Author's Accepted Manuscript

www.elsevier.com/locate/talanta

A reagentless and reusable electrochemical aptamer-based sensor for rapid detection of ampicillin in complex samples

Zhi-gang Yu, Rebecca Y. Lai

PII: S0039-9140(17)30886-X
DOI: http://dx.doi.org/10.1016/j.talanta.2017.08.057
Reference: TAL17853

To appear in: *Talanta*

Received date: 18 July 2017
Revised date: 17 August 2017
Accepted date: 17 August 2017

Cite this article as: Zhi-gang Yu and Rebecca Y. Lai, A reagentless and reusable electrochemical aptamer-based sensor for rapid detection of ampicillin in complex samples, *Talanta,* http://dx.doi.org/10.1016/j.talanta.2017.08.057

A reagentless and reusable electrochemical aptamer-based sensor for rapid detection of ampicillin in complex samples

Zhi-gang Yu[a,b], Rebecca Y. Lai[b]*

[a]College of Chemistry and Chemical Engineering, Yangtze Normal University, Fuling, Chongqing 400715, China

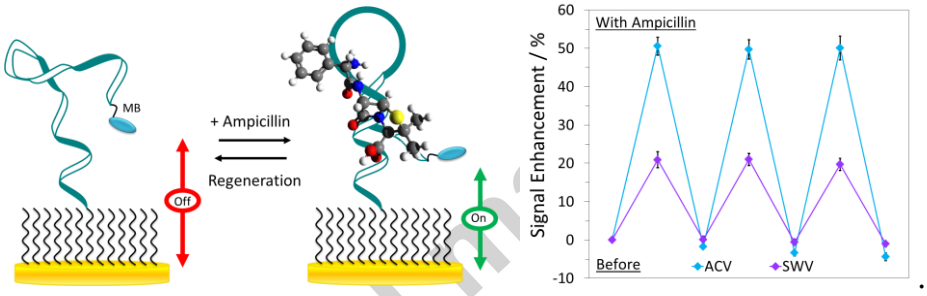
[b]Department of Chemistry, University of Nebraska-Lincoln, Lincoln, Nebraska 68588-0304, United States

*Corresponding author. Tel.: +1-402-472-5340; fax: +1-402-472-9402. E-mail address: rlai2@unl.edu

Abstract

We report the design and fabrication of a "signal-on" electrochemical aptamer-based (E-AB) sensor for detection of ampicillin. The signaling of the sensor is based on target binding-induced changes in the conformation and flexibility of the methylene blue-modified aptamer probe. The sensor's response is fast, signal saturation can be reached in ~200 sec. Since all the sensor components are surface-immobilized, it is regenerable and can be reused for at least three times. It has demonstrated good specificity and is capable of differentiating between ampicillin and structurally similar antibiotics such as amoxicillin. More importantly, it is selective enough to be employed directly in complex samples, including serum, saliva, and milk. Although both alternating current voltammetry (ACV) and square wave voltammetry (SWV) are suitable sensor characterization techniques, our results show that ACV is better suited for target analysis. Even under the optimal experimental conditions, the limit of detection of the sensor obtained in ACV (1 µM) is significantly lower than that obtained in SWV (30 µM).

Graphical Abstract:



Keywords: Ampicillin; Electrochemical aptamer-based sensor; Methylene blue; Alternating current voltammetry; Square wave voltammetry; Self-assembled monolayer

Introduction

Ampicillin (AMP) (Fig. 1A) is a penicillin-like antibiotic that is commonly used to treat certain bacterial infections such as pneumonia, bronchitis, and ear, lung, skin, and urinary tract infections (UTI) [1]. Owing to its importance as a broad-spectrum antibiotic, pharmacokinetics and pharmacodynamics studies of AMP have been performed in the past decades [2,3]. Quantification of AMP in biological fluids such as serum, urine, and saliva could help determine the optimal therapeutic concentration as well as the most effective method of administration [4,5]. Traditionally, this class of antibiotics is determined by microbiological assay [6]. While sensitive, these assays are not very specific or quantitative. To circumvent this deficiency, a wide range of analytical techniques, including high performance liquid chromatography, mass spectrometry, and surface enhanced Raman spectroscopy, have been employed to analyze AMP in complex samples [7-9]. A variety of immunoassays are also available for quantification of AMP [10-12]. Despite the high sensitivity and specificity, most are multi-step processes and cannot be used with complex samples without pretreatment or modification. There are merits in developing sensors that are capable of real time target analysis in realistically complex samples.

In the past decade, a wide selection of folding- and dynamics-based electrochemical biosensors ideal for real world applications have been developed. One of the more well-studied sensors is the electrochemical DNA (E-DNA) sensor [13-15]. These sensors are reagentless, they do not require the addition of an exogenous reagent such as an enzyme or a substrate to generate the signal. Furthermore, since all of the sensor components are surface-immobilized, they are regenerable and reusable. Most of them are sensitive, specific, and selective enough to be used directly in a wide array of biological, environmental, and food samples [13-17]. The same folding- and dynamics-based detection strategy can be applied for detection of small molecules and proteins via the use of target-specific aptamers [18-21]. To date, several electrochemical aptamer-based (E-AB) sensors have since been developed for detection of small molecules such as cocaine, and proteins such as vascular endothelial growth factor [19-21]. Aptamers capable of recognizing AMP are available, however, no E-AB sensor has yet been designed for detection of AMP in complex samples [22].