Talanta 134 (2015) 607–618

Contents lists available at ScienceDirect

Talanta

journal homepage: www.elsevier.com/locate/talanta

Generation of non-multilinear three-way voltammetric arrays by an electrochemically oxidized glassy carbon electrode as an efficient electronic device to achieving second-order advantage: Challenges, and tailored applications

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ARTICLE INFO

Article history:
Received 23 October 2014
Received in revised form
28 November 2014
Accepted 29 November 2014
Available online 11 December 2014

Keywords:
Second-order advantage
Paracetamol
Norepinephrine
Uric acid
Simultaneous quantification
Uncalibrated interference.

ABSTRACT

For the first time, several second-order calibration models based on artificial neural network-residual bilinearization (ANN-RBL), unfolded-partial least squares-RBL (U-PLS/RBL), multidimensional-partial least squares-RBL (N-PLS/RBL), multivariate curve resolution-alternating least squares (MCR-ALS), and parallel factor analysis 2 (PARAFAC2) were used to exploiting second-order advantage to identify which technique offers the best predictions for the simultaneous quantification of norepinephrine (NE), paracetamol (AC), and uric acid (UA) in the presence of pteroylglutamic acid (FA) as an uncalibrated interference at an electrochemically oxidized glassy carbon electrode (OGCE). Three-way differential pulse voltammetric (DPV) arrays were obtained by recording the DPV signals at different pulse heights. The recorded three-way arrays were both non-bilinear and non-trilinear therefore, the observed shifts in the recorded DPV data were corrected using correlation optimised warping (COW) algorithm. All the algorithms achieved the second-order advantage and were in principle able to overcome the problem of the presence of unexpected interference. Comparison of the performance of the applied second-order chemometric algorithms confirmed the more superiority of U-PLS/RBL to resolve complex systems. The results of applying U-PLS/RBL for the simultaneous quantification of the studied analytes in human serum samples were also encouraging.

1. Introduction

The recent developments in multi-dimensional analytical instrumentation and data collection are producing data arrays of increasing complexity, which are particularly useful for quantitative analysis in complex mixtures. It is apparent that this progress towards multi-way data offers theoretical and practical advantages from an analytical point of view [1–3]. For example, whereas zeroth-order univariate calibration cannot detect sample components producing an interfering signal, first-order calibration, which operates using a vector of data per sample, may compensate for these potential interferents, provided they are included in the calibration set, a property known as the "first-order advantage" [4]. Going one step further, second-order data lead to three-way arrays which can be uniquely decomposed, allowing relative concentrations and profiles of the individual components in the different domains to be extracted directly. In this way, analytes can

Abbreviations: ANN, artificial neural network; RBL, residual bilinearization; U-PLS, unfolded-partial least squares; N-PLS, multidimensional-partial least squares; MCR-ALS, multivariate curve resolution-alternating least squares; PARAFAC2, parallel factor analysis 2; NE, norepinephrine; AC, paracetamol; UA, uric acid; FA, pteroylglutamic acid; OGCE, oxidized glassy carbon electrode; COW, correlation optimised warping; UPW, ultrapure water; EIS, electrochemical impedance spectroscopy; ATLD, alternating trilinear decomposition; SWATLD, self-weighted ATLD; APTLD, penalized ATLD; GRAM, generalized rank annihilation method; DTLD, direct trilinear decomposition; BLLS, bilinear least-squares; EFA, evolving factor analysis; AFOM, analytical figures of merit; NAS, net analyte signal; LOD, limit of detection; SEL, selectivity; SEN, sensitivity; SVD, singular value decomposition; OLS, ordinary least squares; EJCR, elliptical joint confidence region; SIMPLISMA, simple interactive self-modeling mixture analysis.

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http://dx.doi.org/10.1016/j.talanta.2014.11.066
0039-9140/

be quantified even in the presence of unknown interferents which are not included in the calibration set. This property has been generally recognized as the "second-order advantage", a term coined in 1994 [4], although the first experimental demonstration of this advantage was reported in 1978, when perylene was determined in mixtures with anthracene, by suitable processing of fluorescence excitation-emission matrix data [5]. Second-order data for a given sample can be easily produced in a variety of ways, either in a single instrument or by resorting to instrument hyphenation.

Multi-way analysis allows direct separation of the measured signals into the underlying contributions from individual analytes. In second-order domain, three-way data generates a matrix from a single chemical sample. The processing of second-order data have attracted the attention of chemometricians in recent years for a variety of reasons: (1) they are now abundantly produced by modern analytical instruments, (2) they show peculiar mathematical characteristics which distinguish them from first order data, and (3) they provide analytical chemists with the important second-order advantage, an intrinsic property which permits analyte quantitation in the presence of unexpected sample components (i.e., components not present in the calibration set of samples) [6].

Norepinephrine (NE) is an important catecholamine neurotransmitter and is secreted by the adrenal medulla. It is released as a metabotropic neurotransmitter from nerve endings in the sympathetic nervous system and some areas of the cerebral cortex. Many diseases are related to changes of its concentration [7]. Thus the quantitative determination of NE in biological fluids for medical control and in pharmaceutical formulations for quality control analysis is important. Besides various methods such as spectrophotometry [8], capillary electrophoresis [9], gas chromatography [10] and high-performance liquid chromatography [11]; electrochemical methods have also been employed for detection of NE [12–15].

Paracetamol (AC) is the most extensively employed drug as pain reliever and fever reducer. However, overdoses of AC cause liver and kidney damage and may lead to death [16,17]. Several methods have been used for the determination of AC including spectrophotometry [18], chromatographic methods [19] and electroanalysis by modified electrodes [20–24].

Uric acid (UA or 2,6,8-trihydroxypurine) is the primary end product of purine metabolism. Physiological UA serum level is in the range of 4.1–8.8 mg (per 100 mL) and with urinary excretion typically in the range of 250–750 mg (per 100 mL). Its abnormal concentration level in the human body will lead to several diseases such as hyperuricaemia, gout, leukemia, lesch-nyhan syndrome and pneumonia [25]. Therefore, the development of a rapid, selective and simple method is very important for its determinations in routine analysis. Due to the advantages of low cost, fast response, simple instrumentation, high sensitivity, facile miniaturization, and low power requirement, numerous voltammetric methods for determination of UA have been developed [26–28].

AC administration increases brain serotonin levels [29] and serotonin is known to play a role in NE release in the brain [30]. Also, pteroylglutamic acid (FA) works primarily in the brain and nervous system and is necessary for the synthesis of NE and serotonin in the nervous system. Also, some substances like nonsteriodal anti-inflammatory drugs such as AC can inhibit FA from being absorbed or used by the body. Likewise, when taken for long periods of time, AC can also increase the need for FA [31]. On the other hand UA is naturally present in the body therefore, after drugs ingestion NE, AC, UA, and FA can be found in biological fluids. FA is one of the usual interferences in the simultaneous determination of NE, AC, and UA because the oxidation peak potential for FA was very close to those of NE, AC, and UA therefore, FA was considered as interference in the simultaneous determination of NE, AC, and UA.

In the present study, we introduced an efficient electroanalytical method based on exploiting second-order advantage from DPV data for the simultaneous quantification of AC, NE, and UA in the presence of FA as uncalibrated interference. Glassy carbon electrode (GCE) is made up of special type of carbon which fabricated by pyrolsis of polymer resin exhibits good electrical conductivity with well-defined surfaces. The chemical and electrochemical pretreatment shows significant changes in physical and electrochemical properties of GCE. In particular, electrochemical activation of GCE results in the generation of surface functional groups which could be used as capacitor electrodes. By the use of GCE we weren't able to determine low concentrations of the analytes of interest therefore, this problem prompted us to use an electrochemically oxidized GCE (OGCE) for quantifying the analytes of interest. As expected, electrochemical oxidation of GCE increases porosity of GCE and improves electron transfer kinetic between analyte and electrode. Because of the non-linear behavior of the recoded second-order data, three hybrid second-order algorithms including ANN-RBL, U-PLS/RBL, and N-PLS/RBL were used and their prediction performance was compared with MCR-ALS and PARAFAC2. The observed shifts in the recorded data were corrected by the use of COW algorithm. Finally, according to the obtained results, U-PLS/RBL was chosen as the best algorithm for the simultaneous quantification of the studied analytes in human serum samples as experimental cases.

2. Theoretical and experimental considerations

2.1. Theoretical details

2.1.1. Generating second-order DPV data

In this work, the pulse height (ΔE) in DPV was changed to obtain electrochemical second-order data. The theory behind the proposed procedure will be briefly discussed. The current signal intensity in DPV can be obtained using the following equations [32]:

$$\delta_i = \frac{n\mathrm{FAD}_O^{1/2}C_O^*}{\pi^{1/2}(\tau-\tau')^{1/2}}\left[\frac{P_A(1-\sigma^2)}{(\sigma+P_A)(1+P_A\sigma)}\right] \quad (1)$$

$$P_A = \xi\exp\left[\frac{n\mathrm{F}}{\mathrm{RT}}\left(E+\frac{\Delta E}{2}-E^{0'}\right)\right] \quad (2)$$

$$\sigma = \exp\left(\frac{n\mathrm{F}}{\mathrm{RT}}\frac{\Delta E}{2}\right) \quad (3)$$

$$\xi = \left(\frac{D_O}{D_R}\right)^{1/2} \quad (4)$$

where ΔE is the pulse height and other symbols have their conventional meanings. For a typical electrochemical reaction, a data vector can be obtained by sweeping the potential at constant ΔE and τ. Applying a different ΔE and sweeping potential at the constant τ, will produce different data vectors, i.e., sweeping the potential and applying different pulse heights (ΔEs) at a constant pulse duration in DPV produces a non-bilinear second-order data.

2.1.2. Second-order calibration algorithms

Second-order data can be processed by a variety of algorithms for analyte quantitation. Those classified as trilinear are: (1) parallel factor analysis (PARAFAC) [33], (2) different versions of alternating trilinear decomposition (ATLD) [34], such as self-weighted ATLD (SWATLD) [35] and penalized ATLD (APTLD) [36], (3) generalized