Discover the Latest Advancements in Electrochemical Detection of Ascorbic Acid: Overcoming Challenges with Precision and Insight

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Graphical abstract:

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Abstract:
In this research paper, the authors conducted a study on graphene oxide by directly applying it to the glassy carbon electrode (GCE) using cyclic voltammetric techniques. This process resulted in a modified electrode called reduced graphene oxide modified electrode (ErGO/GCE). The electrochemical behavior of ErGO/GCE towards ascorbic acid (AAB) was investigated using differential pulse anodic stripping voltammetry (DP-ASV). The differential pulse voltammetry analysis demonstrated that AAB could be detected with high selectivity and sensitivity on ErGO/GCE, exhibiting a peak at 0.312 V. The detection limits for AAB were found to be 0.36 µM, suggesting that ErGO/GCE is well-suited for detecting this analyte due to its excellent sensitivity and selectivity. Furthermore, the researchers successfully applied the proposed method to analyze AAB in pharmaceutical preparations.

Keywords: DP-ASV, ErGO/GCE modified electrode, Ascorbic acid, The updated detection of a common medication, voltammetric method

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Introduction
Vitamin C, also known as AAB–ascorbic acid, is commonly added to various foods and beverages to enhance the appearance of preserved items, owing to its antioxidant properties (Moon et al., 2020). Additionally, it serves as a stabilizer for color and flavor and inhibits browning (Kimbrough, 1976). Within the human body, AAB plays a crucial role in the biosynthesis of carnitine, collagen, and neurotransmitters (Naidu, 2003). The European Food Safety Authority has determined that the average dietary intake of AAB in healthy adults is approximately 90 mg/day (EFSA Panel on Dietetic Products, 2013).

Excessive levels of AAB can lead to undesirable gastrointestinal and renal effects, attributed to inflammatory reactions and the excretion of oxalate through urine, respectively (de Faria et al., 2020). On the one way, a deficiency in AAB can result in scurvy, a condition associated with impaired collagen synthesis (Elgailani et al., 2017). Moreover, insufficient AAB intake can negatively impact the immune system, iron absorption, cholesterol, and protein metabolism (Vissers et al., 2013).

Several analytical techniques have been employed for the determination of AAB, including coulometric (Kyaw, 1978), high-performance liquid chromatography (HPLC) with electrochemical colorimetry (Washko et al., 1989), electrochemical (Badea et al., 2015), amperometric (de Faria et al., 2020), capillary electrophoresis (Versari et al., 2004), gas
chromatography (F. O. Silva, 2005), and liquid chromatography (Oliveira & Watson, 2001). However, most of these methods suffer from drawbacks such as low sensitivity, high cost, complexity, and the desire for trained personnel. They also involve extended sample preparation and time-consuming procedures, except for the electrochemical technique (Habibi et al., 2011; López-Pastor et al., 2020). To address these issues, researchers have extensively utilized electrochemical sensors incorporating carbon nanotubes (CNTs), metal oxide nanoparticles, graphene oxide, and conducting polymers to detect AAB in various real-life samples (Dalmasso et al., 2012; K. K. H. De Silva et al., 2018; Farghali & Ahmed, 2015; Gupta et al., 2013; W. Liu et al., 2020; Marlinda et al., 2020). These sensors offer advantages such as affordability, rapid and efficient analysis, as well as strong sensitivity and selectivity.

Graphene (denoted Gr) includes a flat one-layer of sp²-carbon atoms attached and structured in a honey-house lattice network. It has gained abroad interest for its excellent features consisting of its ideal 2D structure (Novoselov et al., 2005), electronic properties (Novoselov et al., 2007), mechanical properties (Lee et al., 2008), and thermal properties (Balandin et al., 2008). Furthermore, GO and rGO, graphene oxide and reduced graphene oxide, respectively, have also been applied in changed electrodes. rGO has stronger conductivity than the GO materials and preserves the unique features discovered in the pristine Gr. Several records are connected to the usage of rGO in the electrochemistry fields (K. K. H. De Silva et al., 2018; Lourenção et al., 2009). Various techniques consisting of chemical reduction (J. Liu et al., 2017; Stankovich et al., 2007), thermal reduction (Gao et al., 2009; Lin et al., 2010), photocatalytic reduction (X. Liu et al., 2013), (Williams et al., 2008), and electrochemical reduction (Toh et al., 2014) have been applied to produce rGO materials. The electrochemical reduction process of GO is a relatively easy, saving, quick, and green pathway. This path would preserve several of the source features of pristine Gr and exploit novel functionalities of rGO materials, along with various NPs or substances (Toh et al., 2014). rGO materials generated by applying this path are called “electrochemically reduced graphene oxide” (ErGO). Until this time, the updated voltammetric investigation of AAB using an ErGO working device has not been recorded extensively.

In this study, an electrode changed with ErGO is illustrated. The obtained changed electrode was employed for the detection of AAB through the DP-ASV technique.

Material and apparatus

Materials and chemical agents

Graphite, potassium permanganate (KMnO₄), sodium acetate (NaCH₃COO), sodium citrate (Na₃C₆H₅O₇), sodium dihydrogen phosphate (NaH₂PO₄), sodiumhydrogen phosphate
(Na$_2$HPO$_4$), acetic acid (CH$_3$COOH), ascorbic acid (C$_6$H$_5$O$_6$), citric acid (C$_6$H$_8$O$_7$), boric acid (H$_3$BO$_3$), ammonia solution (NH$_3$OH, 25%), and ascorbic acid (C$_6$H$_5$O$_6$ 99%) were received from Merck (Germany). Sodium nitrate (NaNO$_3$), ethanol (C$_2$H$_5$OH), hydroperoxide (H$_2$O$_2$, 35%), and potassium hydroxide (or KOH) were received by Daejung (Korea). Phosphate-buffer solution (denoted as PBS) (pH = 6) was formed from 1 M NaH$_2$PO$_4$ and 1 M Na$_2$HPO$_4$. The Britton-Robinson buffer solutions (or B–RBS)' pH between 2.0 - 6.0 were formed by 0.50 M H$_3$BO$_3$, 0.50 M H$_3$PO$_4$, and 0.50 M CH$_3$COOH. The needed pH = 6.0 buffer solution was adjusted employing 1.0 M KOH or 1.0 M H$_3$PO$_4$ solutions.

Methods

A CPA-HH5 (denoted as Computerized Polarography Analyzer) (in Vietnam) was applied for all voltammetry tests. All tests were carried out in a cell with 03 working electrodes: a working GCE with (d = 2.80 ± 0.10 mm) employed for applying the changed electrode as the functional device, an Ag/AgCl/3 M KCl played a role as a reference device and a platinum wire worked as an auxiliary device. All tests were carried out at room conditions.

Formation of ErGO-modified GCE

GO was formed by applying the modified Hummers procedure (Toh et al., 2014), (Toh et al., 2014). Reduced graphene oxide or rGO was directly formed on the working GCE through an electrochemical reduction process. GO was employed by CV with an applied potential range between 0 V - 1.5 V in 0.20 M PBS buffer with pH = 7.0.

Results and discussion

Linear range, the limit of detection (LOD), and limit of quantitative (LOQ)

The AAB determination with the modified ErGO/GCE

The sole investigation of AAB was carried out employing the DP-ASV technique, in which the content of 01 substance was increased in the 0.20 MBRBS solution pH 3.2 (Figure 1). As could be illustrated in Figure 1 the current peak of the AAB compound with fixed content remained. It leads to the oxidation procedures being carried out solely. The peak current rises in the AAB content range between 0.2 μM - 4.4 μM. The obtained regression equations are in the following:

\[ I_{p,AAB} = (0,06 \pm 0,03) + (0.426 \pm 0.009) \cdot C_{AAB}, \quad r = 0.997, \quad p < 0.001 \]  \hspace{1cm} (1)

In the scope between 0.2 μM - 4.4 μM, the single-procedure LOD determined applying 3σ/s for AAB is 0.360 μM.
Figure 1. The DP-ASV lines on ErGO-modified GCE for (a) AAB at different contents (0.2 - 4.4 µM). Experimental conditions: potential range: -0.2 V - 0.8 V, PBS solution buffer, pH 7, scan rate: 0.1 V/s

The comparison of the obtained results and the previous reports

A summarization of the suggested technique with other voltammetric techniques for AAB investigation is reported in Table 1. It would be noticed that the LOD of AAB from the applied technique is smaller than those outcomes with employed electrodes in previous reports (Habibi et al., 2011; Yang et al., 2014). Overall, the modified ErGO/GCE is proven to be an efficient electrode for the AAB examination

Table 1. Summarization of various reports related to several electrodes for the examination of targeted analytes

<table>
<thead>
<tr>
<th>Electrode</th>
<th>Method</th>
<th>LOD (µM)</th>
<th>REF</th>
</tr>
</thead>
<tbody>
<tr>
<td>MWCNTs /GCE</td>
<td>SWV</td>
<td>0.01</td>
<td>(Gupta et al., 2013)</td>
</tr>
<tr>
<td>MWCNTs dispersed in polyhistidine/GCE</td>
<td>DPV</td>
<td>0.76</td>
<td>(Dalmasso et al., 2012)</td>
</tr>
<tr>
<td>SWCNTs/carbon-ceramic electrode</td>
<td>DPV</td>
<td>3</td>
<td>(Habibi et al., 2011)</td>
</tr>
<tr>
<td>Au-Ag bimetallic NTs in a chitosan matrix/GCE</td>
<td>Amperometry</td>
<td>2</td>
<td>(Yang et al., 2014)</td>
</tr>
<tr>
<td>MnFe$_2$O$_4$@CNT-N/GCE</td>
<td>DPV</td>
<td>1.8</td>
<td>(Fernandes et al., 2015)</td>
</tr>
<tr>
<td>Electrode</td>
<td>Method</td>
<td>LOD (µM)</td>
<td>REF</td>
</tr>
<tr>
<td>---------------</td>
<td>--------</td>
<td>----------</td>
<td>--------------------</td>
</tr>
<tr>
<td>CuO-Gr/CPE</td>
<td>DPV</td>
<td>0.011</td>
<td>(Khoshhesab, 2015)</td>
</tr>
<tr>
<td>ErGO/GCE</td>
<td>DP-ASV</td>
<td>0.36</td>
<td>This work</td>
</tr>
</tbody>
</table>

*Note: “-” not found*

**Conclusions**

The working paper illustrates that the employed electrode could be applied for the detection of AAB. The outcomes also illustrate that the changed electrode shows good electrocatalytic activity to the electrooxidation property of AAB by increasing the peak currents. Large peaks of AAB would be collected applying the DP-ASV technique, and this could help their sole investigation on the ErGO-modified GCE. The good analyzed outcome and easy formation of ErGO-modified GCE make it an interesting material for strongly sensitive properties and selective features of electrochemical sensors, 0.36 µM and 1.08 µM for LOD and LOQ, respectively. The accurate detection of ascorbic acid (AAB) through electrochemical methods and other analytical techniques presents formidable challenges in the field. These challenges encompass the intricate task of distinguishing ascorbic acid from a complex matrix of interfering substances and the imperative need for advancing sensor technology to achieve heightened sensitivity and specificity for its detection, thereby facilitating its broader application in diverse scientific and clinical contexts. In addition, the scientific value of this work should be considered because it is a valuable material or reference for the next studies about detecting AAB in general and through electrochemistry techniques specifically.

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**References**


