Study on Oxidation State Dependent Electrocatalytic Ability for I\(^{-}/I_3^{-}\) Redox Reaction of Reduced Graphene Oxides

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Abstract: The influence of oxidation states on electrocatalytic abilities for I\(^{-}\) reduction is discussed for reduced graphene oxide (rGO), which is a promising catalyst of counter electrodes for dye-sensitized solar cells (DSSCs). The oxidation states of rGO can be controlled via a photothermal reduction process by varying exposure times. The results reveal that the rGO with lower oxidation state shows smaller charge transfer resistance at the electrode/electrolyte interface and better electrocatalytic ability attributed to higher standard heterogeneous rate constant (k\(^0\)) and reasonable electrochemical surface area (A\(_s\)) for I\(^{-}\) reduction, both are quantitatively determined by a rotating disk electrode system using the Koutecký–Levich equation.

Keywords: Counter electrode · Dye-sensitized solar cells · Electrocatalytic ability · Photothermal reduction process · Reduced graphene oxide

1 Introduction

Carbonaceous materials, such as graphite, diamond, fullerene, amorphous carbon, active carbon, carbon black (CB), carbon nanotubes (CNT), and graphene (GN) were widely studied in different fields, owing to the fact that the physical and chemical properties of carbon vary greatly with its form. Some allotropic forms of carbon attract a lot of attention for electrochemical applications, such as fuel cells [1], lithium-ion batteries [2], hydrogen storages [3], sensors [4], and supercapacitors [5], owing to their outstanding electrical conductivity, chemical durability, and electrocatalytic ability. These materials possess a high potential to replace expensive materials and thereby reducing the cost of the pertinent devices and also enhancing the performance and durability of such devices.

A counter electrode (CE) in a dye-sensitized solar cell (DSSC) has the roles of collecting electrons from the photoanode and reducing the triiodide ions (I\(_3^{-}\)) to iodide (I\(^{-}\)) in the electrolyte. Typically, a conducting glass with Pt catalytic layer is used as the CE of a DSSC, due to its outstanding electrocatalytic activity for I\(_3^{-}\) reduction, excellent electronic conductivity, and good chemical durability [6]. The Pt-coated conducting glass, which can be fabricated by various methods, e.g., thermal decomposition [7], sputtering [6,8], electrochemical deposition [9], chemical deposition [10], electroless deposition [11], is most commonly used CEs in DSSCs. However, the high cost of Pt is an issue to be considered for cost-effective fabrication of DSSCs, although it shows a high catalytic activity for I\(_3^{-}\) reduction and an excellent electronic conductivity. Replacement of Pt with other cheaper materials is required for reducing the production cost of the cells, especially when the production is in mass scale. Several carbonaceous materials, e.g., activated carbon [12], CB [13], acetylene-black spheres [14], hard carbon spherule [15], graphite [16], and multiwalled carbon nanotube (MWCNT) or single-walled carbon nanotube (SWCNT) [17] have become potential materials to substitute Pt, because of their low cost, high conductivity, and good catalytic ability for the reduction of I\(_3^{-}\). Recently, Wu et al. proposed nine kinds of carbonaceous materials as the catalysts of CEs for DSSCs [18]. Murakami et al. employed CB as the catalyst for I\(_3^{-}\) reduction on a conducting glass for DSSCs and found the charge-transfer resistance for the thicker carbon layer is about one-third that of the Pt layer [13].

Ever since Novoselov and Geim have discovered the Noble Prize winning two dimensional GN in 2004 [19], it has been introduced in various electrochemical devices, such as sensors [20], lithium-ion batteries [21], and capacitors [22]. In the research of electrochemical properties of GN, some interesting results were reported. In 2008, the first electroanalytical investigation of GN was performed by Infield et al. [23], and the high electrocatalytic activity of GN towards the oxidation of I\(_3^{-}\) was demonstrated. Since then, a considerable research effort has been devoted to the electrocatalytic studies of GN and the oxidation states of GN have been investigated by different authors. Especially, the structural character of GN has been modified by chemical reduction [24], photochemical reduction [25,26], ultrasonic treatment [27], and other methods [28], which would change the physical and chemical properties of GN like the redox activity, electronic conductivity, and photocatalytic activity, etc. Therefore, it is of significant importance to study the influence of oxidation states of GN towards the electrocatalytic activities of GN.

In this study, we report the oxidation states dependency of the electrocatalytic activity of reduced graphene oxide on the counter electrodes for DSSCs. The results reveal that the rGO with lower oxidation state shows smaller charge transfer resistance at the electrode/electrolyte interface and better electrocatalytic ability attributed to higher standard heterogeneous rate constant (k\(^0\)) and reasonable electrochemical surface area (A\(_s\)) for I\(_3^{-}\) reduction, both are quantitatively determined by a rotating disk electrode system using the Koutecký–Levich equation.

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Full Paper

2.2 Preparation of GO and rGO with Various Oxidation States

GO was prepared from graphite powder using a modified version of Hummers’ method [27]. Briefly, graphite powder (2 g), NaNO₃ (1 g), and H₂SO₄ (46 mL) were mixed in an ice bath and then KMnO₄ (6 g) was added slowly. Once mixed, the solution was transferred to a water bath and stirred at 35 °C for approximately 1 h, forming a thick paste. Deionized water (80 mL) was added and then the solution was stirred for 1 h at 90 °C. Finally, more deionized water (200 mL) was added, followed by the slow addition of H₂O₂ (30%, 6 mL). The warm solution was filtered and washed sequentially with 10% HCl (200 mL) for three times and deionized water (200 mL) for one time. The filter cake was dispersed in water through mechanical agitation and then stirred overnight. The dispersion was left to settle and the supernatant (clear yellow dispersion) was subjected to dialysis for 1 month, resulting in a stock solution having a GO concentration of approximately 0.17 mg mL⁻¹. The stable dispersion was filtered through an alumina membrane (Anopore, Pore size = 0.2 μm, Whatman, UK) and left to dry for several days. The GO paper was then carefully peeled from the filter and stored under ambient conditions.

The rGO with various oxidation states were controlled by applying modified photothermal reduction with various exposure times [26]. Briefly, aqueous GO solution (2 mg mL⁻¹) was prepared and subjected to steady-state Xe lamp irradiation (250 W) with different exposure times (6–72 h). Based on our experiment results, the oxidation state of rGO cannot be further altered by increasing the exposure time longer than 72 h (data not shown). Hence, the maximum exposure time is fixed at 72 h in this work. Photothermal reduction provides a gradual transformation from GO to rGO, this implies that the oxidation state of rGO can be controlled by the exposure time. The electrocatalytic ability of rGO with various oxidation states for reducing I₃⁻ can be explored systematically.
2.3 Characterization of GO and rGO with Various Oxidation States

Aqueous solutions with 2 wt% of GO and rGO under various exposure times (6–72 h) were used for depositing the corresponding films on the ITO glasses, by drop coating technique. The structure of graphite powder and GO were investigated by transmission electron microscopy (TEM, JEM–1230, JEOL, Tokyo, Japan). UV-visible spectrophotometer (UV-vis, V-570, Jasco Inc., Maryland, USA) was used to investigate the color change from GO to rGO in aqueous solutions. The degrees of defects of the GO and rGO under various exposure times were estimated by Raman spectra, using a Dimension Raman system with 532 nm laser source (P2, Lambda Solution, Inc., USA). The contact angles of the films with GO and rGO under various exposure times on the ITO substrate were measured using a contact angle system (FTA125, First Ten Angstroms, Inc., Portsmouth, UK).

2.4 Electrochemical Measurements for GO and rGO with Various Oxidation States

CV was performed to investigate electrocatalytic abilities of the electrodes with GO and rGO under various exposure times. The CV was carried out in a three-electrode electrochemical system, by using either GO or rGO prepared under various exposure times as the working electrode, Pt foil as the counter electrode, and an Ag/Ag⁺ electrode as the reference electrode in an ACN solution containing 10.0 mM I⁻, 1.0 mM I₂, and 0.1 M LiClO₄.

A symmetric cell was used to investigate the electrocatalytic abilities for both GO and rGO, via EIS, applying a frequency range from 10 mHz to 100 kHz [28]. The cell consisted of an iodide/triiodide (I⁻/I₃⁻) electrolyte, precisely 0.1 M LiI, 0.6 M DMPII, 0.05 M I₂, and 0.5 M tBP in MPN/ACN (volume ratio = 1:1), and two identical electrodes each with an area of 1 cm², separated by a Surlyn spacer film of 60 μm thick. Impedance spectroscopic data were obtained, optimized by fitting to an equivalent circuit, using Z-View software [29].

Tafel polarization curves were obtained with the same symmetric cell by using an electrochemical potentiostat (scan rate: 100 mV s⁻¹). The exchange current density (J⁰) of an electrode, which can be obtained from the Tafel zone, is tantamount to the electrocatalytic ability of the electrode [30]. The value of J⁰ can be estimated from the extrapolated intercepts of the anodic and cathodic branches of the corresponding Tafel curves at the current density axes.

Intrinsic heterogeneous rate constant (k⁰) and effective electroactive surface area (Aₑ) for the reduction reaction of I⁻ on GO and rGO under various exposure times were quantitatively determined by a RDE system as mentioned above, by using the Koutecký-Levich equation. The RDE system was equipped with a modulated speed rotator (PINE Instrument Company) and was connected to a potentiostat (model 900B, CH Instruments, Texas, USA). GCEs (working area: 0.247 cm², Part #AFE7R9GGC, PINE Instrument Company) were deposited with thin films of GO or rGO under various exposure times by drop coating (15 μL, 30 μg catalyst); the electrodes were dried at ambient temperature. The GCEs coated with these films had served as the working electrode, Pt foil as the counter electrode, and an Ag/Ag⁺ as the reference electrode. The electrolyte used was an ACN based solution, containing 0.1 M LiClO₄ and 1.0 mM TBAI₃. CV curves were obtained by scanning the potential of the working electrode from −0.8 to 1.0 V (vs. Ag/Ag⁺) at a scan rate of 50 mV s⁻¹ (in absence of rotation). Five linear sweep voltammetric (LSV) curves (not shown) were acquired for the above electrode by controlling the rotating speed (50, 100, 200, 400, 600, and 800 rpm), at a scan rate of 1 mV s⁻¹ (scan range: 0.1 to −0.6 V vs. Ag/Ag⁺). A corresponding current, i, could be obtained from each of the LSV curves at the formal potential (E⁰). Thus, plots of i⁻¹ vs. a⁻¹/2 were made for the GCE coated with the films of GO or rGO under various exposure times.

3 Results and Discussion

3.1 Morphologies and Characteristics of As-Prepared GO

Transmission electron microscopy (TEM) images of the graphite powder and as-prepared GO are shown in Supporting Information Figure S1a and b, respectively. Graphite powder shows a dense-planar structure with several stacked layers, and GO possesses a planar sheet structure, like ultra-thin paper, with several monolayers stacked together. Figure S1c presents the Raman spectra of graphite powder and as-prepared GO, containing D band (1355 cm⁻¹, ring breathing mode from sp² carbon rings, A₁g mode) and G band (1579 cm⁻¹, planar configuration sp² bonded carbon with bond-stretching motion, E₂g mode). Raman spectra show higher ratio of D peak intensity to G peak intensity (I_D/I_G) for the as-prepared GO than that for graphite powder, implying the successful exfoliation of graphite powder to form GO [31].

3.2 Oxidation States of GO and rGO Under Various Exposure Times of Photothermal Reduction: Raman Spectra and Contact Angle Analysis

GO was further reduced to rGO by a photothermal reduction process [26]. Figure 1 displays the normalized absorbance spectra of GO and rGO obtained after a photothermal reduction process for 24 h. Supporting Information Figure S2 shows clearly that the color of GO suspensions changes from brown to black and the absorbance increases as well after undergoing a photothermal reduction process. Moreover, the normalized absorbance spectra of rGO with other exposure times of photothermal reduction process are also shown in Supporting Information Figure S3, suggesting no change of rGO absorb-
These results confirm that GO was successfully reduced by the photothermal reduction process. Supporting Information Figure S4 shows the photographs of GO and rGO obtained by being subjected to the photothermal reduction process with various exposure times (6 to 72 h). The color gradually changes from the original light brown to black with increasing exposure time. It can be inferred that the oxidation state of rGO is a function of the exposure time in the photothermal reduction process. In order to identify the oxidation state of rGO, Raman spectra was used to observe the degree of graphitization of rGO with various exposure times, as shown in Figure 2a. The intensity ratio of $I_D/I_G$ can serve as an approximate index for evaluating the graphitic quality of carbon materials. The $I_D/I_G$ value shows the decreasing trend as a function of the exposure time during the photothermal reduction process, as shown in Figure 2b. Larger value of $I_D/I_G$ is attributed to the abundant oxygen-based functional groups attached on the surface of rGO, accompanying the decoration of heteroatoms and thus producing amorphous carbon. With the increase in the exposure time of the photothermal reduction process, oxygen functionalities gradually diminished from the surface of rGO, resulting in a lower level of oxidation state, and further leading to the decreased value of $I_D/I_G$. Moreover, the contact angle of water on the films of GO and rGO with various exposure times of the photothermal reduction process were obtained and shown in Figure 3. The water contact angle of the GO film is 22.4°, implying the good hydrophilic property for GO owing to its abundant oxygen-based functional groups on the surface. Also, the contact angle shows the increasing trend as a function of the exposure time during the photothermal reduction process, indicating that the removal of the oxygen-containing groups on the surface of rGO can be achieved by the photothermal reduction process, as confirmed earlier by Raman spectra as well.

Accordingly, it can be inferred that the oxidation state of rGO is tunable by controlling the exposure time of the photothermal reduction process. Therefore, the electrocatalytic ability of rGO with various oxidation states for reducing $I_3^-/C_0$ can be explored systematically using this method.

### 3.3 Cyclic Voltammetry Analysis of the Electrocatalytic Ability for $I_3^-/C_0$ Reduction at the GO and rGO Electrodes Under Various Exposure Times

CV was performed to understand the reaction kinetics and electrocatalytic abilities of the GO and rGO electrodes with various exposure times. Figure 4 shows the CVs of the electrodes containing the GO and rGO with various exposure times, and the corresponding parameters are listed in Table 1. In a DSSC, the photoexcited electrons from the dye are injected into the TiO$_2$ conduction band. The oxidized dye is then reduced by the $I^-_3$ in the electrolyte and the resulting $I_3^-$ is reduced at the CE. The redox reactions at the photoanode and the CE are shown in Equation 1 and 2, respectively:
It can be seen in Table 1 that the peak current densities ($J_p$) increase with increasing exposure time of the photothermal reduction process; this increase in the $J_p$ is tantamount to the increase in the electrocatalytic ability of the electrodes for reducing $I_3^-$. The result of CV measurement indicates that diminishing the oxidation state of rGO, namely, removing oxygen-based functional groups, can efficiently facilitate the kinetics of $I^-/I_3^-$ redox reaction, and thus inducing higher $J_p$ value. The enhanced electrocatalytic activity of rGO electrodes with longer exposure time is certainly associated with the decreasing oxidation extent of GO and increasing active site for reducing $I_3^-$. This result indicates that rGO can be successfully prepared by the photothermal reduction and further used as the catalyst for catalyzing the $I^-/I_3^-$ reduction. Moreover, the value of $J_p$ for the electrode with rGO-72hr (4.70 mA cm$^{-2}$) is much higher than that of the Pt-coated electrode (2.55 mA cm$^{-2}$, as shown in Supporting Information Figure S5a). This suggests that rGO is a potential catalyst for replacing the conventional Pt catalyst in DSSCs.

$$3I^- \rightarrow I_3^- + 2e^- \quad (1)$$

$$I_3^- + 2e^- \rightarrow 3I^- \quad (2)$$

![Fig. 3](image3.png) Contact angle images and the trend of the contact angle value for GO and rGO under various exposure times (6 to 72 h) for the photothermal reduction process.

![Fig. 4](image4.png) Cyclic voltammograms of the electrodes with GO and rGO under various exposure times (6 to 72 h) for the photothermal reduction process, recorded in an electrolyte containing 1.00 mM LiI, 1.00 mM $I_2$, and 0.1 M LiClO$_4$ in ACN, at a scan rate of 100 mV s$^{-1}$.

<table>
<thead>
<tr>
<th>Samples</th>
<th>$J_p$ (mA cm$^{-2}$)</th>
<th>$R_s$ ($\Omega$ cm$^2$)</th>
<th>$R_c$ ($\Omega$ cm$^2$)</th>
<th>$k'$ (cm s$^{-1}$)</th>
<th>$A_e$ (cm$^2$ g$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>GO</td>
<td>0.49</td>
<td>46.5</td>
<td>272.6</td>
<td>0.38 x 10$^{-3}$</td>
<td>598</td>
</tr>
<tr>
<td>rGO-6hr</td>
<td>0.74</td>
<td>22.0</td>
<td>51.4</td>
<td>0.53 x 10$^{-3}$</td>
<td>1483</td>
</tr>
<tr>
<td>rGO-12hr</td>
<td>1.64</td>
<td>16.0</td>
<td>12.1</td>
<td>0.62 x 10$^{-3}$</td>
<td>1467</td>
</tr>
<tr>
<td>rGO-24hr</td>
<td>3.19</td>
<td>16.5</td>
<td>7.8</td>
<td>0.87 x 10$^{-3}$</td>
<td>1576</td>
</tr>
<tr>
<td>rGO-48hr</td>
<td>4.27</td>
<td>12.4</td>
<td>9.3</td>
<td>1.08 x 10$^{-3}$</td>
<td>1490</td>
</tr>
<tr>
<td>rGO-72hr</td>
<td>4.70</td>
<td>11.5</td>
<td>4.0</td>
<td>2.83 x 10$^{-3}$</td>
<td>1372</td>
</tr>
</tbody>
</table>

[a] CV measurement; [b] EIS analysis; [c] RDE analysis.
Also, the result shows that the rGO exhibits a promising potential for replacing conventional Pt CEs in DSSCs. In order to further investigate the resistance of the substrate and charge transfer behaviors of the GO and rGO electrodes with various exposure times of the photothermal reduction process, electrochemical impedance spectra (EIS) and Tafel polarization analysis were performed with symmetric cells.

3.4 Electrochemical Impedance Spectra of the GO and rGO Electrodes Under Various Exposure Times

The charge transfer resistance (\( R_{\text{ct}} \)) for the reduction of \( \text{I}_3^- \) at the electrode/electrolyte interface was investigated by EIS, using a symmetric cell composed of two identical electrodes. Figure 5 shows Nyquist plots of the electrodes with GO and rGO with various exposure times of the photothermal reduction process; the equivalent circuit is shown in the figure as an inset. A Nyquist plot of a symmetric cell is associated with three parts of the impedance. The values of ohmic series resistance (\( R_s \)) of the substrate and its catalytic layer, charge transfer resistance at the interface of the electrode with the electrolyte (\( R_{\text{ct}} \)) and Warburg resistance (\( Z_W \)) in the electrolyte were evaluated by fitting the impedance spectra in the equivalent circuit, and are given in Table 1. The Nyquist plot of a symmetric cell is associated with three parts of the impedance. The result reveals that the \( R_s \) value dramatically reduced from 46.5 to 11.5 cm\(^2\) for GO to rGO with the photothermal reduction after 72 h. It is therefore concluded that GO is gradually reduced to rGO with increasing exposure time of the photothermal reduction process, and the electric conductivity is incrementally enhanced as well. The result also indicates that the enhancement on the conductivity of rGO decreases after 12 hr, implying that most of GO would be reduced by the photothermal reduction process after half day. The value of \( R_{\text{ct}} \) is in general consistent with the electrocatalytic ability of the corresponding CEs, i.e., the value of \( R_{\text{ct}} \) would be higher with smaller value of \( J_p \). The trend of \( R_{\text{ct}} \) values is in good agreement with the values of \( J_p \) obtained from the corresponding CV measurements. The lowest value of \( R_{\text{ct}} \) was observed for the electrode with rGO-72hr due to the high electrocatalytic activity for reducing \( \text{I}_3^- \). It is worth mentioning that the values of \( R_s \) and \( R_{\text{ct}} \) for the electrode with rGO-72hr are close to that of the Pt-coated electrode (\( R_s = 7.23 \text{ cm}^2 \) and \( R_{\text{ct}} = 4.21 \text{ cm}^2 \)), implying that rGO can be successfully prepared by the photothermal reduction process and further used as the low-cost catalyst of CEs for DSSCs.

3.5 Tafel Polarization Plots for the GO and rGO Electrodes Under Various Exposure Times

Tafel polarization analysis was performed in order to further confirm the electrocatalytic property of the GO and rGO electrodes with various exposure times of the photothermal reduction process. Figure 6 shows plots of the logarithmic current density (log \( J \)) versus the potential (V), i.e. Tafel curves, obtained with the same symmetric cell measured in EIS. Theoretically, there are three zones for each of the Tafel curves; the curve at low potentials (\(|V| < 120 \text{ mV}\)) represents the polarization zone, the one at the middle potentials (with a sharp slope) represents the Tafel zone, and the other at the high potentials (horizontal part) represents the diffusion zone. The exchange current density (\( J_0 \)) of an electrode obtained from the Tafel zone is tantamount to the electrocatalytic ability of the electrode [30b].

The value of \( J_0 \) can be estimated from the extrapolated intercepts of the anodic and cathodic branches for the corresponding Tafel curves at the current density axes. The slopes of the cathodic and anodic branches in the Tafel zone are higher for the electrode with rGO-72hr than those of GO and rGO with other exposure times, indicating the highest value of \( J_0 \) for the former case ac-
The values of \( k^0 \) and \( A_e \) were extracted by using the modified Koutecký–Levich equation, which relates the current to the rotating speed; Equation 4 can be written as follows [30a]:

\[
i^{-1} = \left[ nF A_e k^0 C_0 \right]^{-1} + \left[ 0.62nF A_e D^{2/3} v^{-1/6} \omega^{1/2} C_0 \right]^{-1}
\]  

where \( i \) is the current obtained from the LSV curves at \( E^0 \), \( A_e \) is the active surface area, \( k^0 \) is the standard heterogeneous electrochemical rate constant for \( \Gamma^- \) reduction reaction, \( C_0 \) is the concentration of \( \Gamma^- \) equal to 1.0 mM, \( D \) is the diffusion coefficient of \( \Gamma^- \) equal to \( 3.62 \times 10^{-4} \text{ cm}^2 \text{ s}^{-1} \), \( v \) is the kinematic viscosity of ACN equal to \( 4.71 \times 10^{-2} \text{ cm}^2 \text{ s}^{-1} \), and \( \omega \) is the angular velocity converted from the rotating speed.

Figure 7 shows plots of \( i^{-1} \) vs. \( \omega^{-1/2} \) for the RDE with thin films of GO and rGO with various exposure times. The values of \( k^0 \) and \( A_e \) for these films were respectively calculated from the intercept and the slope of the plot using Koutecký–Levich equation after fitting the data in the plots. The parameters of \( k^0 \) and \( A_e \) are listed in Table 1. GO shows smaller \( k^0 \) and \( A_e \) than those of the rGO with various exposure times; this result can be explained by the poor electric conductivity and less active site for GO with high level oxidation states. The \( k^0 \) value of rGO gradually increases with extending the exposure time of photothermal reduction process, implying that the intrinsic electrocatalytic activity for reducing \( \Gamma^- \) can be facilitated for the rGO with lower oxidation state. However, the \( A_e \) value of rGO gradually decreases with further increases of the exposure time from 24 to 72 h. This interesting result may be caused by aggregation of rGO with less functional group on its surface, which can be ob-
served in Supporting Information Figure S4 (in the case of rGO-72 hr). The result suggests that the dominant parameter for controlling the electrocatalytic ability of rGO for reducing $I_{3}^{-}$ is $k^{0}$ instead of $A_{e}$.

4 Conclusions

Home-made rGO with various oxidation states can be controlled by applying a modified photothermal reduction under various exposure times. With increasing the exposure time of the photothermal reduction process, oxygen functionalities gradually diminished from the surface of rGO, resulting in the lower level of oxidation state and further decreased the ratio of $I_{2}/I_{3}^{-}$, obtained from Ramen spectra. The $J_{p}$ value of rGO obtained from CV increased with increasing photothermal reduction exposure time, indicating the facilitated kinetics of $I^{-}/I_{3}^{-}$ redox reaction, due to the more reduction of rGO oxidation states by more removal of oxygen-based functional groups, and therefore the more active site on rGO for reducing $I_{3}^{-}$. The trend of CV is in good agreement with that obtained from the EIS analysis and Tafel polarization curve. The $R_{0}$ values obtained from the EIS decreased from GO to rGO-72 hr, suggesting that the GO was gradually reduced to rGO with increasing photothermal reduction exposure time, and the corresponding electric conductivity was simultaneously enhanced incrementally. Moreover, higher $k^{0}$ and $A_{e}$ values measured by the RDE were obtained for rGO with various exposure times with reference to those of GO, due to the better electric conductivity and more active sites for rGO with low level oxidation states. The RDE results suggest that the dominant parameter for controlling the electrocatalytic ability of rGO for reducing $I_{3}^{-}$ is $k^{0}$ other than $A_{e}$.

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