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Engineering of Pt@Ni,N-doped graphene electrocatalyst based on recycled water bottles waste as an efficient cathode material for PEM fuel cells

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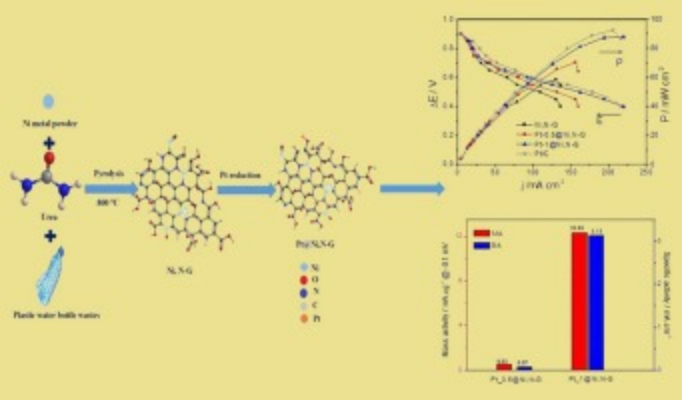
Highlights

- Development of sustainable electrocatalysts for PEM fuel cells derived from spent water bottles
- Evaluating of electrochemical and physicochemical properties of prepared nano-composite catalyst
- Pt-1@Ni,N-G has promising features as a graphene based electrocatalyst with low content of Pt.

Abstract

Fuel cells (FCs) represent one of the most promising clean power sources. Their cost represents a considerable challenge to their widespread commercialization because of their sensitivity and the expensive cost of the employed platinum electrocatalysts. Accordingly, one feature of a fuel cell device that many interested parties are looking for is the engineering of catalysts with a low Pt content. Herewith, Pt supported on nickel loaded nitrogen-doped graphene (Pt@Ni,N-G) was synthesized through a simple pyrolysis process of mineral water bottles waste with urea and nickel metal at 800 °C followed by loading Pt in different proportions via a simple reduction method in aqueous solution at room temperature. The physicochemical and electrochemical properties of a promising Pt@Ni,N-G cathode electrocatalyst for polymer electrolyte membrane fuel cell (PEMFC) were characterized and evaluated using a number of specialized techniques. The results showed that the high Pt@Ni,N-G content enhanced the catalytic activity of the oxygen reduction reaction (ORR), with a specific activity of 3.13 $mA cm^{-2}$ and a mass activity of 12.35 $mA \mu g^{-1}_{Pt}$ at −0.1 V. A maximum power density of 87.8 $mW cm^{-2}$ was achieved which is closest to that of commercial Pt/C. Designing Pt@Ni,N-G electrocatalysts by recycling waste bottles is not only expected to reduce the cost of fuel cells by ~30% to raise the industry standard but also protect the environment and humanity from harmful pollutants.

Graphical abstract



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Introduction

Ecofriendly energy storage and conversion devices are severely needed to solve worldwide energy crises. Typical fuel cells are devices that convert chemical energy from hydrogen into electricity. In the 1970s, liquid hydrogen was used by NASA as a fuel for space shuttles, more

recently, in fuel cell electric vehicles (FCEVs), military applications, power plants, and housing power [1]. FCs outperform combustion engines in terms of operational efficiency, with an electrical energy conversion efficiency of 60% or higher and lower emissions and noise levels [2]. Based on the type of used electrolyte, fuel cells are classified into polymer electrolyte fuel cells (PEMFCs), alkaline fuel cells (AFCs), phosphoric acid fuel cells (PAFCs), molten carbonate fuel cells (MCFCs), and solid oxide fuel cells (SOFCs). Among different type of fuel cells, polymer electrolyte membrane (PEM) fuel cell is being developed primarily for transportation applications, along with stationary and portable fuel cell uses [3,4]. The widespread use of FCs is still limited by their sensitivity and the high cost of using Pt electrocatalysts [5]. To reduce its cost, the Pt loading should be reduced by increasing the activity of the oxygen reduction reaction (ORR) catalyst, which has five times slower kinetics than the hydrogen oxidation reaction (HOR) at the anode. Therefore, in the last decade, tremendous progress has been made in improving ORR activity [[6], [7], [8], [9], [10], [11], [12], [13], [14], [15]] to accelerate its kinetics and reduce the Pt loading by combining with other transition metals such as Ni, Ag, Fe, or Co [6,10,14,15].

Generally, Pt-based nanoparticles typically tend to aggregate, due to their high surface energy [16]. Therefore, to reduce their agglomeration and enhance catalytic activity, it is required to use materials with a high surface area as anchoring sites for Pt-nanoparticles [13]. Nevertheless, the catalytic activity, stability, and performance of catalysts towards ORR are significantly affected by support materials. Therefore, some desired properties of suitable support materials are required, such as good conductivity, high surface area, and high porosity [16]. However, carbon-based materials have distinctive properties to be used as a support material for nanoscale catalysts. Carbon black is frequently employed among the available carbon support materials [17,18]. However, the cathode's operating circumstances are complex, such as the electrode's high overpotentials [19]. As a result, this situation leads to the degradation of the support material, separating and aggregating Pt nanoparticles, lowering their performance and shortening the catalyst's lifetime [16]. Therefore, research on novel support materials made from carbon structures is geared to improve ORR catalysts' stability and catalytic activity.

Plastic waste poses severe environmental hazards since it degrades slowly. Still, because it primarily consists of carbon, it can be the most readily accessible precursor for producing carbon-based electrocatalysts. Upcycling plastic waste can be a simple and cost-effective approach to synthesizing carbon-based ORR electrocatalysts [[20], [21], [22], [23]]. As a result, both the inadequate management of plastic waste and the energy crisis can be addressed at once. Generally, carbon-based Pt-free electrocatalysts comprise more than 99% of a carbon backbone, in which defects containing nitrogen and metals coordinated with

nitrogen are embedded into the graphitic/graphene-like structure [24]. Graphene is often used as a support material in energy applications due to its superior conductivity, extraordinary layered structure with high specific surface area, and high chemical and thermal durability [[25], [26], [27], [28], [29]].

Herein, nickel and nitrogen-doped graphene (Ni,N-G) is produced from the thermal decomposition of PET bottles waste with urea with nickel metal. This stable and high surface area substrate supports Pt nanoparticles and enhances its performance as cathodic electrocatalysts in HFCs. When N atoms are added to the graphene structure, the electron density of the nearby C atoms is reorganized, causing an electrophilic center to form next to the N atoms. This alters the geometry and enhances the graphene's ability to donate electrons [[30], [31], [32]]. Compared to previously used methods, the proposed N-G synthesis offers several benefits, including simple reaction setup and operating stages. This quick, easy, and environmentally friendly one-pot process can be easily industrial scaled up. Accordingly, fuel cell commercialization may become more likely if mass-produced at a low cost and scale. Using cyclic voltammetry (CV) and linear scan voltammetry (LSV), the electrochemical evaluation of the produced Pt@Ni,N-G composite electrocatalysts was carried out, illustrating the possible use of these materials in PEMFCs.

Section snippets

Preparation of Pt@GN/Ni

In a process similar to that described by Elessawy et al., Ni,N-G (3 wt% of Ni metal, 99%, Sigma–Aldrich) was synthesized from polyethylene terephthalate (PET) plastic water bottle wastes by pyrolyzing them with urea and Ni metal at 800 °C for 1 h [23,32]. The final powder product was collected and finely ground. Pt@Ni,N-G catalysts with two Pt metal loading ratios 0.9 and 1.8 wt% were synthesized by gently mixing Ni,N-G powder and chloroplatinic acid solution (H_2PtCl_6.8wt.%H_2O, Sigma-Aldrich) …

Results and discussion

Ni,N-G was synthesized from PET plastic water bottle waste by pyrolyzing them with urea and Ni metal (3 wt% of bottle waste) at 800 °C for 1 h. According to the previous studies on the thermal decomposition of PET and urea [32,34,35], the carbon skeleton of PET starts to fracture after 400 °C, while graphitic carbon nitride (g-C_3N_4) can be formed from urea above

550°C [36,37]. Above 600°C, nickel acts as a catalyst to produce GN sheets by supporting the interaction between the pyrolyzed…

Conclusion

The proposed study showed that the Pt-1@Ni,N-G cathode electrocatalyst prepared from the thermal dissociation of polyethylene terephthalate bottles waste with urea and nickel is a promising electrocatalyst with activities for FCs. The electrochemical results show that the ORR activity and durability of N-doped graphene electrocatalysts can be greatly enhanced by the addition of Pt, even in low Pt loading. Furthermore, the catalyst porosity has an additional effect whereas, Pt-1@Ni,N-G has a…

Data availability statements

All data generated or analyzed during this study are included in this published article.…

CRediT authorship contribution statement

Noha A. Elessawy: Writing – original draft, Visualization, Validation, Project administration, Methodology, Investigation, Formal analysis, Conceptualization. **Sami A. Al-Hussain:** Formal analysis, Writing – review & editing. **Arafat Toghan:** Writing – original draft, Writing – review & editing, Conceptualization, Methodology, Visualization, Investigation, Formal analysis, Data curation, Validation, Supervision, Project administration.…

Declaration of competing interest

The authors declare no competing interests.…

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