±14.64) % (n=7). It is concluded that the method can be applied to determine uric acid in urine or a serum sample.

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