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Electrochemical Sensing of Uric Acid Using Glassy Carbon Modified with Multiwall Carbon Nanotubes Dispersed in Polyethylenimine

Alejandro Gutiérrez,[a] Maria L. Lozano,[b] Laura Galicia,*[a] Nancy F. Ferreyra,[c] and Gustavo A. Rivas*[c]

Abstract: This work reports the advantages of using glassy carbon electrodes modified with multiwall carbon nanotubes (MWCNT) dispersed in polyethylenimine (PEI). The presence of MWCNTs wrapped by PEI largely facilitated the strong adsorption of uric acid (UA) and allowed its highly sensitive and selective quantification even in the presence of high excess of ascorbic acid. The selected conditions for the electrochemical sensing were 5 s accumulation at −0.300 V under stirring and quantification in a 0.050 M phosphate buffer solution pH 7.40 by differential pulse voltammetry adsorptive-stripping after medium exchange. The platform allowed the successful application in the quantification of UA in urine.

Keywords: Carbon nanotubes · Non-covalent functionalization · Carbon nanotubes dispersion · Polyethylenimine · Uric acid · Electrochemical sensor

1 Introduction

Uric acid (UA), the major nitrogenous compound in urine, is the primary end product of purine metabolism in the human body. The presence of abnormal UA levels is a sign of gout, hyperuricemia or Lesch–Nyhan syndrome. Therefore, simple, fast and sensitive methods are highly required for the quantification of this bioanalyte [1].

Several analytical methods have been proposed for the determination of UA, ranging from the simple colorimetric methods based on commercially available enzymatic kits [2] to fluorometry and chemiluminescence [3], flow-injection [4], or even high-resolution separation methods, such as capillary electrophoresis [5] high-performance liquid chromatography (HPLC) [6] and ion-exclusion HPLC [7].

The electrochemical sensors have demonstrated to be an interesting alternative for the quantification of UA. However, they present the disadvantage of the interference of other electroactive compounds like ascorbic acid (AA), which is oxidized at potentials similar to UA [8–19]. Several strategies have been proposed to overcome this problem [8,20] involving the modification of electrodes with polymeric membranes [10,15,21–25], nanoparticles [19,26–31], cyclodextrines [19,20,32] and carbon nanostructures [9,10,12–19,25,28,31,33–38]. Rodríguez et al. [25] have reported the highly selective and sensitive UA quantification in the presence of AA using glassy carbon electrode (GCE) modified with multiwall carbon nanotubes (MWCNT) dispersed in polylysine (Plys) by differential pulse voltammetry-adsorptive stripping. The electrocatalytic activity of MWCNT deposited on GCE allowed an important decrease in the overvoltage for AA oxidation making possible the clear separation of the oxidation processes of AA and UA. The use of cationic polymers as dispersing agents of MWCNTs present two advantages, the efficient dispersion of the nanotubes and the favorable electrostatic accumulation of bioanalytes, once immobilized at the top of the electrodes, demonstrating an important advantage compared to other dispersing agents [39].

In this work we propose the use of GCE modified with MWCNTs dispersed in polyethylenimine (PEI), for the highly sensitive and selective UA quantification by Differential Pulse Voltammetry-Adsorptive Stripping (DPV-AdS) in the presence of a large excess of AA.

2 Experimental

2.1 Reagents

Ascorbic acid (AA) was obtained from Fluka. Uric acid (UA) was purchased from Quimica Meyer. Polyethylenimine (PEI, average MW 750,000, catalog number P-3143)

[a] A. Gutiérrez, L. Galicia
Universidad Autónoma Metropolitana Iztapalapa, Depto. de Química
Av. Michoacán y la Purísima, Col. Vicentina, C.P. 09340, México
*e-mail: lgl@xanum.uam.mx

[b] M. L. Lozano
Tecnológico de Estudios Superiores del Oriente del Estado de México, Depto. de Ingeniería Ambiental
Col. Barrio de Tecamachalco, La Paz, Estado de México, C.P. 56400, México

[c] N. F. Ferreyra, G. A. Rivas
INFIQC, Departamento de Físico Química, Facultad de Ciencias Químicas, Universidad Nacional de Córdoba
5000 Córdoba, Argentina
*e-mail: grivas@fcq.unc.edu.ar

was purchased from Sigma. Multiwall carbon nanotubes powder (MWCNT, (30±15) nm diameter, 5–20 microns length) were obtained from NanoLab (USA). Other chemicals were reagent grade and used without further purification. Ultrapure water ($\rho = 18\ M\Omega\,cm$) from Millipore-MilliQ system was used for preparing all the solutions. A 0.050 M phosphate buffer solution pH 7.40 was used as supporting electrolyte.

2.2 Apparatus

The electrochemical measurements were performed with a 100B potentiostat (BAS). Glassy carbon electrodes (GCE, CHI 3 mm diameter) either bare or modified with PEI or the dispersion of MWCNTs in PEI were used as working electrodes. A platinum wire and a Ag/AgCl, 3 M NaCl (BAS, Model RE-5B) were used as counter and reference electrodes, respectively. All potentials are referred to the latter. A magnetic stirrer provided the convective transport during the amperometric measurements.

2.3 Modification of the Working Electrodes

2.3.1 Preparation of MWCNT-PEI Dispersion

It was obtained by mixing 1.0 mg of MWCNTs within 1.0 mL of PEI solution (1.0 $mg\,mL^{-1}$ prepared in 50/50 v/v ethanol/water) followed by sonication for 15 min.

2.3.2 Preparation of Glassy Carbon Electrodes Modified with MWCNT-PEI (GCE/MWCNT-PEI)

The glassy carbon electrodes were polished with alumina slurries of 1.0, 0.30, and 0.05 μm for 2 min each; cycled five times in a 0.050 M phosphate buffer solution pH 7.40 between −0.300 V to 0.800 V and finally modified by dropping 20 μL of MWCNT-PEI dispersion on the top. The solvent was allowed to evaporate at room temperature for 1h. The modified electrodes were cycled for ten times between −0.300 V and 0.800 V at 0.050 $V\,s^{-1}$ before starting the electrochemical experiments. The GCE modified with PEI (GCE/PEI) was prepared in a similar way by dropping 20 μL of a 1.0 $mg\,mL^{-1}$ PEI solution (prepared in 50/50 v/v ethanol/water and sonicated for 15 min).

2.4 Procedure

The quantification of UA involved the following steps:

- *Preconcentration of UA:* was performed at −0.300 V for a given time in a UA solution prepared in 0.050 M phosphate buffer solution pH 7.40 under stirring.
- *Washing:* with 0.050 M phosphate buffer solution pH 7.40 for 10 seconds.
- *Stripping:* in a 0.050 M phosphate buffer solution pH 7.40 by differential pulse voltammetry (DPV). The DPV parameters are the following: scan rate of 0.020 $V\,s^{-1}$, pulse width of 50 mV, 100 ms pulse period, 2 s setting time and a pulse amplitude of 50 mV, without stirring.

All the experiments were conducted at room temperature.

3 Results and Discussion

Figure 1 shows cyclic voltammograms obtained for a mixture of 1.00×10^{-3} M AA + 1.00×10^{-4} M UA at different electrodes, GCE (red), GCE/PEI (blue) and GCE/MWCNT-PEI (orange) at a scan rate of 0.050 Vs^{-1}. At GCE there is a broad peak at around 0.6 V since AA and UA are oxidized at very close potentials, indicating that is not possible to use the bare GCE to detect a mixture of these compounds. Similar behavior was observed at GCE/PEI, with a broad peak at around 0.3 V that involves the two oxidation processes. Compared to bare GCE, there is a shifting to more negative potentials due to the facilitated interaction of ascorbate and urate anions at the positively charged PEI-modified GCE. On the contrary, the potentiodynamic profile obtained for AA + UA mixture at GCE/MWCNT-PEI displays two very well-defined current peaks at −0.050 V and 0.253 V for the oxidation of AA and UA, respectively. The assignment of the signals was performed by comparison with the electrochemical response obtained for each analyte at GCE/MWCNT-PEI under the same concentration (not shown). In addition to the decrease in the oxidation overvoltage, there is a significant enhancement in the oxidation currents. In fact, the AA oxidation currents change from 12.8 μA and 12 μA at GCE and GCE/PEI, respectively, to 44 μA at GCE/MWCNT-PEI. In the case of UA the currents change from 1.7 μA and 1.6 μA at bare GCE and GCE-PEI, respectively, to 21 μA at GCE/MWCNT-PEI. Therefore,

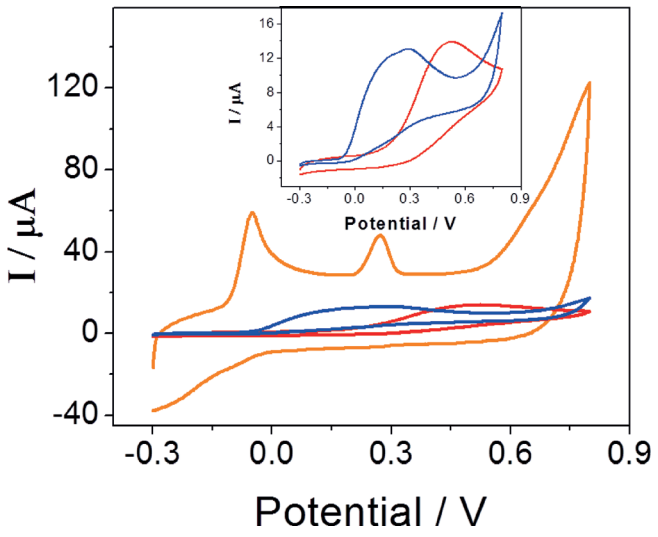

Fig. 1. Cyclic voltammograms for 1.00×10^{-3} M AA + 1.0×10^{-4} M UA at GCE (red), GCE/PEI (blue) and GCE/MWCNT-PEI (orange). Supporting electrolyte: 0.050 M phosphate buffer pH 7.40. Scan rate: 0.050 Vs^{-1}.

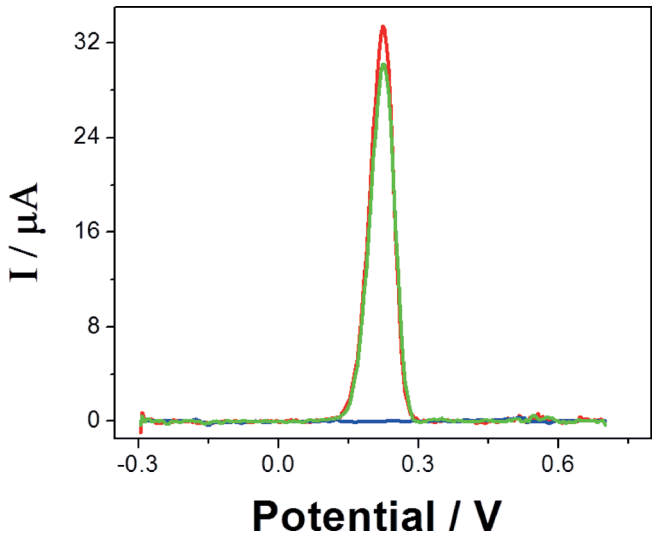

Fig. 2. DPV recordings obtained at GCE/MWCNT-PEI in a 0.050 M phosphate buffer solution pH 7.40 after accumulation of 1.00×10^{-3} M AA (blue), 5.0×10^{-5} M UA (green) and 1.00×10^{-3} M AA + 5.0×10^{-5} M UA (red) and medium exchange. Accumulation conditions: 5 seconds at −0.300 V. Stripping conditions: Scan rate: 0.020 V s^{-1}, pulse width: 0.050 V, pulse period: 100 ms, setting time: 2 s; pulse amplitude: 0.050 V.

the modification of GCE with MWCNT-PEI offers the great advantage of a clear definition of AA and UA oxidation processes due to the catalytic activity of MWCNTs and the presence of the positively charged polymer that support the MWCNTs. The catalytic effect of MWCNTs and the increment in the electroactive area also makes possible a large increase in the AA and UA oxidation currents.

Based on the important separation of the oxidation peak potentials for AA and UA, (0.283 V), we evaluated the adsorptive behavior of UA and AA at GCE/MWCNT-PEI to improve the simultaneous determination of both compounds. Figure 2 depicts DPV recordings in a 0.050 M phosphate buffer solution pH 7.40 obtained after accumulation of 1.00×10^{-3} M AA (blue), 5.0×10^{-5} M UA (red) and 1.00×10^{-3} M AA + 5.0×10^{-5} M UA (green) at GCE/MWCNT-PEI for 5 seconds at −0.300 V and medium exchange. Even when the concentration of AA during the accumulation step was 20 times higher than that for UA, no response is observed for AA, indicating a poor adsorption at the electrode surface. The DPV obtained after the adsorption of UA shows a clear signal at 0.224 V (peak current = 31 ± 2) µA, due to the strong adsorption of UA. The DPV obtained after the preconcentration step in a mixture of 1.00×10^{-3} M AA + 5.0×10^{-5} M UA shows a peak at 0.224 V with an associated current of (34 ± 3) µA, clearly demonstrating that the presence of a large excess of AA does not affect the oxidation of UA. These results are a clear evidence that it is possible to perform the determination of UA even in the presence of a large excess of AA due to the different adsorption behavior of both analytes at GCE/MWCNT-PEI.

We studied the influence of the potential and time during the preconcentration step. Figure 3A shows the optimization of DPV-AdS with medium exchange for 5.0×10^{-5} M UA oxidation as a function of the preconcentration potential between −0.300 and 0.000 V. The highest signal was obtained at −0.300 V and this potential was selected for further work. The influence of the preconcentration time at −0.300 V for 5.0×10^{-5} M UA is displayed in Figure 3B. The oxidation current increases up to 5 s and then it trends to level off. Thus, the selected conditions for the determination of UA by anodic adsorptive stripping were: 5 s preconcentration at −0.300 V at GCE modified with 1.0 mg mL^{-1} MWCNT-PEI dispersion.

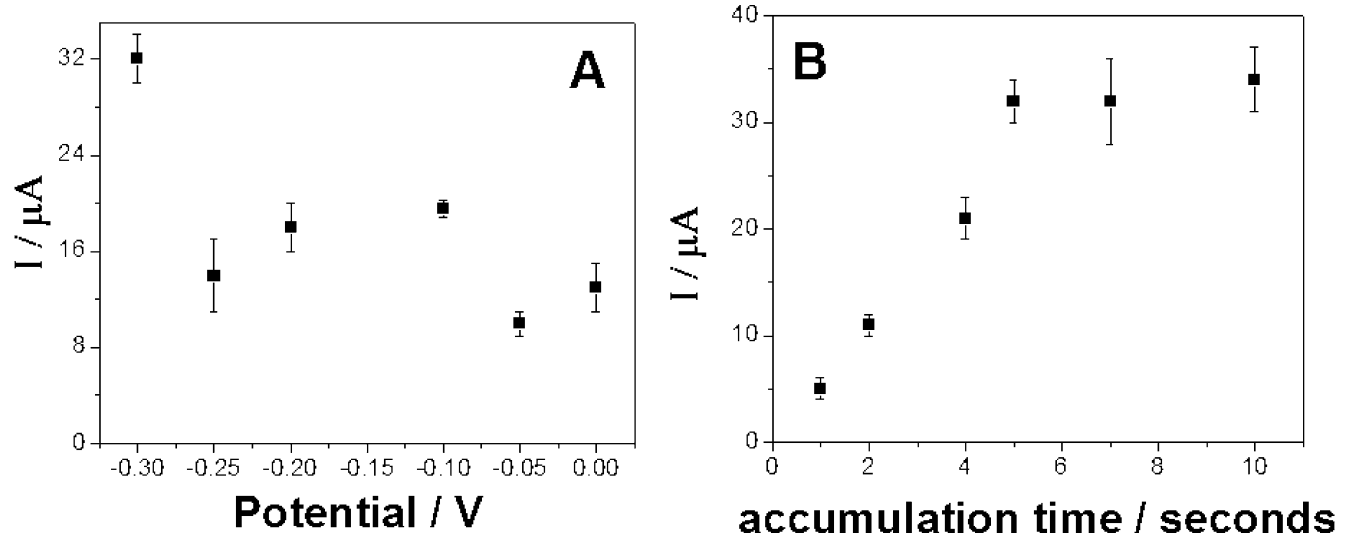

Fig. 3. Variation of the oxidation peak current of 5.0×10^{-5} M UA at GCE/MWCNT-PEI, obtained by DPV-AdS with medium exchange, as a function of: A) Accumulation potential for 5 sec and B) Accumulation time at a potential of −0.300 V. Supporting electrolyte 0.050 M phosphate buffer solution pH 7.40.

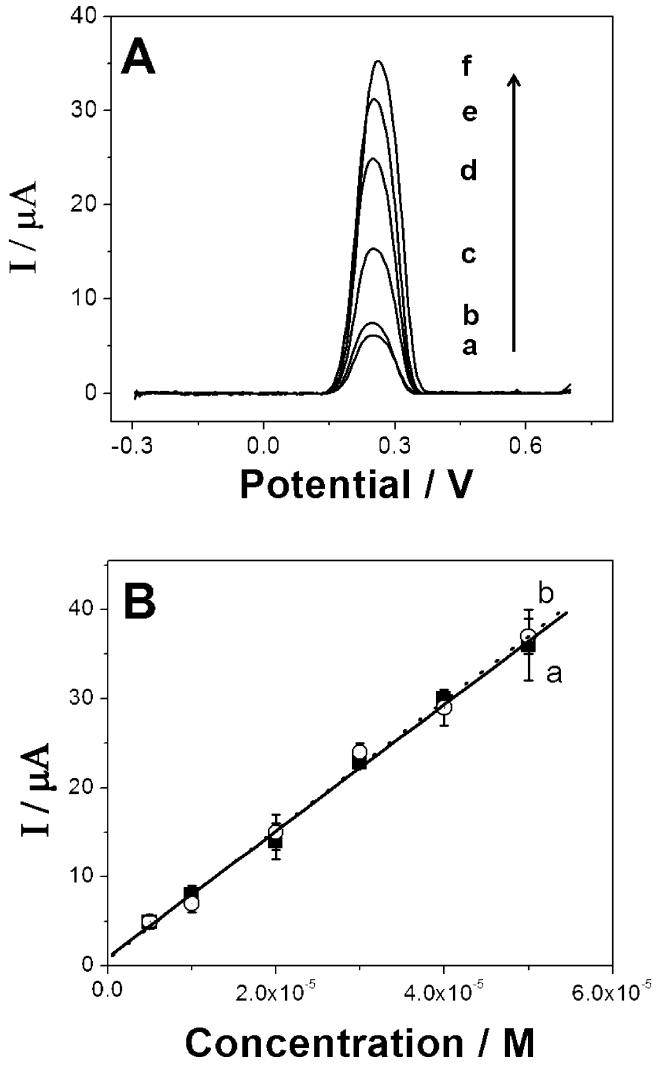

Fig. 4. A) DPVs obtained in a 0.050 M phosphate buffer solution pH 7.40 at GCE/MWCNT-PEI after accumulation of UA: 0.5×10^{-5} M (a), 1.0×10^{-5} M (b), 2.0×10^{-5} M (c), 3.0×10^{-5} M (d), 4.0×10^{-5} M (e) and 5.0×10^{-5} M (f) in the presence of 1.00×10^{-3} M AA. B) Calibration plots for UA obtained in the absence (empty circles) and presence (full circles) of 1.0×10^{-3} M AA by DPV-AdS and medium exchange. Other conditions as in Figure 2.

Figure 4A shows DPV recordings obtained in a 0.050 M phosphate buffer solution pH 7.40 at GCE/MWCNT-PEI for different concentrations of UA from 0.5×10^{-6} M to 5.0×10^{-5} M in the presence of 1.00×10^{-3} M AA after 5 s accumulation at −0.300 V. A clear definition of the oxidation process is observed for the whole range of concentrations, even the more diluted ones. Figure 4B shows the calibration plot for UA obtained in the absence (empty circles) and presence (full circles) of 1.0×10^{-3} M AA obtained by DPV-AdS after medium exchange under the optimal conditions. There is a linear relationship between peak current and UA concentration from 0.5×10^{-6} to 5.0×10^{-5} M. The sensitivity for UA in the absence and presence of AA are $(7.2\pm0.2)\times10^{5}$ µAM^{-1} ($r^2=0.9994$) and $(7.2\pm0.3)\times10^{5}$ µAM^{-1} ($r^2=0.997$), respectively. The detection limit is 1×10^{-7} M (taken as $3.3\times\sigma/S$, where σ is the standard deviation of the blank signal and S the sensitivity), and the quantification limit is 3×10^{-7} M (taken as $10\times\sigma/S$). These results clearly demonstrate that, based on the different adsorptive behavior of UA and AA at GCE/MWCNT-PEI, it is possible the highly sensitive and selective quantification of UA in mixtures of UA and AA.

The *RSD* for the determination of 4.0×10^{-5} M UA using five different electrodes modified with the same MWCNT-PEI dispersion was 3.0%. Analogous experiments using five electrodes prepared with five different dispersions gave a *RSD* of 2.0%.

The GCE/MWCNT-PEI was used for the quantification of UA in urine without pretreatment, just with a previous dilution with 0.050 M phosphate buffer pH 7.40 (50 times). The concentration of UA in the urine sample obtained with the proposed sensor was (17.0 ± 0.2) mg mL^{-1} (average of five determinations), which is in excellent agreement with the value reported by the clinical laboratory that supplied the sample, 16.8 mg mL^{-1} (using the enzymatic method with spectrophotometric quantification). We also evaluated the percentage of recovery after the addition of 2.0×10^{-5} M UA to the diluted urine sample and the values obtained ranged between 98 and 105%, demonstrating an excellent performance.

Table 1 presents a comparison of the analytical performance of the proposed sensor with other electrochemical sensing methodologies for the quantification of UA using nanostructures. The analysis of the results summarized in the table indicates that our sensor is highly competitive, with detection limits and sensitivities even better than the most relevant sensors [9,13,18,19,25,27,32,40]. Other authors [14,15,41] have reported better detection limits, although they used more complicated protocols.

4 Conclusions

In summary, the combination of the electrocatalytic activity of MWCNTs and the positive charge of the polymer used as efficient dispersing agent of MWCNTs have allowed the highly sensitive and selective quantification of UA even in the presence of large excess of AA in a simple and fast way. The electrode was successfully used for the determination of UA in urine samples demonstrating an excellent agreement with the value informed using conventional methods. These characteristics make GCE/MWCNT-PEI an interesting analytical tool for the quantification of UA in the presence of AA, and open the doors to new challenges in the electroanalytical determination of other bioanalytes and further practical applications.

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Table 1. Comparison of the analytical performance of the proposed sensor with other electrochemical sensing methodologies for the quantification of uric acid using nanostructures.

Modified electrode	Detection	Analyte	Sample	Analytical performance	Ref.
CCE/MWCNT	CV, DPV and amperometry +0.30 V	UA. No Interference AA, DA	Urine	*LOD*: 1.4 μM, *LR*: 0.5–10.0 μM Sensitivity: 1.5 μA μM^{-1}.	[9]
Au/MWCNT	Amperometry +0.40 V	UA. No Interference of creatinine, urea, ammonia, AA	Urine	*LOD*: 0.1 μM; *LR*: up to 1.8 mM Sensitivity: 92 μA mM^{-1}	[19]
SPCE/PAA-MWCNT	DPV	AA, NE, UA	AA + NE + UA pure solution	*LOD*: 0.458 μM *LR*: 0–30 μM Sensitivity: 8.295 μA μM^{-1}	[13]
Au/MWCNT-Chit/ PAMAM/DNA	DPV	DA and UA Interferent evaluated: AA	AA + DA + UA	*LOD*: 0.07 μM *LR*: 0.5–100 μM	[15]
GCE/MWCNT-Plys	DPV-AdS with medium exchange	UA No Interference of AA	Urine	*LOD*: 2.2 μM *LR*: 10–80 μM Sensitivity: 0.5 μA μM^{-1}	[25]
GCE/MWCNT/SDS	DPV	AA, UA, DA	AA + DA + UA	*LOD*: 0.4 μM *LR*: 4–30μM Sensitivity: 1.17458 μA μM^{-1}	[18]
CPE/MWCNT/α-CD	DPV	UA and DA	DA + UA	*LOD*: 5.0 μM *LR*: 5.0–40.0μM Sensitivity: 0.325 μA μM^{-1}	[32]
AU/MWCNT/AuNP/uricase	CV	UA	Serum	*LOD*: 0.01 mM; *LR*: 0.01–0.08 mM. Sensitivity: 0.44 mA mM^{-1}.	[27]
GCE/fMWCNT/Q	CV	UA, LD, Tyra	Urine	*LOD* 0.575 μM *LR*: 1-125 μM Sensitivity: 0.078 μA μM^{-1}	[40]
GCE/La-MWCNT	CA	AA, DA, UA, NO_2^-	Urine serum	*LOD*: 0.015 μM *LR*: 0.04–810 μM	[41]
GCE/ MWCNT@PDOP@PtNP	CV, DPV	UA and DA	AA + DA + UA	*LOD*: 0.12 μM *LR*: 0.3–13 μM Sensitivity: 1.03 μA μM^{-1}	[16]
GCE/MWCNT-FeNAZ-chit	DPV	AA, DA, UA, Trp	Urine, serum	*LOD*: 0.033 μM *LR*: 0.23 nM–83.3 μM Sensitivity: 4.0 μA μM^{-1}	[14]
GCE/MWCNT-PEI	DPV-AdS with medium exchange	UA	Urine	*LOD*: 0.1 μM *LR*: 5–50 μM Sensitivity: 7.5×10^2 μA mM^{-1}	This work

Analytes: UA: uric acid; AA: ascorbic acid; DA: dopamine; NE: norepinephrine; LD: levodopa, Tyra: tyramine; Trp: tryptophan. **Electrodes:** CCE: carbon-ceramic electrode, SPCE: screen printed carbon electrode, Au: gold; GCE: Glassy carbon electrode; CPE: carbon paste electrode. **Polymers:** Plys: polylysine; PAA: polyacrylic acid; PAMAM: poly(amidoamine ; Chit: chitosan; α-CD: α-cyclodextrine; Q: quercetin. **Others:** MWCNT: multiwalled carbon nanotubes; SDS: sodium dodecyl sulfate; AuNP: gold nanoparticles; fMWCNT: functionalized MWCNT; FeNAZ: iron ion-doped natrolite zeolite; PDOP: polydopamine; PtNPs: Pt nanoparticles; La-MWCNT: Lanthanum-MWCNT nanocomposite. **Techniques:** CV: Cyclic Voltammetry; DPV: Differential Pulse Voltammetry; CA: chronoamperometry; DPV-AdS: Differential Pulse Voltammetry-Adsorptive Stripping; *LOD*: Limit of detection; *LR*: linear range.

References

[1] S. Wang, Q. Xu, G. Liu, *Electroanalysis* **2008**, *20,* 1116.
[2] D. Martínez-Pérez, M. L. Ferrer, C. Reyes-Mateo, *Anal. Biochem.* **2003**, *322*, 238
[3] J. Yu, S. Wang, L. Ge, S. Ge, *Biosens. Bioelectron.* **2011**, *26*, 3284.
[4] R. C. Matos, M. A. Augelli, C. L. Lago, L. Angnes, *Anal. Chim. Acta* **2000**, *404*, 151.
[5] Y. Tanaka, N. Naruishi, H. Fukuya, J. Sakata, K. Saito, S. Wakida, *J. Chromatogr. A* **2004**, *1051*, 193.
[6] X. Dai, X. Fang, C. Zhang, R. Xu, B. Xu, *J. Chromatogr. B* **2007**, *857*, 287.
[7] D. Iveković, M. Japec, M. Solar, N. Živković, *Int. J. Electrochem. Sci.* **2012**, *7*, 3252.
[8] S. Prakash, T. Chakrabarty, A. M. Rajesh,, V. K. Shahi, *Measurement* **2012**, *45*, 500.
[9] B. Habibi, H. Pezhhan, M. H. Pournaghi-Azar, *Microchim. Acta* **2010**, *169*, 313.
[10] Y. Yu, Z. Chen, B. Zhang, X. Li, J. Pan, *Talanta* **2013**, *112*, 31.
[11] F. Sekli-Belaidi, D. Evrard, P. Gros, *Electrochem. Commun.* **2011**, *13*, 423.
[12] F. d. A. d. S. Silva, C. Bezerra Lopes, L. Tatsuo Kubota, P. Rodrigues Lima, M. Oliveira Fonseca Goulart, *Sens. Actuators B* **2012**, *168*, 289.
[13] S.-H. Huang, H.-H. Liao, D.-H. Chen, *Biosens. Bioelectron.* **2010**, *25*, 2351
[14] M. Noroozifara, M. Khorasani-Motlagh, R. Akbari, M. Bemanadi Parizi, *Biosens. Bioelectron.* **2011**, *28*, 56
[15] X. Liu, Y. Peng, X. Qu, S. Ai, R. Han, X. Zhu, *J. Electroanal. Chem.* **2011**, *654*, 72.
[16] M. Lin, H. Huang, Y. Liu, C. Liang, S. Fei, X. Chen, C. Ni, *Nanotechnology* **2013**, *24*, 065501.
[17] Y. J. Yang, W. Li, *Biosens. Bioelectron.* **2014**, *56*, 300.
[18] J. Zhang, Z. Zhu, J. Zhu, K. Li, S. Hua, *Int. J. Electrochem. Sci.* **2014**, *9*, 1264.
[19] J. Wang, W.-D. Zhang, *J. Electroanal. Chem.* **2011**, *654*, 79.
[20] M. Ramírez-Berriozabal, L. Galicia, S. Gutiérrez-Granados, J. Sandoval, P. Herrasti, *Electroanalysis* **2008**, *20(15),* 1678.
[21] F. Y. Zhang, Z. H. Wang, Y. Z. Zhang, Z. X. Zheng, C. M. Wang, Y. L. Du, W. C. Ye, *Talanta,* **2012**, *93*, 320.
[22] P. Shi, X. Miao, H. Yao, S. Lin, B. Wei, J. Chen, X. Lin, Y. Tang, *Electrochim. Acta* **2013**, *92*, 341.
[23] L. Zhang, D. Yang, L. Wang, *Electrochim. Acta* **2013**, *111*, 9.
[24] S. Chitravathi, B. E. Kumara Swamy, G. P. Mamatha, B. S. Sherigara, *J. Electroanal. Chem.* **2012**, *667*, 66.
[25] M. C. Rodríguez, J. Sandoval, L. Galicia, S. Gutiérrez, G. A. Rivas, *Sens. Actuators B* **2008**, *134*, 559.
[26] G. Rivas, M. D. Rubianes, M. L. Pedano, N. F. Ferreyra, G. L. Luque, M. C. Rodríguez, S. A. Miscoria, *Electroanalysis* **2007**, *19(7–8)*, 823.
[27] N. Chauhan, C. S. Pundir, *Anal. Biochem.* **2011**, *413*, 97.
[28] T.-Q. Xu, Q.-L. Zhang, J.-N. Zheng, Z.-Y. Lv, J. Wei, A.-J. Wang, J.-J. Feng, *Electrochim. Acta* **2014**, *115*, 109.
[29] B. Kaura, T. Pandiyanb, B. Satpatic, R. Srivastava, *Colloid Surf. B*, **2013**, *111*, 97.
[30] L. Zhang, W.-J. Yuan, B.-Q. Hou, *J. Electroanal. Chem.* **2013**, *689*, 135.
[31] J. Yan, S. Liu, Z. Zhang, G. He, P. Zhou, H. Liang, L. Tian, X. Zhou, H. Jiang, *Colloid Surface B*, **2013**, *111*, 392.
[32] S. M. Ghoreishi, M. Behpour, M. H. M. Fard, *J. Solid State Electrochem.* **2012**, *16*, 179.
[33] M. A. Raj, S. A. John, *Anal. Chim. Acta* **2013**, *771*, 14.
[34] Q. Lian, Z. He, Q. He, A. Luo, K. Yan, D. Zhang, X. Lu, X. Zhou, *Anal. Chim. Acta* **2014**, *823*, 32.
[35] Y. Bu, W. Dai, N. Li, X. Zhao, X. Zuo, *J. Energy Chem.*, **2013**, *22*, 685.
[36] M. Mallesha, R. Manjunatha, C. Nethravathi, G. S. Suresh, M. Rajamathi, J. S. Melo, T. V. Venkatesha, *Bioelectrochemistry* **2011**, *81*, 104.
[37] A. Gutiérrez, S. Gutiérrez-Granados, G. García, L. Galicia, G. Rivas, *ECS Transactions* **2010**, *2,* 369.
[38] A. Gutiérrez, S. Gutiérrez, G. García, L. Galicia, G. Rivas, *Electroanalysis* **2011**, *23,* 1221.
[39] E. N. Primo, F. A. Gutierrez, G. L. Luque, P. R. Dalmasso, A. Gasnier, Y. Jalit, M. Moreno, M. V. Bracamonte, M. Eguílaz Rubio, M. L. Pedano, M. C. Rodríguez, N. F. Ferreyra, M. D. Rubianes, S. Bollo, G. A. Rivas, *Anal. Chim. Acta* **2013**, *805*, 19.
[40] J. B. Raoof, R. Ojani, M. Amiri-Aref, M. Baghayeri, *Sens. Actuators B* **2012**, *166–167*, 508.
[41] W. Zhang, R. Yuan, Y.-Q. Chai, Y. Zhang, S.-H. Chen, *Sens. Actuators B* **2012**, *166–167*, 601.

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