

Research Article

Enhanced Electrocatalytic Activity of Pt Particles Supported on Reduced Graphene Oxide/Poly(3,4-ethylenedioxythiophene) RGO/PEDOT Composite towards Ethanol Oxidation

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Catalysts in fuel cells are normally platinum based because platinum exhibits high electrocatalytic activity towards ethanol oxidation in acidic medium. However, bulk Pt is expensive and rare in nature. To reduce the consumption of Pt, a support material or matrix is needed to disperse Pt on its surface as micro- or nanoparticles with potential application as anode material in direct ethanol fuel cells (DEFCs). In this study, a composite material consisting of platinum particles dispersed on reduced graphene oxide/poly(3,4-ethylenedioxythiophene) (RGO/PEDOT) support was electrochemically prepared for ethanol oxidation in sulfuric acid electrolyte. PEDOT, a conductive polymer, was potentiodynamically polymerized from the corresponding monomer, 0.10 M EDOT in 0.10 M HClO₄ electrolyte. The PEDOT-modified electrode was used as a substrate for exfoliated graphene oxide (EGO) which was prepared by electrochemical exfoliation of graphite from carbon rod of spent batteries and subsequently reduced to form RGO. The Pt/RGO/PEDOT composite gave the highest electrocatalytic activity with an anodic current density of 2688.7 mA·cm⁻² at E = 0.70 V (versus Ag/AgCl) towards ethanol oxidation compared to bare Pt electrode and other composites. Scanning electron microscopy (SEM) revealed the surface morphology of the hybrid composites while energy dispersive X-ray (EDX) confirmed the presence of all the elements for the Pt/RGO/PEDOT composite.

1. Introduction

Fuel cells are basically open thermodynamic systems that operate on the basis of electrochemical reactions and consume reactant from an external source, simply its fuel. In the recent years, proton exchange membrane fuel cells (PEMFCs) have been extensively studied and emerged as one of the potential systems, which not only provide clean energy but also offer good commercial feasibility and portability [1].

Direct alcohol fuel cell (DAFC) is a variety of PEMFCs which uses alcohols such as methanol and ethanol. Alcohols have low molecular weight and can be stored in dilute concentrations as fuel has been noted for portability and

transportation applications. Direct ethanol fuel cells (DEFCs) have spurred more and more interest in recent years due to ethanol's intrinsic advantages such as low toxicity, renewability, and its easy production in great quantity by the fermentation from sugar-containing raw materials [2].

Moreover, catalysts such as precious metals and non-precious metals are needed for the oxidation of ethanol. Platinum (Pt) has currently been regarded as the best catalyst for fuel cell electrochemical reaction. However, its high commercialization cost and scarcity could hinder its advantages. Moreover, pure Pt is not the most efficient anodic catalyst for DEFC for the reason that Pt itself is known to be rapidly poisoned on its surface by strongly adsorbed species

coming from the dissociative adsorption of ethanol [3]. To circumvent the limitation of bulk noble metal catalysts, good support materials are being explored for the dispersion of these nanoparticles such as carbon-based materials [4, 5] and conductive polymers [6, 7]. The use of support materials could not only reduce the cost of the noble metals but also improve the efficiency and poisoning tolerance of the electrocatalysts [8].

Graphene, a novel carbon allotrope, is an atomically thin sheet of hexagonally arranged carbon atoms which has attracted a lot of interest since its discovery by Geim and Novoselov in 2004 [9]. Aside from its unique structure, it also offers unique properties such as high conductivity and one of the fastest available electron transfer capabilities. Theoretical surface area of graphene is reported to be $\sim 2630 \text{ m}^2/\text{g}$, surpassing that of both carbon nanotubes and graphite [10]. It is also reported that graphene remains stable over a vast range of temperatures that is essential for reliability in many energy related applications [9]. Graphene is a nearly perfect two-dimensional sheet that there are almost no functional groups in its plane except for the edges. As a result, the interaction between graphene and noble metal nanoparticles is not very strong. To further improve the performance of graphene as the support in fuel cells, one of the best choices is to introduce other carbon materials into graphene.

Conducting polymers, such as poly(3,4-ethylenedioxythiophene) (PEDOT), are widely investigated as support matrices because of their ability to effectively disperse metallic nanoparticles evenly avoiding metallic agglomeration, thus enhancing the stability of metal nanoparticles for ethanol oxidation relevant to direct ethanol fuel cell applications [6]. We have reported the use of PEDOT as host matrix for the dispersion of Pt microparticles [11, 12]. These studies demonstrate better dispersion of the Pt particles on the electrochemically prepared PEDOT support matrix giving enhanced electrocatalytic activities towards ethanol oxidation compared to the bare Pt electrodes.

Combining the advantages of PEDOT and graphene, several studies have investigated the use of PEDOT/graphene composite as support matrix for the dispersion of Pt [13] and Pd [8] nanoparticles for ethanol oxidation. In both studies, graphene oxide was synthesized through the well-known Hummer's method [14] which requires strong oxidizing agents to oxidize graphite to graphene that could be tedious and not environment friendly. Recently, electrochemical exfoliation [15] has received a great deal of interest because of its fast and straightforward approach and accessibility in most laboratories for the synthesis of graphene oxide.

In this paper, we present the electrochemical preparation and characterization of Pt/RGO/PEDOT hybrid composite with enhanced electrocatalytic behavior while reducing its cost of preparation. Electrochemical methods were used to fabricate the composites. Morphological and elemental analyses of the composites were investigated using scanning electron microscopy (SEM) and energy dispersive X-ray (EDX) analysis, respectively. Performance evaluation of the Pt/RGO/PEDOT and other composites towards ethanol oxidation was monitored using cyclic voltammetry.

2. Materials and Methods

2.1. Chemicals. Carbon rod, a graphite source, was extracted from used battery (Eveready 1.5 V, Indonesia). Sulfuric acid (Fluka, Germany) and perchloric acid (Fluka, Germany) were of analytical grade and diluted with ultrapure water to prepare 0.5 M and 0.1 M of the electrolytes, respectively. Dimethylformamide and 3,4-ethylenedioxythiophene (>97%, Sigma-Aldrich, Singapore) were used as received. Hexachloroplatinic (IV) acid hexahydrate (>99.99%, Sigma-Aldrich, Singapore) was dissolved in 0.5 M sulfuric acid. A solution of 0.1 M sulfuric acid was used as electrolyte for ethanol (Scharlau, Spain). All solutions were prepared using Milli-Q ultrapure water (Milli-Q SP-TOC; $\geq 18.2 \text{ M}\Omega \text{ cm}^{-1}$) and sonicated to achieve homogeneous solution.

2.2. Electrochemical Exfoliation of Graphite to Produce Graphene Oxide. Electrochemical exfoliation of graphite was conducted using a power supply in a simple two-electrode cell. Carbon rod, a source of graphite, was first extracted from used battery (1.5 V Eveready), cleaned by soaking in 0.5 M H_2SO_4 for five (5) minutes, and rinsed with ultrapure water. The cleaning procedure was repeated until no visible redox peaks from 0.0 to 1.0 V (versus Ag/AgCl) are evident in the cyclic voltammetry in 0.1 M H_2SO_4 electrolyte. The clean carbon rod (anode) and platinum wire (cathode) were connected to the positive and negative terminal of the power supply, respectively, using alligator clips and conducting wires. The electrodes, separated with a horizontal distance of about 2.5 cm and with a length of about 1.0 cm in contact with the electrolyte, were immersed in 0.5 M H_2SO_4 electrolyte solution.

The electrochemical exfoliation process [15] was carried out by applying a DC conditioning bias of +1 V for 30 minutes and then a DC exfoliating bias of +10 V for 1 minute. The resulting mixture after electrochemical exfoliation was filtered in a 200 nm ashless filter paper and washed with ultrapure water by gravity filtration. The filtrate was observed until there is no evident sign of sulfate. Then the filter paper was washed with dimethylformamide and transferred to a vial. The solution was ultrasonicated for at least 30 minutes to prevent agglomeration of graphene oxide. The dispersed graphene oxide solution was subjected to centrifugation at 3000 rpm for 30 minutes to remove unwanted large particles produced during the exfoliation process. The solution was decanted to obtain the graphene oxide and was dried at 80°C in an oven. The dried sample was then labelled "EGO."

2.3. Electrochemical Synthesis of Pt/RGO/PEDOT. Electrochemical preparation and measurements were conducted using a potentiostat (eDAQ, Australia) under control of a dedicated software in a classical three-electrode cell. Glassy carbon electrode (GCE), Ag/AgCl, and Pt rod were used as the working electrode, reference electrode, and counter electrode, respectively. In all the experiments, the GCE was polished mechanically with Al_2O_3 slurry using emery paper (grade 1200–1500) and then rinsed thoroughly with ultrapure water in an ultrasonic bath prior to use.

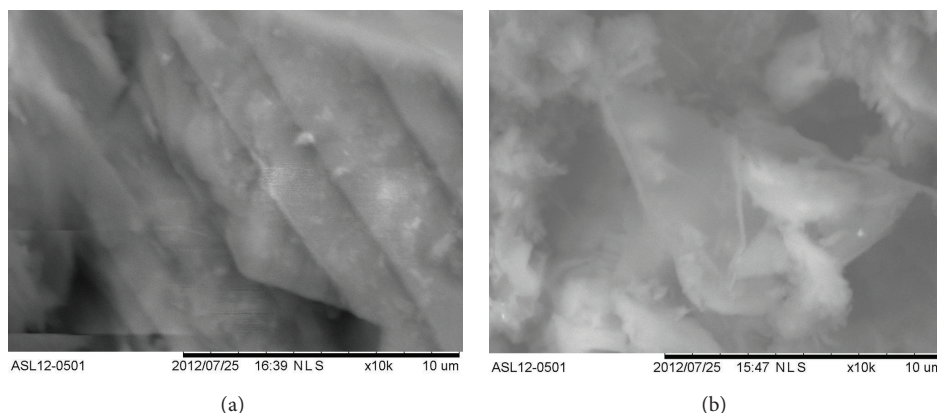


FIGURE 1: SEM image of (a) graphite and (b) electrochemically exfoliated graphene oxide (EGO) at $\times 10,000$ magnification.

The polymerization solution consisted of 10 mM of EDOT monomer and 0.1 M HClO_4 electrolyte. Potentiodynamic polymerization was carried out on a GCE using a potential range of 0.0 to 1.1 V (versus Ag/AgCl) at a scan rate of $50 \text{ mV} \cdot \text{s}^{-1}$ for 10 cycles producing a conductive PEDOT.

A volume of $20 \mu\text{L}$ of EGO colloid ($0.5 \text{ mg} \cdot \text{mL}^{-1}$) was drop-casted on the surface of PEDOT-coated GCE and was oven-dried at 90°C for 10 minutes. Electrochemical reduction of EGO in the dried EGO/PEDOT composite on GCE substrate was carried out by applying an electrode potential of -1.3 V to 0.2 V in 1.0 M sodium phosphate buffer solution ($\text{pH} = 4.12$). The reduced graphene oxide (RGO) incorporated with PEDOT was labelled as “RGO/PEDOT.”

For the electrodeposition of Pt particles, the RGO/PEDOT composite electrode was immersed in a 5.0 mM of $\text{H}_2\text{PtCl}_6 \cdot 6\text{H}_2\text{O}$ solution in 0.5 M H_2SO_4 electrolyte. Potentiodynamic deposition of Pt was carried out with a potential range of -1.1 V to 0.4 V to reduce the Pt salt to metallic particles. The Pt-based composite obtained using this sequence of procedures is labelled “Pt/RGO/PEDOT.” other combination of Pt-based composite such as “Pt/PEDOT/RGO” was obtained by depositing RGO first on GCE and then depositing the electropolymerized PEDOT.

2.4. Surface Characterization of the Support Matrix and Composites. Scanning electron microscopy (SEM) studies of graphite and electrochemically exfoliated graphene oxide were conducted using Hitachi TM-3000 Table Top Microscope (UST Research Center for the Natural and Applied Sciences, Manila). The surface morphology and elemental analyses of the composites were investigated using a JEOL JSM 5310 SEM with an accelerating voltage of 15 keV and energy dispersive X-ray (EDX) (De La Salle University, Manila) analysis, respectively.

3. Results and Discussion

Graphene oxide is obtained from electrochemical exfoliation of graphite derived from carbon rod from used battery. Electrochemical exfoliation consists of two parts. The first part is the condition DC biasing between the graphite sheets

TABLE 1: Resistance measurement of graphite and EGO.

	Graphite	EGO
Applied current	0.015 A	0.015 A
Measured voltage	1 V	0.408 V
Resistance	66.67Ω	27.2Ω

where the sulfate ions intercalate and damp. The second part is the exfoliation DC biasing, wherein the penetrated sulfate ions strip the carbon rod producing graphene oxide in the electrolyte solution [15]. The SEM image of pristine graphite (Figure 1(a)) from used battery revealed a flat surface with multilayered sheets stacked together. The width of the sheet is around $1\text{--}2 \mu\text{m}$. After electrochemical exfoliation using a conditioning bias of +1 V for 30 minutes and an exfoliation bias of +10 V for 1 minute, the ordered stacking of the sheets, characteristic of pristine graphite, has disappeared (Figure 1(b)) with a seemingly “crumpled-sheet-like” and “wrinkled” morphology. The SEM image is characterized by wrinkling of graphitic planes which could be attributed to disruptions on the edges of the graphene planes due to the introduction of the oxygen moieties [16]. Electrochemical exfoliation presents a possible low cost and faster method of producing graphene oxide with a similar surface morphology obtained using Hummer’s method [14].

Conductivity of graphene was reported to be excellent because of its unique electronic properties, which include zero mass Dirac fermion, ambipolar electric field effect, and extremely high carrier mobility [9]. In this study, resistance was measured using an improvised four-point probe technique. Resistance of the EGO was compared to the resistance of commercially available graphite powder to evaluate if the former is less resistive than the latter. As shown in Table 1, a constant current of 15 mA was applied and the voltages were measured. Based on Ohm’s law, the computed resistances were 66.67Ω and 27.2Ω for graphite and EGO, respectively. EGO is more than twice less resistive than the graphite powder. Thus, the EGO obtained is relatively conductive.

Electrochemical synthesis of Pt/RGO/PEDOT was verified through its corresponding CV profiles during each step.

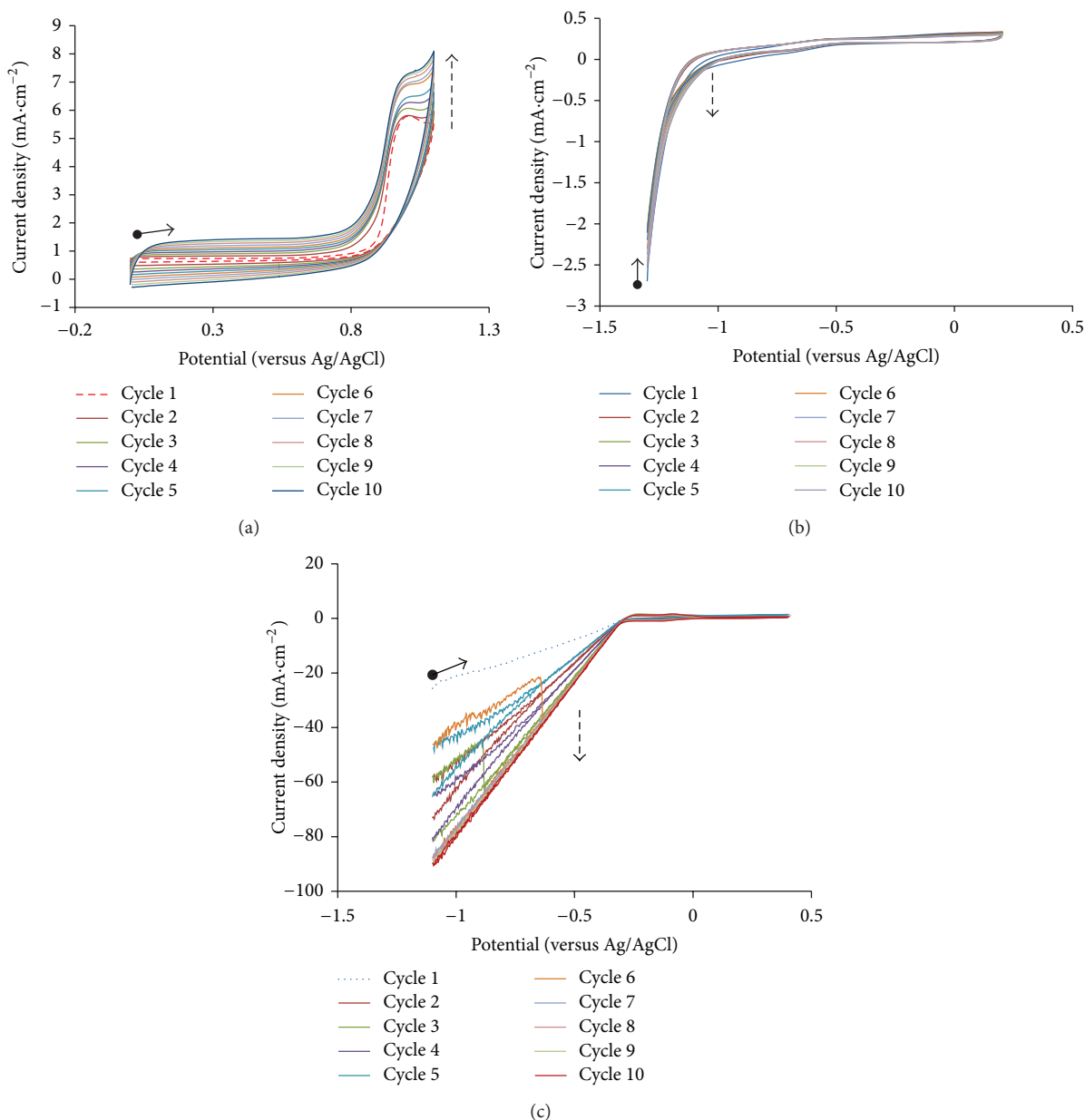


FIGURE 2: CV profiles of (a) electropolymerization of EDOT, (b) reduction of graphene oxide (GO), and (c) deposition of platinum on RGO/PEDOT composite.

A potential range from 0 V to 1.1 V was carried out during the electropolymerization process of EDOT. As presented in Figure 2(a), the growth CV of EDOT on GCE increases as the number of cycles increases as shown by a dashed upward arrow. This indicates that a conductive film is formed on the surface. To prepare RGO/PEDOT composite, a potential range of -1.3 V to 0.2 V was carried out to reduce GO producing RGO. As depicted in Figure 2(b), the cathodic current increases indicative of the reduction of RGO as shown by a dashed downward arrow. This also indicates that RGO is becoming more conductive. To prepare Pt/RGO/PEDOT, a potential of -1.1 V to 0.4 V was carried out for the deposition of Pt particles. Figure 2(c) presents the CV profile for the

deposition of platinum on the RGO/PEDOT composite with the characteristic hydrogen adsorption-desorption peaks [11, 12]. It is worthy of note that the reduction of GO to produce RGO could also take place at this negative potential.

With the unavailability of a glassy carbon electrode as a substrate for SEM, a gold wire (area = 0.0824 cm²) was used instead as a substrate for Pt/RGO/PEDOT and Pt/PEDOT/RGO to characterize their surface morphologies. As shown in Figure 3(a), a rough, globular, and porous surface indicative of PEDOT is visibly seen in the SEM micrograph with bright microparticles dispersed on the conductive polymer support (inset image). These bright microparticles are attributed to Pt particles. The PEDOT support contains

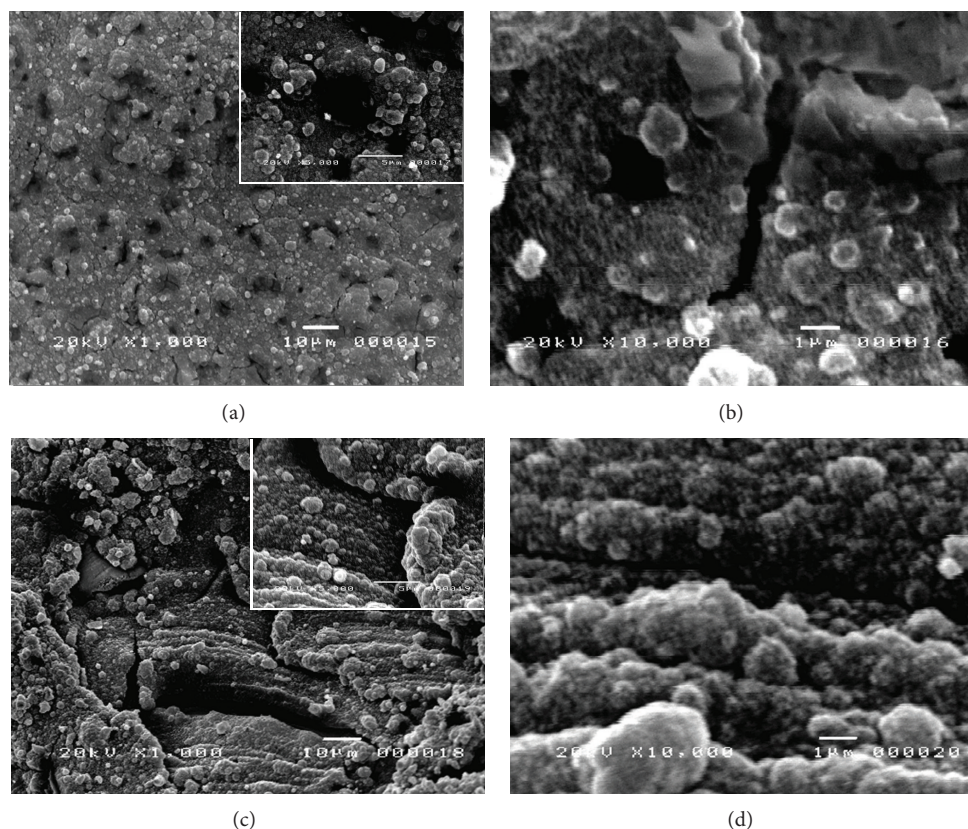


FIGURE 3: SEM images of Pt/PEDOT/RGO at (a) $\times 1,000$ and $\times 5,000$ (inset) magnification and (b) $\times 10,000$ magnification and of Pt/RGO/PEDOT at (c) $\times 1,000$ and $\times 5,000$ (inset) magnification and (d) $\times 10,000$ magnification.

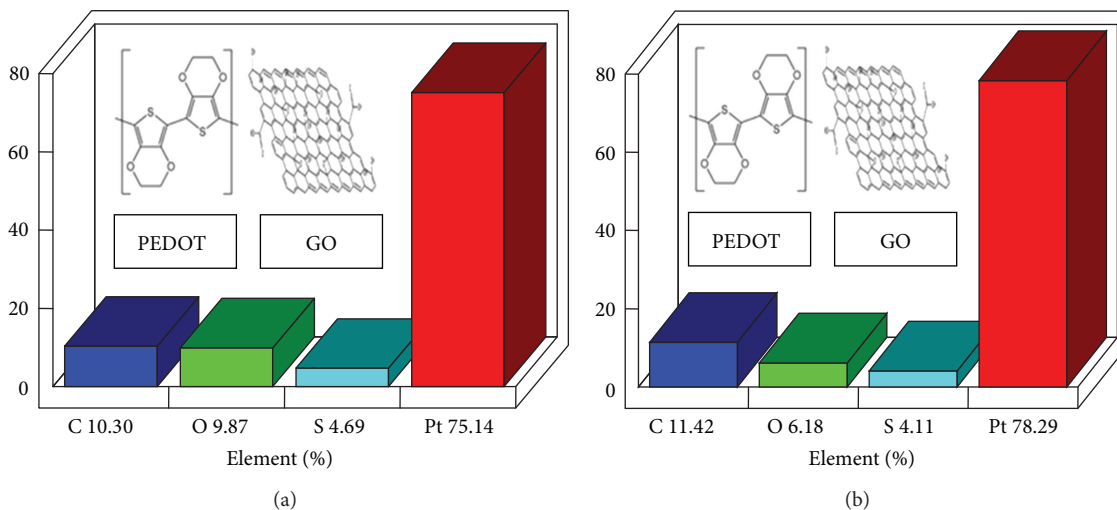


FIGURE 4: Elemental analyses of (a) Pt/PEDOT/RGO and (b) Pt/RGO/PEDOT.

microstructural globules formed by polymerization of EDOT monomers on RGO surface that helps in the dispersion of platinum metallic particles. Figure 3(b) is the high magnification SEM micrograph of Pt/PEDOT/RGO at $\times 10,000$ magnification. Traces of folds were seen along with the porous and globular structures which indicate the codeposition of

PEDOT and RGO. A rough PEDOT surface is coated on RGO by π - π interactions [13]. Careful inspection of Figure 3(c) revealed a layered peeled-off coating of RGO on the surface. Globular traces were also observed due to the presence of PEDOT in the composite. Figure 3(d) was observed to have almost a similar morphology as in Figure 3(b) with the

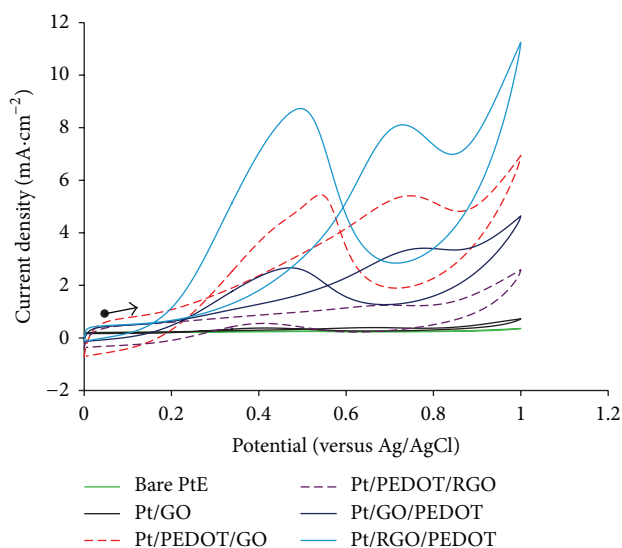


FIGURE 5: CV profiles for the oxidation of 1.0 M ethanol solution on bare Pt electrode (PtE) and Pt particles dispersed on different composite electrodes supported on glassy carbon electrode.

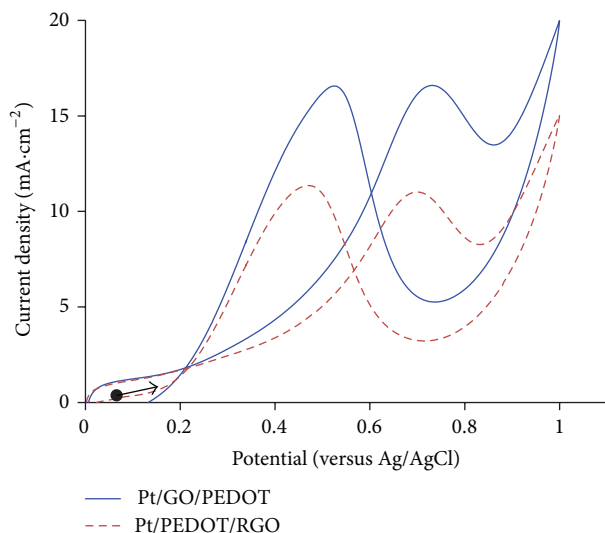


FIGURE 6: CV profiles for the oxidation of 1.0 M ethanol solution on Pt/RGO/PEDOT and Pt/PEDOT/RGO composites supported on gold substrate.

characteristic globular morphology of PEDOT and folded and crumpled-like morphology of graphene. Bright particles are observed which could easily be assigned to platinum on the composite as verified by energy dispersed X-ray (EDX) analysis. Patra and Munichandraiah [7] reported that the interactions between Pt nanoparticles and PEDOT were stronger compared with those of Pt nanoparticles on a smooth substrate like highly ordered pyrolytic graphite and glassy carbon.

Elemental composition using EDX analysis is necessary to support the SEM micrographs of the composites presented

in Figure 3. As shown in Figure 4(a), the percentage elemental composition for Pt is 75.14% which confirms that platinum was deposited on the surface of Pt/PEDOT/RGO. The presence of carbon and oxygen is attributable to RGO and PEDOT. Meanwhile, the presence of sulfur and oxygen confirms the deposition of PEDOT on both composites, that is, Pt/PEDOT/RGO (Figure 4(a)) and Pt/RGO/PEDOT composites (Figure 4(b)). Interestingly, the percentages of sulfur and oxygen for Pt/PEDOT/RGO composite are higher compared to the Pt/RGO/PEDOT. Though it is plausible to correlate the elemental analysis with the stacking sequence of the two composites, it is more likely that the sequential deposition of PEDOT and RGO yields a hybrid composite.

The fabricated hybrid composites were electrochemically evaluated as potential electrocatalysts for ethanol oxidation. The anode of DEFCs is where oxidation of ethanol takes place. The oxidation of ethanol dictates the electron flow in the system; greater oxidation peak current in the CV profile could lead to greater efficiency of the fuel cell system. As shown in Figure 5, the Pt/RGO/PEDOT composite exhibits the highest electrocatalytic activity toward ethanol electrooxidation with an oxidation peak at around 0.70 V indicative of ethanol oxidation and a current density of $2689 \text{ mA} \cdot \text{cm}^{-2}$. Overlay of other stacking sequences such as Pt/PEDOT/RGO is also presented in the CV profile. Pt/PEDOT/RGO composite exhibits inferior electrocatalytic activity towards ethanol electrooxidation with a current density of $74.7 \text{ mA} \cdot \text{cm}^{-2}$ compared to Pt/RGO/PEDOT. The inferior electrocatalytic activity of Pt/PEDOT/RGO compared to Pt/RGO/PEDOT can be explained as follows. Graphene is more electroactive and conductive in its reduced form (i.e., RGO). Thus, applying an oxidation potential to electropolymerize EDOT to produce PEDOT could also oxidize the previously synthesized RGO forming mostly oxidized graphene (GO) instead of RGO. Electrooxidation of ethanol on bare platinum ($20 \text{ mA} \cdot \text{cm}^{-2}$) and Pt/GO ($45.5 \text{ mA} \cdot \text{cm}^{-2}$) gave weak anodic peak currents compared to Pt dispersed on the hybrid composites.

The SEM studies of Pt/PEDOT/RGO and Pt/RGO/PEDOT composites were done on gold wire to investigate the surface morphology because the cylindrical GCE available in our laboratory is not suitable for SEM analysis. Thus, ethanol electrooxidation was also done in these samples. As shown in Figure 6, Pt/RGO/PEDOT composite still exhibits higher electrocatalytic activity toward ethanol oxidation than Pt/PEDOT/RGO.

4. Conclusion

In this study, the preparation of RGO/PEDOT composite as a support matrix for platinum microparticles was done electrochemically, routing faster and greener way of fabrication. Graphene oxide was fabricated from electrochemical exfoliation of graphite from carbon rod of used batteries which promotes a possible low cost and faster and greener production of graphene oxide. SEM analysis of EGO was observed to have a folded and wrinkled morphology which could indicate the electrochemical exfoliation of graphite to graphene-based materials.

The synthesis of Pt/RGO/PEDOT was monitored using cyclic voltammetry. Pt/RGO/PEDOT gave the highest electrocatalytic activity towards ethanol oxidation compared to bare Pt electrode and other composites. This study also demonstrates the utility of electrochemistry as a faster and simpler method of fabricating hybrid composite-modified electrode as potential anode materials for direct ethanol fuel cell application.

Conflict of Interests

There is no conflict of interests in this study.

Acknowledgment

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