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80167-7322(18)36709-6
https://doi.org/10.1016/j.molliq.2019.02.109
MOLLIQ 10510
Journal of Molecular Liquids
21 December 2018
14 February 2019
22 February 2019

Please cite this article as: F.M. Aqlan, M.M. Alam, A.M. Asiri, et al., Fabrication of selective and sensitive Pb2+ detection by 2,2'-(-(1,2-phenylenebis(azaneylylidene))bis(methaneylylidene))diphenol by electrochemical approach for environmental remediation, Journal of Molecular Liquids, https://doi.org/10.1016/j.molliq.2019.02.109

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Fabrication of selective and sensitive Pb²⁺ detection by 2,2'-(-(1,2-

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Abstract:

The Tetradentate Schiff base was easily prepared from *o*-phenylenediamine and salicylaldehyde and used as a ligand for the detection of heavy metal ions in phosphate buffer medium by electrochemical approach. To fabricate the lead (Pb^{2+}) ion sensor, a slurry of the synthesized 2,2'-(-(1,2-phenylenebis(azaneylylidene))bis(methaneylylidene))diphenol (TDL) ligand was deposited on glassy carbon electrode (GCE) as uniform thin layer. The calibration curve of the proposed Pb^{2+} ion sensor in form of a current vs. concentration plot is found to be linear over a Pb^{2+} ion concentration range of 0.1nM~0.01mM. The sensitivity of the sensor is very high (8.84 µAµM⁻¹cm⁻²) and detection limit is as low as 98.02±0.50 pM (Limit of detection is 316.73 pM). The projected Pb^{2+} sensor with active TDL shows reliability, precise reproducibility and short response time (16.0 sec.) during sensing performances. Above all, it was showed successive applicability to detect real environmental samples. In the field sensor development, this methodology using TDL onto GCE might be ae efficient and reliable innovative technique for the detection of heavy metal ions by electrochemical methods.

Keywords: 2,2'-(-(1,2-phenylenebis(azaneylylidene))bis(methaneylylidene))diphenol; Pb²⁺ cation sensor; Glassy carbon electrode; Electrochemical method; Environmental safety

1. Introduction

The Schiff bases are nitrogenous organic compounds, and known as imines or azomethines. They are utilized in optical and electrochemical sensors, and additionally in different chromatographic strategies, to empower recognition of upgraded selectivity and affectability [1-3]. These can be shaped as bi-tri-and tetradentate ligands containing nitrogen and other electron donor (molecules). This class of compound is effortlessly orchestrated and proved effective as

sensors to detect metal ion and as well as a ligand to the synthesis of numerous organometallic complexes [4-14]. The liquid crystals with anisotropic properties and the semiconducting polymers have been synthesized from Schiff bases [15,16].

Nowadays, the overwhelming metal particles are pulled with exceptional consideration on account of the perilous poisonous impacts towards the earth and the human being [17,18]. Specially, Pb^{2+} is substantially more lethal than other metals and known as neurotoxin. The exposure of Pb^{2+} ion is responsible for a number of syndromes in human including, neurobehavioral disorders, neurological damage, cognitive dysfunction, hypertension, renal impairment and the diseases of liver and kidney. In case of infants and children, it can be delayed the mental and physical growth [19-22]. The environmental lead (Pb^{2+}) ion contamination is effective due to effluent of the household, paints, pharmaceuticals, hair colorants, colors, mechanical poisons, composts, and others [23-27].

According to the prescription of the World Health Organization (WHO), the acceptable level of Pb^{2+} ion in drinking water should be $10\mu g/L$ as maximum [28]. The most extreme worthy level of Pb^{2+} in blood is 10.0 mg/L, as per the Center for Disease Control and Prevention [29]. To ensure the healthy life, the sensitive and high selective method to detect Pb^{2+} ion in food chain including drinking water and bio-system is necessary. The existing methods such as plasma atomic emission spectroscopy [30], electrochemical techniques [31], colorimetric [32], anodic stripping voltammetry [33], atomic absorption spectroscopy (AAS) [34], biochemical [35] and surface enhanced Raman spectroscopy [36] are potentially applied to detect Pb^{2+} ion. Considering some significant issues such as cost, heavy and complicated instrumentation, time of detection and non-portability, the most of above mentioned methods except electrochemical technique are not suitable. To overcome these disadvantages of traditional methods, the researches based on

electrochemical technique have conducted applying various organometallic nanocomposites as sensing elements for reliable, sensitive and selective detection of various heady metal ions [37-41].

Up to date, a number of researches have reported to the detection of toxic Pb2+ with various sensing elements such as nanocomposites of metal oxides, organometallic nanocomposites, organic composites with NCT and polymer. It has reported that the nanocomposites of Ni-ZnO and SnO₂-Sb₂O₃ have been applied to the selective detection of Pb²⁺ by adsorption method [42,43]. The graphene oxide (GO)/carbon nanotube (CNT)/poly(O-toluidine) NCs and glutathione-functionalized gold nanoparticles/multi-walled carbon nanotubes (MWCNT) NCs have been found as highly active materials on GCE to detect Pb²⁺ ion in phosphate buffer medium as reported previously [44,45]. Besides this, the detection and quantification of Pb²⁺ ion have executed by applying the polymer and biomolecules [46-49]. Moreover, the ligands have found to be applied for selective detection of heavy metal ions such as Cu²⁺ and Hg²⁺ reported by previous authors [50,51]. Since, it has been evidently proved that the ligands are active sensing element to detect heavy metal ions. Therefore, the aim of this study is to the development of an electrochemical sensor selective to metal ion by applying synthesized organic ligand (TDL) on GCE.

In this novel research, TDL were implemented to the development of Pb^{2+} ion sensor. The synthesized organic ligand was deposited on a GCE with electrically conductive nafion binder (5% nafion suspension in ethanol). Then, the assembled electrochemical sensor was potentially applied to detect Pb^{2+} ion in phosphate buffer medium. The analytical performances of Pb^{2+} ion such as sensitivity, detection limit, linear dynamic range, response time, reproducibility and stability were investigated in details. Finally, it was tested in real environmental samples to conform its applicability.

2. Experimental section

2.1 Materials and methods

All the solvents and the reagents in the form of analytical grade were purchased from Sigma-Aldrich Company (USA) and used as received. As part of this study, the analytical grade inorganic salts including Ga(NO₃)₃, MnSO₄, SnCl₂, CrCl₃, HgCl₂, AsCl₃, Ce(NO₃)₂, Co(NO₃)₂ and PbCl₂ were used to the preparation of the heavy metal ions solutions and received from the similar Sigma-Andrich. Besides this, monosodium, disodium phosphate buffer and conductive binding agent nafion (5% nafion suspension in ethanol) were purchased as axillary reagents. ¹H and ¹³C NMR spectra was recorded in CDCl₃ solution on a Bruker Avance 600 MHz spectrometer. Infrared spectra was performed on PerkinElmer spectra 100 FTIR spectrometer. Elemental analyses were performed at the Microanalytical Unit, Cairo University, Cairo, Egypt, and the found values were within ±0.4% of the theoretical values. Melting point was determined in open capillary tube in a Stuart Scientific melting point apparatus and was uncorrected. To assemble the desire electrochemical cell, the Keithly electrometer from USA was used as the dominating equipment and the main source of constant supply of current.

2.2 Synthesis of 2,2'-(-(1,2-phenylenebis(azaneylylidene))bis(methaneylylidene))diphenol

The title compound was prepared as shown in Scheme 1 following a reported method [4]. Typically, a mixture of salicylaldehyde (2.44, 20 mmol) and *o*-phenylenediamine (1.08 g, 10 mmol) in absolute ethanol (30.0 ml) was refluxed for 2 h. The obtained orange solid product was filtered off and recrystallized from dioxane. Yield 85% (2.67 g); mp 162-164 °C; Anal. Calcd. For $C_{20}H_{16}N_2O_2$ (316.36) C, 75.93; H, 5.10; N, 8.85. Found C, 76.56; H, 4.78; N, 8.96. IR (Cm⁻¹):

2710 (OH), 1611 (C=N), 1585, 1561, 1480 (C=C), 1276 (C-O). ¹HNMR (δ, ppm): 12.97 (s,2H,=Ar-OH), 8.95 (s,2H, CH=N), 6.96-7.68 (m,12H, Ar-H).



Scheme 1. Synthesis of TDL by condensation process.

2.3 Fabrication of desire working electrode using TDL on GCE

The fabrication of GCE is very sensitive work and the efficiency of the electrochemical responses are depended on the fabricated GCE. Thus, a thick slurry of synthesized TDL in ethanol was formed and in the form of very thin uniform layer, it was deposited on the flat part of GCE. Then, the modified GCE was dried by keeping at room conditions. To enhance the legibility of modified GCE, a drop of nafion was added on it and placed inside a low temperature oven at 35°C for adequate time to dry it entirely. The desire electrochemical sensor was assembled using the

Kiethly electrometer as a source for the constant supply of current and the modified GCE with TDL and a simple Pt-wire were connected with kiethly electrometer where, fabricated GCE and Pt-wire were performed as working and counter electrode respectively. The lead chloride salt was diluted in deionized water to prepare a series of solutions based on concentration ranging as 0.1mM ~ 0.1 nM. During the analysis of the obtained I-V data from assembled electrochemical sensor, a calibration curve was plotted as current vs. concentration of Pb²⁺ ion. Considering the maximum linear segment in the resulted calibration curve, the linear dynamic range (LDR) was defined. The sensitivity of the proposed Pb²⁺ cationic sensor was estimated from the slope of calibration curve and the surface area of GCE (0.0316 cm²). Applying signal-to-noise ratio of 3, the detection limit (DL) of Pb²⁺ ion sensor was calculated. The electrochemical (I-V) analysis of Pb²⁺ ion was performed in phosphate buffer medium and 10.0 mL phosphate buffer solution was taken in measuring beaker as constant throughout the experiment. The projected sensor is simple two electrodes system.

3. Results and discussion

3.1 The detection of Pb²⁺ ion by TDL

To the development of the cationic electrochemical sensor with active TDL and GCE, the fabrication of working electrode is the major concern. For the fabrication of the desire cationic sensor, a slurry of TDL in ethanol was subjected to deposit on GCE in the form of thin uniform layer and the stability of TDL on CGE was enhanced by the addition of a drop of nafion. Since, the nafion is a conductive copolymer commercially available as 5% suspension in ethanol, the resulted working electrode shows suitable stability and enhances electrochemical (I-V) activities with high electron transfer facilities as mentioned in our previous report to detect toxic chemicals [52-57]. The numerous toxic heavy metal ions were analyzed by assembled sensor and these

experiments were executed at 0.1µM of metal ions and potential of $0 \sim +1.5$ V in phosphate buffer medium with pH value of 7.0. The Fig. 1 (a) presents the electrochemical (I-V) responses of Ga^{3+} , Mn^{2+} , Sn^{2+} , Cr^{3+} , Hg^{2+} , As^{3+} , Ce^{2+} , Co^{2+} and Pb^{2+} . From **Fig. 1**(a), it is evidently shown that Pb^{2+} ion exhibits the highest I-V response among all metal ions and based on this, the Pb²⁺ ion is identified as selective to the assembled electrochemical sensor. The assembled electrochemical Pb²⁺ ion sensor based on TDL/binder/GCE is not equally active in all the phosphate buffer medium. Therefore, the sensor was subjected to analyze of Pb²⁺ ion with 0.1µM concentration in a range of buffer medium with pH 5.7 to 8.0 illustrated in Fig. 1(b). Obviously, our assembled Pb^{2+} ion sensor is found maximum responsive at pH 7.0. Subsequently, the lead ion with concentration ranging 0.1 mM to 0.1 nM was applied to I-V analysis as demonstrated in Fig. 1(c). As it is apparent from Fig. 1(c), the electrochemical responses are distinct from lower to higher concentration of Pb²⁺ ion. Thus, a direct relation of I-V responses with concentration of Pb²⁺ ion are experimented as described in previous articles [58-61]. To calibrate the Pb²⁺ cationic sensor, the current data are isolated from Fig. 1(c) at potential +1.5V and a new graph known as calibration curve is plotted using current vs. concentration of Pb^{2+} ion relation as presented in **Fig. 1**(d). As it is perceived from Fig. 1(d), the current data are continuously distributed in a linear manner from concentration of 0.1 nM to 0.01mM and it is defined as linear dynamic range (LDR). Evidently, the resulted LDR is a wider range of concentration. The sensitivity of Pb²⁺ ion sensor is measured from the slop of calibration curve and surface area (0.0316 cm²) of GCE and an noticeable value of sensitivity (8.8386 $\mu A\mu M^{-1} cm^{-2}$) is obtained. Counting the signal to noise ratio of 3, the detection limit is perceived around 98.02±0.50 pM (Limit of detection is 316.73 pM), the result might be high satisfactory. To identify the linearity of LDR, a plot current vs. log (conc. of Pb2+

ion) is plotted as demonstrated in **Fig. 1**(d) [inset Fig. 1(e)] and the current data is found to be analogous with regression co-efficient R^2 =0.9994, provides the evidence of the linearity of LDR.



Fig. 1. Optimization of sensor analytical performances of TDL/binder/GCE sensor. (a) Selectivity test with analyte concentration 0.1 μ M, (b) the activity test based on pH variation with 0.1 μ M Pb2+ ion (c) I-V responses to Pb²⁺ ion concentration, (d) exploration of calibration curve as current vs. concentration of Pb²⁺ ion [inset log (con.) vs. current].

The sensor reproducibility performance was tested using Pb^{2+} ion solution with concentration of 0.1µM and potential (V) was applied in range of 0 ~ +1.5 V in phosphate buffer

medium of pH 7.0 as illustrated in **Fig. 2**(a). As it is demonstrated in **Fig. 2**(a), the seven runs in an identical conditions are indistinguishable and the precision of current data at potential +1.5 V is equal to 1.31% RSD (relative standard deviation). The intensity of I-V responses are not altered for reproducibility performances even the washing of electrode after each trail. Thus, the test provides the information about the reliability of this Pb²⁺ ion electrochemical sensor based TDL/binder/GCE.



Fig. 2. Evaluation of reliability performance of sensor TDL/binder/GCE. (a) Reproducibility test with Pb^{2+} ion 0.1µM concentration, (b) long time performances test of Pb^{2+} ion sensor, (c)

response time for Pb^{2+} ion $0.1\mu M$ concentration and (d) interference effect test (Concentration: $0.1\mu M Pb^{2+}$ ion, $0.1\mu M As^{3+}$ ion, $0.1\mu M Co^{2+}$ ion and $0.1\mu M Hg^{2+}$ ion)

The long term stability performance of Pb²⁺ cationic sensor with TDL/binder/GCE was tested as presented in Fig. 2(b). To investigate the stability, the reproducibility performances were repeated for elongated time around seven days at applied potential $0 \sim +1.5$ V and 0.1 μ M concentration of Pb^{2+} ion. The similar achievement is perceived as in reproducibility test in **Fig.** 2(a). Therefore it can be summarized that the projected Pb²⁺ cationic electrochemical sensor is able to perform through long period in phosphate buffer medium. The response time is another important identification of an electrochemical sensor and it provides the information about the efficiency. Thus, this test was performed at 0.1μ M concentration of Pb²⁺ ion as shown in Fig. 2(c). It is observed that the projected Pb²⁺ ion sensor based on TDL/binder/GCE shows a very efficient response time around 16.0 sec. The interference effect of projected Pb²⁺ cationic sensor was experimented during the analysis of Pb^{2+} ion with other toxic heavy metal ions such as As^{3+} . Co^{2+} and Hg^{2+} ions as illustrated in Fig. 2(d). From Fig. 2(d), it is clearly shown that the electrochemical (I-V) response of Pb^{2+} ion is not changed in present of other heavy metal ion. Therefore, it is evidently proved that the proposed Pb²⁺ ion sensor based no TDL/binder/GCE is free of interference effect due to other metal ions. To find out the validity of this research, a comparison in term of sensitivity, linear dynamic range (LDR) and detection limit (DL) is illustrated in Table 1 [55,66] and the projected Pb^{2+} ion sensor is found as reliable and satisfactory in term of sensitivity, LDR and DL.

Modified GCE	DL	LDR	Sensitivity	Ref.
GO-CNT-POT/GCE	89.0 pM	0.1 nM ~1.0 mM	$8.53 \ \mu A \mu M^{-1} cm^{-2}$	[45]
HNMBSH/GCE	24.0 pM	0.1 nM~1.0 mM	$3.9*10^{-3}\mu A\mu M^{-1}cm^{-2}$	[63]
TDL/Nafion/GCE	98.0 pM	0.1 nM~0.01 mM	$8.84 \ \mu A \mu M^{-1} cm^{-2}$	This work

Table 1. Comparison of analytical performances of Pb^{2+} cation sensors based on TDL/binder/GCE sensor prove by electrochemical method.





Scheme 1. The sensing mechanism of Pb²⁺ ion with TDL/Nafion/GCE by electrochemical method. (1) Bare-GCE, (2) TDL fabricated GCE by using 5% Nafion, and (3) Electrochemical response in presence of Pb²⁺ ions.

Finally, it can be mentioned that the proposed heavy metal, Pb²⁺ cationic electrochemical sensor is performed efficiently in term of various analytical paramters such as sensitivity, linear dynamic range, LOQ, linearity, stability, reproducibility, repeatability, and detection limit etc. Moreover, it was found as good reproducibly, long-term stability in phosphate buffer medium and able to measure real environmental samples very preciously compared to other existing methods [64-80].

3.2 Detection of real environmental samples

Above all, the proposed Pb^{2+} cationic sensor was subjected to detect Pb^{2+} ion in environmental samples for validation, which is demonstrated and presented in **Table 2**. These analyses of real environmental samples were performed using 0.01μ M concentration of Pb^{2+} ion in recovery method. There were various real samples were investigated by fabricated sensor probe in electrochemical method such as industrial waste effluents, collected seawater and mineral water collected from underground well. As it is represented in **Table 2**, the evaluated data are seemed to be satisfactory.

Sample	Added Pb ²⁺ ion	Determined Pb ²⁺ concentration ^a by TDL /GCE			Average	RSD ^c (%)
	concentration	(µM)			recovery ^b	(n=3)
	(µM)				(%)	
		R1	R2	R3		
Industrial effluent	0.01000	0.00969	0.00983	0.01023	99.17	2.83
Sea water	0.01000	0.01062	0.00998	0.01009	102.30	3.35
Well water	0.01000	0.00985	0.00984	0.00987	98.53	0.18

Table 2. Analyses of real environmental samples with TDL/binder/GCE sensor.

^aMean of three repeated determination (signal to noise ratio 3) with by TDL/GCE.

^bConcentration of Pb^{2+} ion determined/Concentration taken. (Unit: μM)

^cRelative standard deviation value indicates precision among three repeated measurements(R1,R2,R3).

4. Conclusion

The tetradental Schiff base TDL (2,2'-(-(1,2-phenylenebis(azaneylylidene)))bis (methaneylylidene))diphenol) was prepared facile way and implemented to the fabrication of electrochemical sensor for successive detection of Pb²⁺ ion in phosphate buffer The Pb²⁺ cationic sensor with TDL/binder/GCE is found as sensitive (8.8386 μ A μ M⁻¹cm⁻²), reproducible and very efficient to detect Pb²⁺ ion in real environmental samples. Besides this, it has ability to measure Pb²⁺ ion in between a wider range of concentration (0.1 nM ~ 0.01 mM) and it has very trace lower

limit of detection (98.02±0.50 pM). This study may pave the way for further developments and real implementation in a large scale with this devices based on Schiff base sensor for the detection of heavy metal ions in aqueous solutions. It is introduced an efficient route for the removal of contaminated heavy metallic ions using Schiff bases modified GCE by electrochemical method for the safety of environmental and healthcare fields.

Acknowledgements

Center of Excellence for Advanced Materials Research (CEAMR) and Chemistry Department, King Abdulaziz University, Jeddah, Saudi Arabia is highly acknowledged for financial supports and research facilities.

References:

- [1] M. Valcarcel, M.D. Laque de Castro, Elsevier, Amsterdam, 1994.
- [2] U. Spichiger-Kelle, Wiley-VCH, Weinheim, 1998.
- [3] J.F. Lawrence, R.W. Frei, Elsevier, Amsterdam, 1976.
- [4] M.E.M. Zayed, R.M. El-Shishtawy, Asian J. Chem. 25 (2013) 2719–2721.
- [5] R. Azadbakht, J. Khanabadi, Tetrahedron, 69 (2013) 3206–3211.
- [6] N. Aksuner, E. Henden, I. Yilmaz, A. Cukurovali, Sens. Actuators B, 134 (2008) 510– 515.
- [7] P. Roy, K. Dhara, M. Manassero, P. Banerjee, Inorg. Chem. Acta, 362 (2009) 2927–2932.
- [8] L. Wang, H. Li, D. Cao, Sens. Actuators B, 181 (2013) 749–755.
- [9] A. Ayman, A. Aziz, J. Lumin., 143 (2013) 663–669.

- [10] T. Anand, G. Sivaraman, D. Chellappa, J. Photochem. Photobiol. A Chem., 281 (2014) 47–52.
- [11] T. Anand, G. Sivaramana, M. Iniya, A. Siva, D. Chellappa, Anal. Chim. Acta, 876 (2015) 1–8.
- [12] K.M. Vengaian, C.D. Britto, K. Sekar, G. Sivaraman, S. Singaravadivel, RSC Adv., 6 (2016) 7668–7673.
- [13] P. Singh, M.A. Quraishi, Measurement, 86 (2016) 114-124
- [14] R.M. El-Shishtawy, H. A. Al-Ghamdi , M.M. Alam, Z. M. Al-amshany , A. M. Asiri , M.M. Rahman, Chem. Eng. J. 352 (2018) 225-231.
- [15] E. Perju, V. Cozan, D. Timpu, M. Bruma, Lquid Crystals, 44 (2017) 798–808.
- [16] F.C. Krebs, M. Jørgensen, Synthetic Met., 142 (2004) 181–185.
- [17] D. Yang, L. Wang, Z. Chen, M. Megharaj, R. Naidu, Electrochim. Acta, 132 (2014) 223–229.
- [18] Y. Wei, C. Gao, F.L. Meng, H.H. Li, L. Wang, J.H. Liu, J. Phys. Chem. C, 116 (2012) 1034–1041.
- [19] L. Patrick, Altern. Med. Rev., 11 (2006) 2–23.
- [20] R.A. Goyer, Environ. Health Perspect., 100 (1993) 177-187.
- [21] T. Snoeijs, T. Dauwe, R. Pinxten, V.M. Darras, L. Arckens, M. Eens, Environmental Pollution, 134 (2005) 123–132.
- [22] Y. Finkelstein, M.E. Markowitz, J.F. Rosen, Brain Res. Rev., 27 1998 168–176.
- [23] P. Karrari, O. Mehrpour, M. Abdollahi, Daru, 20 (2012) 2.
- [24] R. Afshari, A. Emadzadeh, Drug Chem. Toxicol., 33 (2009) 48–49.

- [25] N. Manay, A.Z. Cousillas, C. Alvarez, T. Heller, Rev. Environ. Contam. Toxicol., 195 (2008) 93–115.
- [26] B.P. Zietz, J. Laß, R. Suchenwirth, H. Dunkelberg, Environ. Health Perspect., 118 (2010) A154–A155.
- [27] D.W. Dunbabin, G.A. Tallis, P.Y. Popplewell, R.A. Lee, J. Aust., 157 (1992) 835–836.
- [28] C. Baird, M. Cann, Environmental Chemistry, 3rd ed. W.H. Freeman and Company. New York, USA. (2005) 530–541 (ISBN 13:9780716748779).
- [29] S.G. Gilbert, B. Weiss, Neurotoxicology, 27 (2006) 693–701.
- [30] C.R.T. Tarley, F.N. Andrade, F.O. Midori, M. Z.Corazza, L.F. Mendes, M.G. Segatelli, Anal. Chim. Acta., 703 (2011)145–151.
- [31] X. Yang, J. Xu, X. Tang, H. Liu, D. Tian, Chem. Commun., 46 (2010) 3107–3109.
- [32] Z. Jian, Q.Y. Yun, J.J. Li, J.W. Zhao, RSC Adv., 6 (2016) 25611–25619.
- [33] S.M. Rosolina, J.Q. Chambers, C.W. Lee, Z.L. Xue, Anal. Chim. Acta., 893 (2015) 25–33.
- [34] M.P. Ochsenkuh, K.M. Ochsenkuhn, J. Anal. Chem., 369 (2001) 629–632.
- [35] J. Li, Y. Lu, J. Am. Chem. Soc., 122 (2000) 10466.
- [36] X. Shi, W. Gu, C. Zhang, L. Zhao, L. Li, W. Peng, Y. Xian, Chem. Eur. J., 22 (2016)1 10.
- [37] A.A.P. Khan, A. Khan, M.A. Alam, M. Oves, A.M. Asiri, M. M. Rahman, Inamuddin, Environ. Sci. Pollut. Res., 25 (2018) 27899–27911.
- [38] M.M. Rahman, N.A. Alenazi, M.A. Hussein, M.M. Alam, K.A. Alamry, A.M. Asiri. Adv Polym. Technol. 37 (2018) 3689-3700.

- [39] N.A. Alenazi, M.M. Alam, M.A. Hussein, K.A. Alamry, A.M. Asiri, M.M. Rahman, Polym. Bull. (2018) 1-22.
- [40] A.A.P. Khan, A. Khan, M.M. Alam, A.M. Asiri, J. Uddin, M.M. Rahman, J. Mol. Liq., 279 (2019) 392–399.
- [41] M.A. Hussein, M.M. Alam, N.A. Alenazi1, K.A. Alamry, A.M. Asiri1, M.M. Rahman. J. Polym. Res., 25 (2018) 262.
- [42] M.M. Rahman, S.B. Khan, H.M. Marwani, A.M. Asiri, K.A. Alamry, M.A. Rub, A. Khan, A.A.P. Khan, A.H. Qusti, J. Ind. Eng. Chem., 20 (2014) 1071-1078.
- [43] M.M. Rahman, S.B. Khan, H.M. Marwani, A.M. Asiri, Microchim. Acta, 182 (2015) 579.
- [44] M. Jiang, H.R. Chen, S.S. Li, R. Liang, J.H. Liu, Y. Yang, Y.J. Wu, M. Yang, X.J. Huang, Environ. Sci.: Nano, 5 (2018) 2761-2771.
- [45] A.A.P. Khan, A. Khan, M.M. Rahman, A.M. Asiri, M. Oves, Int. J. Biol. Macromol., 89 (2016) 198-205.
- [46] I. Ahmad, M.N. Arshad, M.M. Rahman, A.M. Asiri, T.A. Sheikh, F.M. Aqlan, Inorg. Chim. Acta, 467 (2017) 297–306.
- [47] D. Yang, X. Liu, Y. Zhou, L. Luo, J. Zhang, A. Huang, Q. Mao, X. Chen, L. Tang, Anal. Methods, 9 (2017) 1976-1990.
- [48] H.Y. Peng, W. Wang, F. Gao, S. Lin, L.Y. Liu, X.Q. Pu, Z. Liu, X.J. Ju, R. Xie, L.Y. Chu, J. Mater. Chem. C, 6 (2018) 11356-11367.
- [49] H.N. Kim, W.X. Ren, J.S. Kim, J. Yoon, Chem. Soc. Rev., 41 (2012) 3210-3244.
- [50] Y. Du, R. Liu, B. Liu, S. Wang, M.Y. Han, Z. Zhang, Anal. Chem., 85 (2013) 3160-3165.
- [51] M.R. Awual, M. Ismael, T. Yaita, S.A. El-Safty, H. Shiwaku, Y. Okamoto, S. Suzuki, Chem. Eng. J., 222 (2013) 67-76.

- [52] M.M. Rahman, M.M. Alam, A.M. Asiri, M.A. Islam, RSC Adv., 7 (2017) 22627–22639.
- [53] M.M. Rahm, M.M. Alam, A.M. Asiri, M.R. Awual, New J. Chem., 41 (2017) 9159-9169.
- [54] M.M. Rahman, M.M. Alam, A.M. Asiri, New J. Chem., 41 (2017) 9938-9946.
- [55] M.M. Rahman, M.M. Alam, A.M. Asiri, M.A. Islam, Talanta, 170 (2017) 215–223.
- [56] M.M. Rahman, M.M. Alam, A.M. Asiri, RSCAdv., 8 (2018) 960–970.
- [57] M.M. Rahman, M.M. Alam, A.M. Asiri, J. Ind. Eng. Chem., 62 (2018) 392-400.
- [58] M.M. Alam, A.M. Asiri, M.T. Uddin, M.A. Islam, M.M. Rahman, RSC Adv., 8 (2018) 12562–12572.
- [59] M.M. Rahman, M.M. Alam, A.M. Asiri, M.A. Islam, Talanta, 176 (2018) 17-25.
- [60] M.M. Rahman, M.M. Alam, M.M. Hussain, A.M. Asiri, M.E.M. Zayed, Environmental Nanotechnology, Monitoring & Management, 10 (2018) 1-9.
- [61] M.M. Rahman, M.M. Alam, A.M.Asiri, J. Ind. Eng. Chem., 65 (2018) 300-308.
- [62] A. Umar, M.M. Rahman, S.H. Kim, Y.B. Hahn, Chem. Comm. 2 (2008) 166-168.
- [63] M.M. Rahman, A.M. Asiri, RSC Adv. 5 (2015) 63252-63263.
- [64] M.R. Awual, M.M. Hasan, G.E. Eldesoky, M.A. Khaleque, M.M. Rahman, M. Naushad, Chem. Eng. J. 290 (2016) 243–251.
- [65] M.R. Awual, N.H. Alharthi, M.M. Hasan, M.R. Karim, A. Islam, H. Znad, M.A. Hossain, M.E. Halim, M.M. Rahman, M.A. Khaleque, Chem. Eng. J. 324 (2017) 130–139.
- [66] M.R. Awual, N.H. Alharthi, Y. Okamoto, M.R. Karim, M.E. Halim, M.M. Hasan, M.M. Rahman, M.M. Islam, M.A. Khaleque, M.C. Sheikh, Chem. Eng. J. 320 (2017) 427–435.
- [67] A.A. Khan, A. Khan, M.M. Rahman, A.M. Asiri, M. Oves, Int. J. Biol. Macromol. 98 (2017) 256-267.
- [68] M.M. Hussain, M.M. Rahman, A.M. Asiri, J. Environ. Sci. 53 (2017) 27-38.

- [69] M.M. Rahman, J. Ahmed, A.M. Asiri, RSC Adv. 7 (2017) 14649-14659.
- [70] M.K. Alam, M.M. Rahman, M. Abbas, S.R. Torati, A.M. Asiri, D. Kim, C.G. Kim, J. Electroanal. Chem. 788 (2017) 66-73.
- [71] M.M. Rahman, V.G. Alfonso, F.F. Santiago, J. Bisquert, A.M. Asiri, A.A. Alshehri, H.A. Albar, Microchim. Acta 184 (2017) 2123-2129.
- [72] M.M. Hussain, M.M. Rahman, M.N. Arshad, A.M. Asiri, ACS Omega 2 (2017) 420-431.
- [73] M.N. Arshad, T.A. Sheikh, M.M. Rahman, A.M. Asiri, H.M. Marwani, M.R. Awual, J. Organomet. Chem. 827 (2017) 49-55.
- [74] M.R. Awual, M. Khraisheh, N.H. Alharthi, M. Luqman, A. Islam, M.R. Karim, M.M. Rahman, M.A. Khaleque, Chem. Eng. J. 343 (2018) 118–127.
- [75] J. Ahmed, M.M. Rahman, I.A. Siddiquey, A.M. Asir, M.A. Hasnat. Sens. Actuators B-Chem. 256 (2018) 383-392.
- [76] M.M. Rahman, J. Ahmed, Biosensors & Bioelectronics, 102 (2018) 631-636.
- [77] M.A. Subhan, P.C. Saha, M.M. Alam, A.M. Asiri, M. Al-Mamun, M.M. Rahman, J. Environ. Chem. Engineering, 6 (2018) 1396-1403.
- [78] A.A.P. Khan, A. Khan, M.M. Alam, A.M. Asiri, J. Uddin, M.M. Rahman. J. Molecular Liquid 279 (2019) 392-399.
- [79] M.R. Karim, M.M. Alam, M.O. Aijaz, A.M. Asiri, M.A. Dar, M.M. Rahman. Talanta 193 (2019) 64-69.
- [80] B.M. Abu-Zied, M.M. Alam, A.M. Asiri, W. Schwieger, M.M. Rahman. Colloids and Surfaces A: Physicochemical and Engineering Aspects 562 (2019) 161-169.

Graphical abstract



Highlights

- ► Tetradentate TDL Schiff-base was prepared by condensation method
- ► TDL/Nafion/GCE Pb²⁺ sensor probe was fabricated
- ► The highest sensitivity (~25.05 µAµM⁻¹cm⁻²) was achieved
- ► The lowest LOD (~37.88±1.89 pM) was found
- ► Environmental and healthcare safety

Chertin Manuel