

## Supporting Information

Article

# Palladium-Nickel Electrocatalysts on Nitrogen-Doped Reduced Graphene Oxide Nanosheets for Direct Hydrazine/Hydrogen Peroxide Fuel Cells

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### *Synthesis of graphene oxide (GO)*

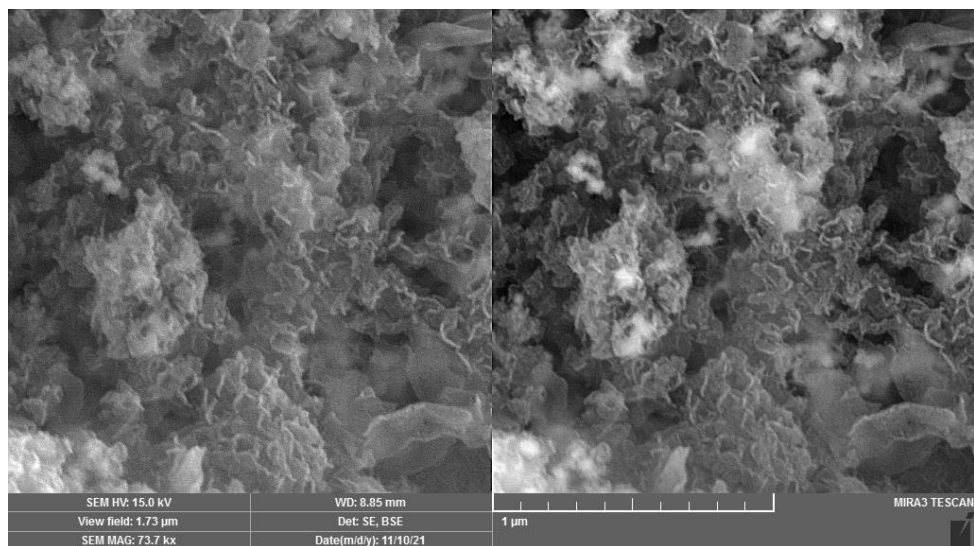
GO was prepared by the modified Hummers method, as previously described [1,2]. A natural graphite powder (5.0 g) was added to a flask containing 120 mL of sulfuric acid solution in an ice bath. 2.5 g of sodium nitrate and 15 g of potassium permanganate were added to the suspension without stirring. After the ice bath was ejected, the solution was stirred at 50 °C for 12 h. Then, 150 mL of ultrapure water was added gradually to the mixture and refluxed at 98 °C for 24 h. Afterwards, 50 mL of aqueous hydrogen peroxide solution was added to this mixture to convert the insoluble manganese species to Mn<sup>2+</sup> ions. Then, the mixture was centrifuged using hydrogen chloride to obtain the GO. The GO was rinsed with Millipore water several times, and a viscous brown dispersion is formed, filtrated, and finally dried at 65 °C under vacuum.

### *Activation of membrane electrode assembly (MEA)*

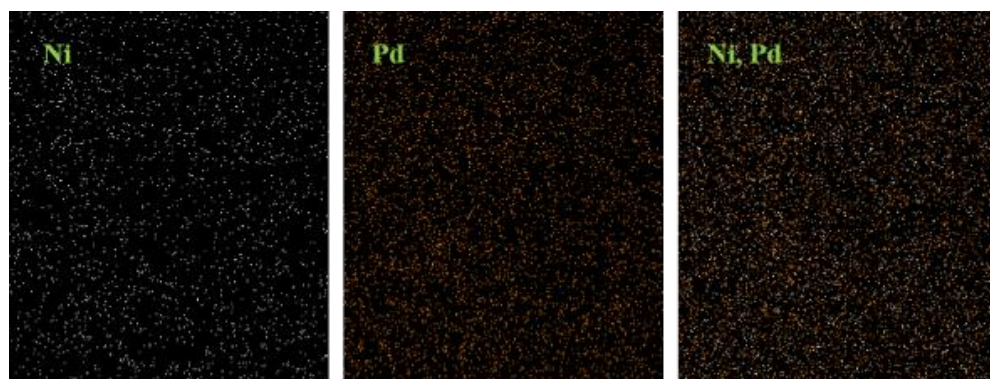
At the first, MEAs were placed in fuel cell set up. Water was flowed from each electrode for 5 h followed then by flowing NaOH 2.0 mol L<sup>-1</sup> from anode for 5 h to convert the cations in the sulfonic groups of the Nafion membrane from H<sup>+</sup> form to Na<sup>+</sup> form. Then, MEAs were activated by feeding alkaline hydrazine solution (NaOH 2.0 mol L<sup>-1</sup> + 1.0 N<sub>2</sub>H<sub>4</sub>) from anode and acidic hydrogen peroxide (H<sub>2</sub>SO<sub>4</sub> 0.5 mol L<sup>-1</sup> + H<sub>2</sub>O<sub>2</sub> 0.5 mol L<sup>-1</sup>) from cathode. The load was applied in steps of 20 mA within the range of 0-200 mA. Each step lasted for 10 min at 60 °C. After activation of MEA, the current density-potential (I-V) and current density-power density (I-P) curves were recorded using homemade electric load system.

**Table S1.** A brief summary of the purity of the used materials.

Material	Formula	Mass percent purity	Cas No.	Source	Country
Graphite powder	C	99.99%	7782-42-5	Sigma Aldrich	USA
Anhydrous palladium chloride	PdCl <sub>2</sub>	59.0-60.0% Pd basis	7647-10-1	Sigma Aldrich	USA
Hydrous Nickel chloride	NiCl <sub>2</sub> .6H <sub>2</sub> O	≥ 98%	7791-20-0	Merck	Germany
Sulfuric acid	H <sub>2</sub> SO <sub>4</sub>	98.0%	7664-93-9	Merck	Germany
Hydrochloric acid	HCl	37.0%	7647-01-0	Merck	Germany
Hydrus hydrazine	N <sub>2</sub> H <sub>4</sub> . H <sub>2</sub> O	98.0%	7803-57-8	Merck	Germany
Hydrogen peroxide	H <sub>2</sub> O <sub>2</sub>	30.0%	499-83-2,7722-84-1	Merck	Germany
Sodium borohydride	NaBH <sub>4</sub>	98.0%	16940-66-2	Alfa Aesar	North America
Urea	CH <sub>4</sub> N <sub>2</sub> O	> 99.0%	57-13-6	Merck	Germany
Potassium permanganate	KMnO <sub>4</sub>	99.0%	7722-64-7	Sigma Aldrich	USA
Nafion solution	C <sub>7</sub> HF <sub>13</sub> O <sub>5</sub> S·C <sub>2</sub> F <sub>4</sub>	5.0%	31175-20-9	Dupont	USA
Sodium hydroxide	NaOH	> 99.9%	1310-73-2	Fluka	Switzerland
2-propanol	C <sub>3</sub> H <sub>8</sub> O	≥ 99.8%		Merck	Germany
Water	H <sub>2</sub> O	Millipore water with conductivity 0.055 μS.cm <sup>-1</sup>	-	-	Iran



**Figure S1.** The BSE images of Pd-Ni/NrGO catalyst.



**Figure S2.** The elemental mapping images of each element in Pd-Ni/NrGO [3], Copyright 2021 American Chemical Society.

## References

- [1]. Hummers, W. S.; Offeman, R. E., Preparation of Graphitic Oxide, *J. Am. Chem. Soc.* **1958**, *80*, 1339.
- [2]. Hirata M.; Gotou, T.; Horiuchi, S.; Fujiwara, M.; Ohba, M., Thin-film particles of graphite oxide 1: High-yield synthesis and flexibility of the particles, *Carbon*, **2004**, *42*, 2929.
- [3]. Hosseini, M. G; Daneshvari-Esfahlan, V.; Wolf S.; Hacker, V., Novel Bimetallic Pd-X (X = Ni, Co) Nanoparticles Assembled on N-Doped Reduced Graphene Oxide as an Anode Catalyst for Highly Efficient Direct Sodium Borohydride–Hydrogen Peroxide Fuel Cells, *ACS App. Energy Mater.*, **2021**, *4*, 6025-6039.