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Second order advantage obtained by spectroelectrochemistry along with novel carbon nanotube modified mesh electrode: Application for determination of acetaminophen in Novafen samples

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ABSTRACT

A matrix-augmentation multivariate curve resolution–alternating least-squares (MA-MCR–ALS) has been conducted on the spectroelectrochemical data of acetaminophen oxidation in order to quantify acetaminophen in Novafen capsule in the presence of unknown interferences. The experiments were carried out using new cheap mesh electrode, namely carbon nanotube modified mesh electrode (CNMME) as optically transparent thin layer electrode (OTTLE). For each sample, a second order spectroelectrochemical data was obtained and MA-MCR–ALS method was applied to analyze these data. Unlike full trilinear models such as PARAFAC, MCR–ALS is flexible in applying trilinearity constraint for each component, a fact which makes it manage deviations from trilinearity of data effectively. This method was employed in both spectral and kinetic augmentation mode of data under examining different trilinear components. However, spectral augmentation was the only setting which allows MA-MCR–ALS to solve the analytical problem achieving the second order advantage. Therefore, here the results of the augmentation in this mode have been described. In order to obtain the best analytical figures of merit in the analysis, different constraints were investigated. The results indicated the accuracy of the proposed method.

1. Introduction

Acetaminophen (N-acetyl-p-aminophenol, 4-acetamidophenol, paracetamol or tylenol) is an analgesic anti-pyretic drug having actions similar to aspirin, and a suitable alternative for patients who are sensitive to aspirin [1]. Overdose of acetaminophen leads to accumulation of toxic metabolites, which may cause severe and sometimes fatal hepatotoxicity and nephrotoxicity [2–4]. The development of analytical techniques for rapid analysis of paracetamol is important for quality and medical control. Many methods have been described for determination of acetaminophen including electrochemistry [5,6], spectrophotometry [7,8], liquid chromatography (LC) [9], chemiluminescence [10,11], amperometric biosensors [12,13], and FTIR-spectrometry [14]. These proposed methods often require complicated operations of pretreatment or expensive instruments which prompt us to develop a chemometrics method for the determination of acetaminophen.

Spectroelectrochemistry (SEC), as a method allowing the simultaneous acquisition of electrochemical and optical information in an electrochemical cell, has been widely used to study the redox behavior of inorganic and organic compounds, to investigate electrode reaction mechanisms, to measure reaction kinetics, and to explore electrode surface phenomena [15]. Semi-transparent materials in the form of mini-grids or mini-meshes of conducting materials as optically transparent electrode (OTE) have found extensive applications in UV–vis and NIR spectroelectrochemistry. Metals primarily available as meshes, transparent glass coatings such as indium–tin oxide (ITO) have been employed [16–18]. Some compounds do not show electrochemical activity on these electrodes. So, in order to solve this problem, these electrodes should be modified. But chemical modification of these electrodes leads to rather significant loss in their optical transparency. Gold and platinum mesh electrodes are not easily modified and cleaned, so seeking for other possible electrodes free of these disadvantages is desirable.

Because of important properties of carbon electrodes, graphite was used for coating steel mesh surface [19–21]. Also, due to interesting properties of nanomaterials [22,23], these materials can be used for modification of carbon mesh electrode.

One of the common methods for generating second-order data (three-way array) is spectroelectrochemistry method. For analyzing three-way array, multiway methods can be used. Useful techniques in this regard are parallel factor analysis (PARAFAC), particularly when the data follow the trilinear structure [24] and multivariate curve resolution–alternating least-squares (MCR–ALS) [25,26] which can handle deviations from trilinearity very efficiently. An interesting property of these models is quantitative measurement of the analyte in the presence of unknown interferents, this property is known as the 'second–order advantage'

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[27,28]. Some examples of studies in which spectroelectrochemical information has been employed to achieve the second-order advantage have been reported in these articles [29–32]. When one of the data dimensions being pH or reaction time, trilinearity may be lost if several species are created by proton-transfer reactions or by progress of a chemical reaction because of linear dependency in the concentration caused by closure relationships. In this case, algorithms such as PARAFAC may obtain incorrect results. However, PARAFAC method can still be useful by appropriate selection of the initial parameters and constraints (e.g., non-negativity) [30]. The main advantage of the trilinearity constraint in MCR–ALS over the methods based on PARAFAC is that for each component applied independently and it is not compulsory to apply it for all the resolved profiles in the augmented mode. In order to perform MCR–ALS method, first, an augmented data matrix is constructed by assembling instrumental data matrices for different samples in the following modes: (1) in the direction of the columns; (2) in the direction of the rows; or (3) in both directions simultaneously, second, some constraints, playing the main role in resolution of data set [33–37], such as non-negativity, unimodality, selectivity and local rank conditions are applied.

The main goal of the present work is to apply the matrix augmentation MCR–ALS (MA-MCR–ALS) method for the analysis of spectroelectrochemical data. As a model, acetaminophen was quantified in Novafen capsule containing an unknown interference by proposed method. Moreover, it has shown how choosing suitable constraints leads to flourish analytical figures of merit in the analysis.

2. Theoretical background

2.1. Description of data matrix

Voltabsorptometry consists of measuring the absorbed in the UV–vis region of the spectrum by the species of interest that are simultaneously

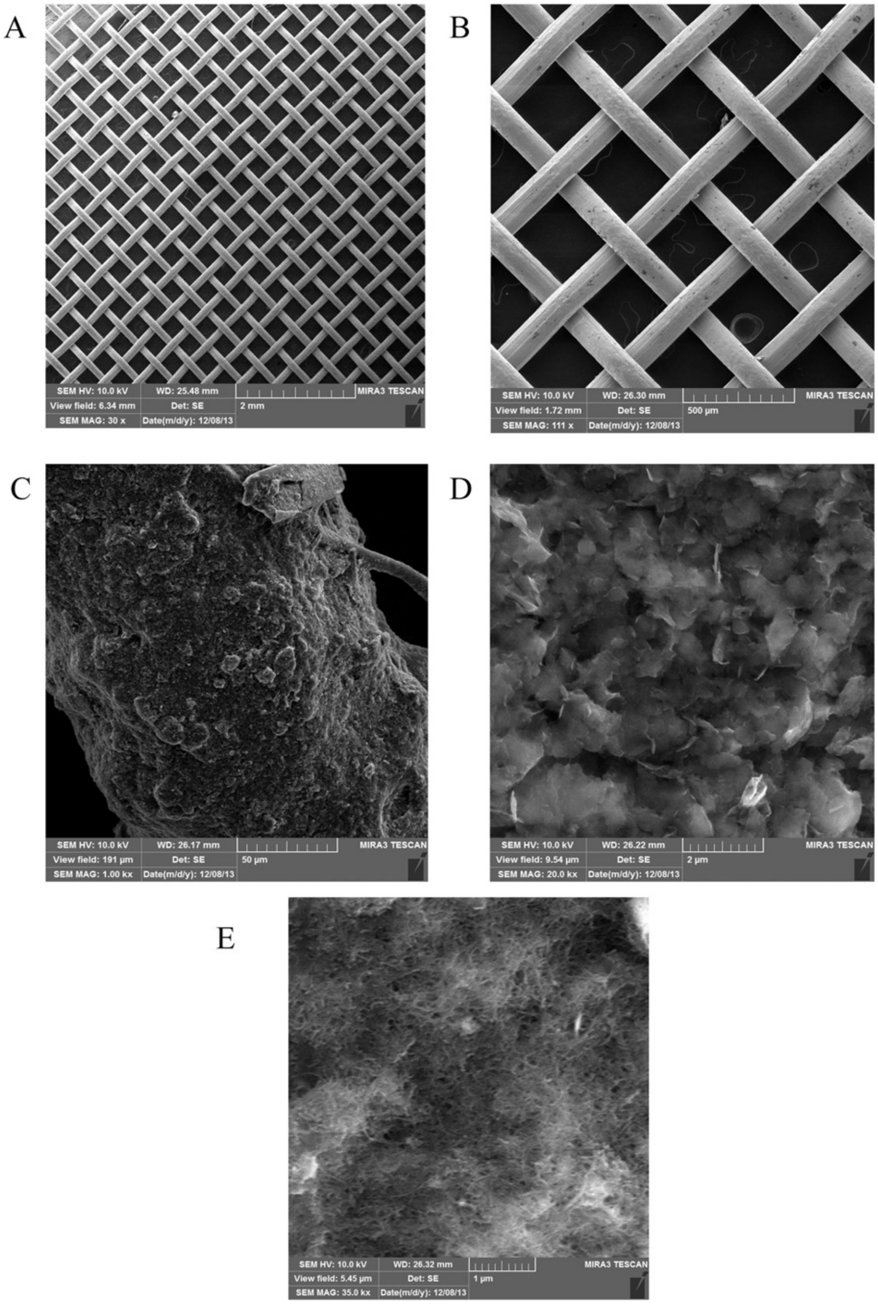

Fig. 1. SEM images of steel meshes (A, B) and modified steel meshes (C–E) with different zooms.