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Supradecoration induced homogenous electrochemical sensing: development of Ru(II) half sandwich complex as isoniazid and rifampicin dual sensor†

Mohd Mustafa,[a] Tabee Jan,[a] Mehdi Rohullah,[b] Mubashir Hussain Masoodi,[c] Noor U. Din Reshi*[d] and Masood Ahmad Rizvi*[a]

Homogenous electrochemical sensing using unmodified electrodes remove electrode fabrication challenges and prove effective for detecting sensitive bio-analytes without chances of surface degradation. This work envisages design and optimization of a ruthenium(II) half-sandwich complex as supradecorated homogeneous electrochemical sensor for simultaneous detection of rifampicin (RIF) and isoniazid (INH) as first-line anti-tuberculosis drugs in aqueous environments. The electrochemical profile of GCE/ruthenium(II) half-sandwich complex sensor was analyzed using cyclic voltammetry, differential pulse voltammetry and electrochemical impedance spectroscopy (EIS). The results indicate significant electrochemical parameters corroborating enhanced sensing propensity of GCE/ruthenium(II) half-sandwich complex over bare GCE for simultaneous estimation of RIF and INH binary mixture. The RIF and INZ analytical figure of merit has been corroborated with their relative supra interactional propensity. Supra interactional propensity has also been predicted to be the plausible mechanism of RIF and INZ electrochemical sensing. Under optimized conditions GCE/ruthenium(II) half-sandwich complex sensor depicted INH detection limits of 1.2 μM, and RIF detection limit of 32 nM. The comparative study of RIF and INZ analytes individually depicted high sensitivity of 24.57 μA μM^{-1} cm^{-2} and 1.69 μA μM^{-1} cm^{-2} under a linear response in the range of 0.29–3.72 μM and 4.9–82.22 μM for RIF and INH respectively. The analytical figure of merit of homogenous sensor has been compared to other GCE modified electrodes for RIF and INZ analytes. A significant antibiotic contaminant recovery of RIF and INZ drugs in pharmaceutical formulations, municipal water supplies and Dal lake water under spiked as well as unspiked conditions was observed portraying real time sensing application propensity. The homogenous GCE/ruthenium(II) half-sandwich complex expresses excellent stability and reproducibility. The GCE/ruthenium(II) half-sandwich complex in the presence of potential redox active biological interfering agents confirmed selectivity towards RIF and INZ analytes.

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Introduction

Electrochemical sensing is a promising tool for real time monitoring of contaminant concentrations for safety assessment and sustainability. Increase in global use of antibiotics for commercial agriculture and for human medicine has developed these into non conventional contaminants with ecotoxicological concerns and adverse effects on human health.[1–3] Their prevalence, risk assessment evaluation and natural attenuation rate has labeled antibiotics as major persistent contaminants challenging environmental sustainability.[4–6] The cosmopolitan distribution of antibiotic residues in surface,[7] waste-waters,[8,9] sediments,[10] crops,[11] and drinking water[12,13] lead to sustained exposure developing antibiotic-resistant bacteria (ARB) and antibiotic-resistant genes (ARG).[8] World Health Organization (WHO), considers ARGs as major threats to human and animal health in the 21st century.[14,15] A comprehensive analytical monitoring of antibiotics in environmental segments especially water bodies is crucial from sustainability, antibiotic resistance and water treatment point of view.[16–18] Bacterial strains resistant to standard first-line TB treatment drugs isoniazid (INH) and rifampicin (RIF) represent practically incurable multidrug-resistant (MDR) TB. MDR-TB strains that are resistant to

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[a] *Department of Chemistry, University of Kashmir, Hazratbal, Srinagar, 190006, J&K, India. E-mail: masoodku2@gmail.com*

[b] *School of Chemistry, University of Hyderabad, Gachibowli, Hyderabad-500046, India*

[c] *Department of Pharmaceutical Sciences, University of Kashmir, Hazratbal, Srinagar, J&K, India*

[d] *Department of Chemistry, Islamic University of Science and Technology, Awantipora, 192122, J&K, India. E-mail: noor.reshi@iust.ac.in*

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