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N/O co-doped porous carbon derived from polyester waste for electrochemical production of H₂O₂†

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Electrochemical synthesis of hydrogen peroxide (H₂O₂) has been considered as an ideal alternative to traditional anthraquinone techniques due to its convenient operation and environmental friendliness. However, the development of catalysts with high activity, selectivity, and stability is still an important challenge. In this study, waste polyethylene terephthalate (PET) was converted into a high-value carbon-based catalyst for the electrochemical production of H₂O₂. Nitrogen–oxygen co-doped porous carbon (NOPC) was obtained by pyrolysis of PET with addition of melamine as a nitrogen source and magnesium oxide (MgO) as a templating agent. The cross-linking reaction during the pyrolysis process tightly bound nitrogen (N) and oxygen (O) active sites via amide group formation. These groups were uniformly distributed within the carbon matrix, accompanied by numerous defects, thereby enhancing both activity and selectivity. The optimized PET-derived NOPC catalysts exhibited a H₂O₂ selectivity of approximately 85% with an electron transfer number close to 2.3, and demonstrated long-term stability for over 10 hours in acidic solution. The electrocatalytic H₂O₂ yield in a custom-built cell reached 261.23 mmol g_{catalyst}⁻¹ h⁻¹ with faradaic efficiency above 80%. This work presents an efficient synthesis method for 2-electron oxygen reduction reaction (2e⁻ ORR) electrocatalysts and provides a sustainable strategy for recycling waste plastics into valuable carbon materials.

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Introduction

Electrochemical synthesis of H₂O₂ by 2e⁻ oxygen reduction reaction (ORR) is considered as an environmentally friendly way for the development of green and sustainable chemistry.¹ The 4-electron ORR process generating water is the main competitive reaction in the electrocatalytic preparation of H₂O₂.² Consequently, electrocatalysts with high activity and selectivity for 2e⁻ ORR have gained increasing attention in recent years.³ Precious metal (Pt, Pd, Au) alloy catalysts such as Pt–Hg, Pd–Hg, and Ag–Hg for H₂O₂ production have been reported,^{4,5} which exhibit excellent catalytic properties due to the weakened binding energy of the intermediates (*OOH).⁶ Unfortunately, the high cost and scarcity of precious metals limit their wide application on a large scale.⁷ On the other hand, carbon-based materials are more promising electrocatalysts for 2e⁻ ORR

due to their advantages of low cost, abundant resources, good conductivity, and stability.⁸ To date, various carbon materials have been explored for the electrochemical production of H₂O₂.^{9,10} It has been reported that the microstructure and composition of carbon will affect the catalytic activity and selectivity.^{11,12} The defect structures, oxygen-containing functional groups, and heteroatomic doping could increase the catalytic activity of carbon materials.^{3,13,14}

Generally, the modification of the pore structure of carbon materials is an effective strategy for enhancing their electrochemical performance. The mesoporous structures improve mass transfer and optimize electron transfer pathways, promoting H₂O₂ production during the 2e⁻ ORR.¹⁵ However, precise regulation of the pore structure remains challenging due to factors such as synthesis methods, heat treatment processes, and material properties.^{7,16,17} Effective pore-making strategies, including templating, activation, and modifications of synthesis conditions, have been developed to address these difficulties.^{16,18,19}

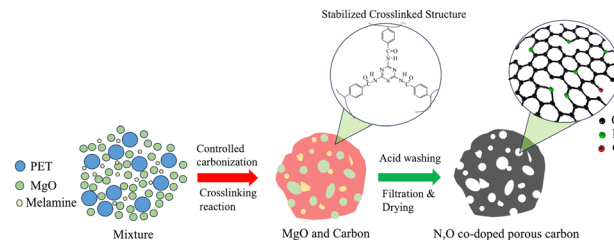
Additionally, heterogeneous atom doping (N,²⁰ P,²¹ S,²² etc.) has been found to adjust the microstructure of carbon materials, thereby modifying the electronic structure of active sites and improving catalytic performance. Kinetic analysis and theoretical calculation revealed that the synergistic interaction between

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nitrogen and oxygen active sites facilitated the conversion of *OOH to H_2O_2 by reducing *OOH adsorption and retaining O–O bonds.²³ Currently, most attention has been paid to carbon-based electrocatalysts for electrosynthesis of H_2O_2 in alkaline media. However, H_2O_2 is more stable under acid conditions than in bases, and is more widely used in acid media with stronger oxidation ability. Consequently, the development of carbon-based catalysts for H_2O_2 production under acid conditions is of great importance.

Polyethylene terephthalate (PET) is one of the most commonly used polymers for packaging and beverage containers in our daily lives. However, owing to its superior stability, PET waste resists biodegradation under natural conditions, which leads to serious environmental and health issues. Recently, the conversion of PET to high-value carbon materials for different applications in the renewable energy field has been explored. Early research on the synthesis of high surface area porous carbon nanosheets through the catalytic carbonization of PET and KOH activation showed that this carbon material served as an ideal electrode for double-layer supercapacitors,²⁴ exhibiting excellent capacitive performance. Mohd Ubaidullah and co-workers promoted this research by converting PET to metal-organic frameworks (MOF-5), and then producing nitrogen-doped mesoporous carbon for supercapacitor applications.¹⁸ Nitrogen-doped microporous carbon produced by the controlled carbonization of PET using molten salts also demonstrated exceptional CO_2 adsorption capacity.¹⁶ Hard carbon anode materials with constructed closed-pore structures derived from PET have been applied in sodium-ion batteries.²⁵ In addition, layered porous nitrogen-doped carbon derived from PET has also been applied as an efficient electrocatalytic carrier for hydrogen evolution reaction (HER) under alkaline conditions.¹⁹ These studies demonstrate that the trash-to-treasure strategy of recycling plastic waste into energy materials might be a promising way to resolve the issue of white pollution. Although PET-derived carbon materials have been applied in many fields, few studies have been reported for the development of electrocatalysts, especially for the electrosynthesis of hydrogen peroxide. Actually, the pyrolysis of PET disrupts weak oxygen-containing bonds, leading to partial collapse of the carbon skeleton and resulting in low-porosity, low-activity carbon materials.¹⁶ Therefore, controlled carbonization of PET to obtain carbon materials with high electrocatalytic activity remains significantly challenging.

Herein, we propose a method to prepare nitrogen–oxygen co-doped porous carbon (NOPC) by carbonization of PET with melamine and MgO. It was found that melamine could react with the intermediates during the PET pyrolysis process to form a stable cross-linked structure. MgO was used as a template agent to modulate the pore structure of the obtained PET-derived carbon. N,O synergistic carbon catalysts with mesoporous structures have been successfully synthesized. The optimized catalyst demonstrated favourable $2e^-$ ORR activity and excellent H_2O_2 selectivity under acidic conditions. Moreover, the catalyst maintained high selectivity for at least 10 hours in acidic electrolytes. This work provides a promising and sustainable method to convert waste PET to NOPC



Scheme 1 Schematic illustration of the conversion process of PET to NOPC for hydrogen peroxide production.

for the electrochemical production of hydrogen peroxide (Scheme 1).

Results and discussion

Material synthesis and characterization

MgO was mixed with PET in different mass ratios, and the mixture was then heated, in order to regulate the additive amount of MgO. The obtained carbon materials after the carbonization process were studied. Fig. S2 (ESI[†]) shows the SEM images and EDS results of the samples of OPC1-800. After acid leaching, irregular macroporous structures were clearly observed on the surface of the samples (Fig. S2a–c, ESI[†]). During heat treatment, PET initially fused and MgO nanoparticles dispersed within the melted PET, leading to molecular rearrangement around the MgO particles. At the same time, some MgO nanoparticles agglomerated and their particle size increased. MgO particles initially encapsulated by carbonation products were removed during HCl leaching, resulting in the formation of a macroporous structure. The EDS result of OPC1-800 exhibited a trace amount of Mg (0.84 at%) and Cl (1.08 at%), indicating that most of MgO had been removed by HCl leaching (Fig. S2d, ESI[†]). The XRD pattern of OPC1-800 showed a broad peak at $2\theta = 23.22^\circ$ owing to the characteristic diffraction peak of the graphite (002) plane, which suggested an amorphous structure in the PET-derived carbon and confirmed the effective removal of MgO again (Fig. S3, ESI[†]).

Nitrogen adsorption–desorption curves and pore size distribution for different PET-derived carbon materials are shown in Fig. S4 (ESI[†]), which exhibits a typical type-IV isotherm. The low adsorption capacity at a lower relative pressure ($P/P_0 < 0.1$) indicated less microporous structure in the obtained carbon. The significant hysteresis loops indicated that all materials had mesoporous structures (Fig. S4a, ESI[†]). The specific surface areas for all OPC samples were about $700 \text{ m}^2 \text{ g}^{-1}$ as listed in Table S1 (ESI[†]), which demonstrated that MgO played an important role in forming the mesoporous structure. The pore size distribution curves indicated that the pore size in the carbon materials was in the range of 3–20 nm (Fig. S4b, ESI[†]). Especially, the pore size at about 10–20 nm was in accordance with the diameter of MgO nanoparticles. The mesopore volume increased with the addition of MgO and reached the maximum value of $1.22 \text{ cm}^3 \text{ g}^{-1}$ when M/P was 1, and then dropped to $1.09 \text{ cm}^3 \text{ g}^{-1}$ for OPC2-800 (Table S1, ESI[†]).



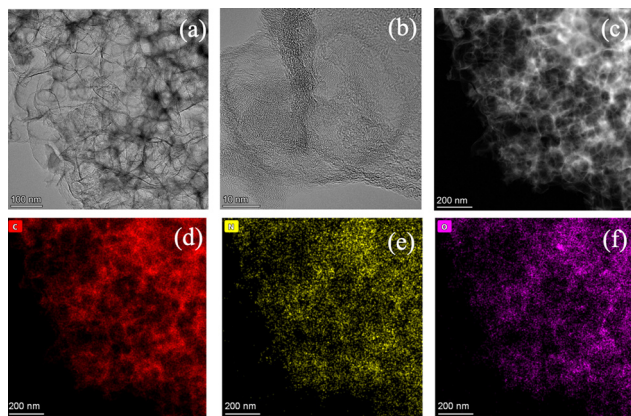


Fig. 2 (a) TEM and (b) HRTEM images of NOPC-0.1. (c) HAADF-STEM image and STEM-EDS mapping profiles for (d) C, (e) N, and (f) O of NOPC-0.1.

introduce nitrogen into PET-derived carbon by cross-linking with the PET degradation products.

XRD patterns of NOPC samples after HCl/HNO₃ washing revealed two broad peaks at $2\theta = 23.22^\circ$ and 43.8° (Fig. S3, ESI[†]), corresponding to the (002) and (100) diffraction planes of graphitic carbon, respectively.³⁴ TEM analysis was conducted to investigate the micromorphology of NOPC. Fig. 2(a) and (b), and Fig. S7 (ESI[†]) show the obvious porous structure of PET-derived carbon. Moreover, the carbon exhibited a low degree of graphitization. High-angle annular dark field-scanning transmission microscopy (HAADF-STEM) and EDS analysis confirmed the plenty of pores and the uniform distribution of C, N, and O elements within the materials (Fig. 2(c)–(f)). N₂ adsorption–desorption isotherms for NOPC samples are shown in Fig. 3(a). NOPC-0.05 and NOPC-0.1 displayed similar curve shapes to those without melamine addition (OPC), showing hysteresis loops at a relative pressure of $0.4 < P/P_0 < 1.0$ (Fig. 3(a)). On the other hand, with the increase of melamine, NOPC-0.3 and NOPC-0.5 exhibited a larger N₂ uptake at a low relative pressure ($P/P_0 < 0.1$) and almost no hysteresis loops, which indicated the presence of more micropores and fewer mesopores. The pore size distribution curves confirmed this change (Fig. 3b). More pores with a size of 3–5 nm formed in the samples of NOPC-0.3 and NOPC-0.5, while there were almost no pores in the range of 10–30 nm in these samples. The reason for this might be that more melamine decomposed without cross-linking with PET degradation products when a higher mass ratio of melamine was applied. Moreover, the specific surface areas of NOPC-0.3 and NOPC-0.5 in Table S1 (ESI[†]) were $718.8 \text{ m}^2 \text{ g}^{-1}$ and $696.5 \text{ m}^2 \text{ g}^{-1}$, respectively, which were much higher than those of NOPC-0.05 ($409.26 \text{ m}^2 \text{ g}^{-1}$) and NOPC-0.1 ($389.77 \text{ m}^2 \text{ g}^{-1}$), confirming more microporous structures again. The average pore size concluded in Table S1 (ESI[†]) indicated that in the absence of melamine, the pyrolysis of PET with MgO resulted in molecular chain breakage and rearrangement, with pore formation primarily attributed to the removal of MgO. Consequently, OPCs exhibited a relatively stable average pore size. However, with the addition of a small

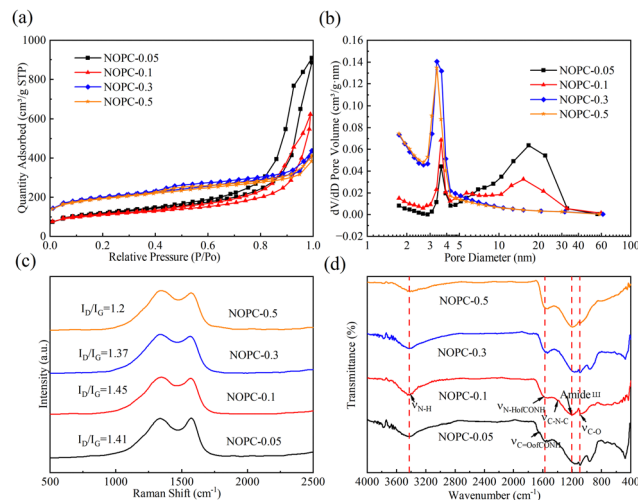


Fig. 3 (a) N₂ adsorption–desorption isotherms, (b) pore size distribution, (c) Raman spectra, (d) FT-IR spectra corresponding to different NOPC samples.

amount of melamine, the average pore size increased, likely due to melamine acting as a chain extender, thereby altering the cleavage and rearrangement of PET molecular chains. With the further increase of melamine content, the pore size decreased. The reason for this might be that more micropores formed, making the average pore size decrease. Additionally, the denser crosslinked network reduced the average pore size.

In Raman spectroscopy (Fig. 3(c)), the peaks at 1350 cm^{-1} and 1576 cm^{-1} corresponded to the D-band and G-band, respectively.^{16,35} The D-band was associated with the introduction of $\text{sp}^3\text{-C}$ bonds and structural defects at edges or basal planes. The G-band was related to graphitic carbon.³⁶ The intensity ratio of the D-band to the G-band (I_D/I_G) was usually used to describe the relative defect content in carbon materials.¹³ The I_D/I_G values for NOPC-0.05, NOPC-0.1, NOPC-0.3, and NOPC-0.5 were 1.41, 1.45, 1.37, and 1.2, respectively, indicating that the defect content dropped with the increase of melamine. These abundant defects were associated with $\text{sp}^3\text{-C}$ and edge sites in mesoporous carbon materials,⁷ which could act as active sites for the adsorption or reaction during the electrocatalytic process.

The infrared spectrum analysis (FT-IR) showed the peak at 1571 cm^{-1} in NOPC-0.1, which was attributed to the N–H stretching vibration of the amide group (Fig. 3(d)). The peak at 1205 cm^{-1} was assigned to the amide III band, while the peak at 1095 cm^{-1} corresponded to the C–O stretching vibration of the amide group. The peaks at 1396 cm^{-1} and 3426 cm^{-1} were associated with C–N–C symmetric stretching and N–H asymmetric stretching, respectively. These functional groups clearly demonstrated the presence of amide groups in the melamine-doped carbonized products. The appearance of amide groups further suggested that the N and O active sites were tightly bonded in the PET-derived carbon materials.³⁷

XPS was used to characterize the surface elements and functional groups, which showed the presence of C, N, and O



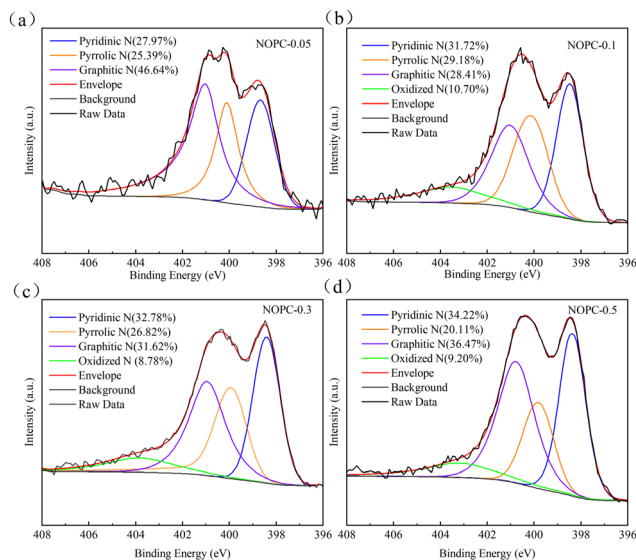


Fig. 4 High-resolution N 1s XPS spectra of different NOPC samples: (a) NOPC-0.05, (b) NOPC-0.1, (c) NOPC-0.3, and (d) NOPC-0.5.

elements in all NOPC samples (Fig. S8, ESI[†]). The content of nitrogen increased with the increase of melamine addition (Table S2, ESI[†]), demonstrating that nitrogen originated from melamine and the effective cross-linking reaction between melamine and PET. The N 1s spectra of the NOPC samples were deconvoluted into four peaks (Fig. 4): pyridinic N (398.48 eV), pyrrolic N (400.11 eV), graphitic N (401.03 eV), and oxidized N (403.53 eV),^{38,39} and their relative atomic percentages are concluded in Table S3 (ESI[†]). NOPC-0.1 exhibited a high percent of pyrrolic N, which was reported to be responsible for the optimal selectivity for 2e⁻ ORR.⁴⁰ Generally, more nitrogen doping would contribute to the 4e⁻ ORR process.^{41,42} Compared to OPC1-800, the oxygen content also increased for NOPC samples, resulting from the acid leaching with HNO₃. The high-resolution C 1s spectra of the NOPC samples shown in Fig. S9 (ESI[†]) could be divided into four distinct peaks: graphite carbon (284.80 eV), C=N (286.00 eV), C=O (288.13 eV), and $\pi \rightarrow \pi^*$ (290.73 eV).^{13,16} NOPC-0.1 was found to contain the highest amount of C=O (Table S4, ESI[†]). It has been reported that C=O bonds from carboxyl and carbonyl groups were considered as the key active sites for 2e⁻ ORR.⁴³ In addition, the high electronegativity of nitrogen atoms caused charge redistribution, which changed the adsorption strength of intermediates on carbon atoms and ultimately affected 2e⁻ ORR performance.^{39,44,45}

Electrochemical characterization

To investigate the electrochemical activity of the catalysts, cyclic voltammetry (CV) curves were measured under N₂ and O₂ atmospheres.⁴⁶ No significant peaks were observed under N₂-saturated conditions for all catalysts. However, distinct reduction peaks were observed in the O₂-saturated electrolyte, indicating the active oxygen reduction capabilities of all catalysts (Fig. S10, ESI[†]). The ORR catalytic activities of different

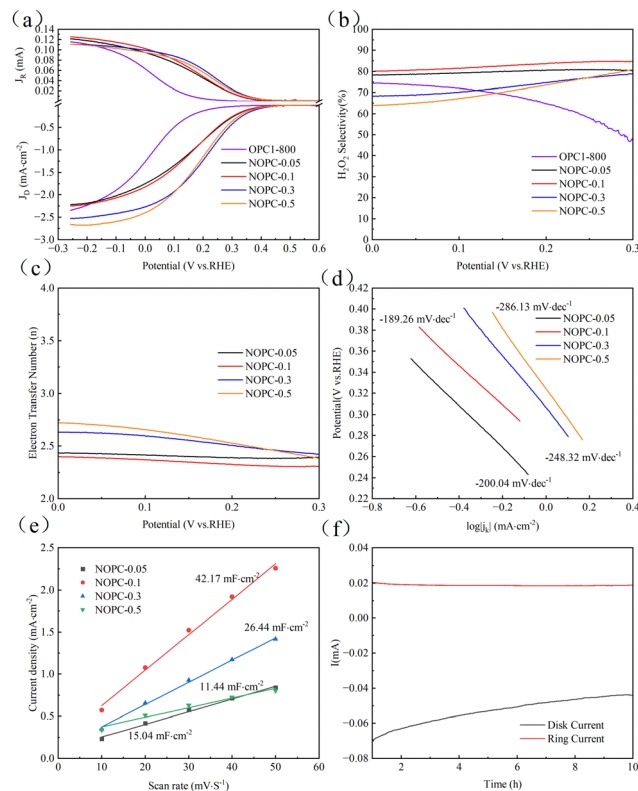


Fig. 5 Electrochemical test of the catalysts in 0.5 M H₂SO₄. (a) RRDE polarization curves of different catalysts. (b) H₂O₂ selectivity and (c) electron transfer number of different catalysts. (d) Tafel slope of different catalysts. (e) Capacitive current density at 0.59 V as a function of scan rate. (f) Stability test of NOPC-0.1 at a fixed disk potential of 0.3 V (vs. RHE).

catalysts were evaluated using linear sweep voltammetry (LSV). As displayed in Fig. 5(a), the LSV curves for all NOPC samples shifted positively to higher potential compared to that of OPC1-800, indicating that the nitrogen doping in carbon materials facilitated enhancement of the ORR catalytic activity. With the increase of melamine amount, an increased oxygen reduction current was observed and the onset potential shifted to higher potentials. The onset potential of OPC1-800 was measured to be 0.259 V, while that of NOPC-0.05, 0.1, 0.3, and 0.5 was determined to be 0.396 V, 0.401 V, 0.444 V, and 0.472 V, respectively. These results indicated that the activities of the NOPC catalysts were significantly enhanced after doping with a small amount of nitrogen.⁹

The H₂O₂ selectivity of different samples is shown in Fig. 5(b). As shown, within the potential range of 0–0.3 V, the H₂O₂ selectivity of NOPC samples was higher than that of OPC1-800. This demonstrated that nitrogen incorporation resulted in an enhanced oxygen reduction activity coupled with improved H₂O₂ selectivity. Among the NOPC catalysts, NOPC-0.1 exhibited the highest H₂O₂ selectivity of 80–84.7% in the potential range of 0–0.3 V. Moreover, the NOPC-0.1 catalyst showed a lower electron transfer number of about 2.3, which was also confirmed by the results calculated from the K–L equation (Fig. 5(c) and Fig. S11(a) and (b), ESI[†]). Catalysts with higher melamine doping (NOPC-0.3, NOPC-0.5) exhibited lower



H₂O₂ selectivity compared to NOPC-0.1. This suggested that moderate N doping helped to improve the H₂O₂ selectivity, but excessive N content facilitated the ORR by the 4e⁻ pathway with the formation of water. As reported, excessive N doping led to the aggregation of isolated active sites, which contributed to the breaking of the O–O bond and favoured the 4e⁻ ORR pathway. However, appropriate N doping limited the number of active sites, favouring the 2e⁻ ORR.⁹

Notably, there was no significant positive relationship between H₂O₂ selectivity and specific surface area. For NOPC-0.1, it had a large average pore size (13.77 nm) and the lowest specific surface area (389.77 m² g⁻¹), but it exhibited the highest selectivity (84.77%). However, a positive correlation between defect levels and selectivity was found. NOPC-0.1 with the highest I_D/I_G value showed the best electrocatalytic activity for H₂O₂ production, since the formation of defects provided active sites for adsorption or reactions, facilitating OOH* accumulation and increasing H₂O₂ selectivity.³⁶ Consequently, for PET-derived N,O-doped carbon materials, comprehensive effects of microstructure and composition on the catalytic activity and selectivity for the 2e⁻ ORR catalysts should be considered.

The Tafel slopes of the catalysts are shown in Fig. 5(d).⁴⁷ The Tafel slopes of NOPC-0.05, NOPC-0.1, NOPC-0.3, and NOPC-0.5 were 200.04 mV dec⁻¹, 189.26 mV dec⁻¹, 248.32 mV dec⁻¹, and 286.13 mV dec⁻¹, respectively. It was noteworthy that NOPC-0.1 had the lowest Tafel slope, indicating the optimum electrical conductivity and the lowest charge transfer resistance.^{3,13}

The electrochemical double-layer capacitance (C_{dl}) was often used to characterize the electrochemical specific surface area.¹³ The C_{dl} values for NOPC-0.05, NOPC-0.1, NOPC-0.3, and NOPC-0.5 were measured from CV curves at different scanning rates (Fig. S12, ESI[†]), which were 15.04 mF cm⁻², 42.17 mF cm⁻², 26.44 mF cm⁻², and 14.44 mF cm⁻², respectively (Fig. 5(e)). Generally, catalysts exhibiting high electrochemical double-layer capacitance values possessed more active sites exposed for O₂ adsorption, leading to enhancement of catalytic activity.⁴⁸ Electrochemical capacitance evaluation of NOPC indicated a linear correlation between the double-layer capacitance and the ring current at 0 V (Fig. S13, ESI[†]). This suggested that NOPC-0.1 with a more mesoporous structure could favour more active sites exposed to the electrolyte and facilitate the 2e⁻ oxygen reduction reaction for hydrogen peroxide generation. The stability of the catalysts under acidic conditions was evaluated by the amperometric *i*-*t* curve test.⁴⁹ The H₂O₂ ring current of NOPC-0.1 remained stable during 10 h of operation, indicating its good electrochemical stability and promising potential for practical application (Fig. 5(f)).

H₂O₂ production demonstration

The optimized PET-derived carbon materials, NOPC-0.1, were evaluated for H₂O₂ production at different applied potentials in the bulk electrolyte. Based on the standard concentration curve of Ce⁴⁺ (Fig. S14, ESI[†]), the accumulated amount of H₂O₂ concentration normalized by catalyst loading amount *versus* reaction time in O₂-saturated 0.5 M H₂SO₄ is shown in Fig. 6(a). It could be observed that the H₂O₂ production amount

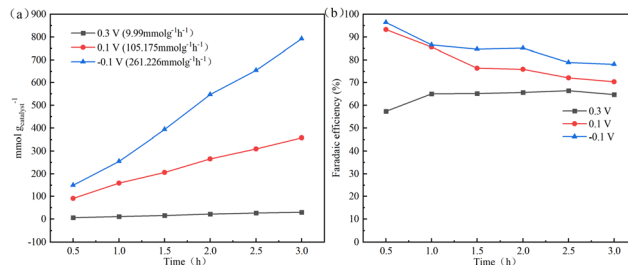


Fig. 6 (a) H₂O₂ production amount normalized by catalyst loading amount over the reaction time at different voltages, (b) the corresponding faradaic efficiencies (FE%).

increased as the potential shifted negatively in the potential range of -0.1–0.3 V. The plots of accumulated H₂O₂ concentration reflected an almost linear relationship between H₂O₂ production amount and reaction time. At -0.1 V, the NOPC-0.1 electrocatalyst achieved a high H₂O₂ generation rate of 261.23 mmol g_{catalyst}⁻¹ h⁻¹, comparable with those catalysts reported recently.⁹ As shown in Fig. 6(b), the faradaic efficiency of NOPC-0.1 was about 86.45% under a potential of -0.1 V, higher than that under the potential of 0.1 and 0.3 V. The faradaic efficiency decreased firstly and then stabilized relatively at about 76.38% and 65.27% when the potential was 0.1 V and 0.3 V, respectively. As anticipated, NOPC-0.1 exhibited a favourable faradaic efficiency for H₂O₂ production. As summarized in Table S5 (ESI[†]), the present NOPC-0.1 exhibited comparable or even better performance than those previously reported catalysts, further confirming the excellent electrochemical properties of NOPC-0.1. The practical demonstration suggested that the PET-derived porous carbon materials might be a promising catalyst for the electrochemical synthesis of H₂O₂.

Conclusions

In conclusion, a N,O co-doped porous carbon material was successfully synthesized, which was achieved through the cross-linking carbonization strategy of PET with melamine and MgO. The prepared NOPC exhibited an abundant mesoporous structure with N doping. The cross-linking carbonization process tightly bound the active sites of the catalysts through the formation of a large number of amide groups consisting of nitrogen and oxygen elements. The well-developed NOPC-0.1 catalyst showed a high H₂O₂ selectivity of 84.77% and long-time stability of 10 hours. Moreover, a high-yield electrosynthesis of H₂O₂ was achieved, with a production rate of 261.23 mmol g_{catalyst}⁻¹ h⁻¹ and an acceptable faradaic efficiency of 86.45%. Additionally, this study provided a promising approach for converting low-cost waste polyester into high-value carbon-based catalyst materials, as well advanced the development of waste-to-value strategies.

Author contributions

M. Luo designed experiments, synthesized materials, performed electrochemical characterization, and wrote the manuscript.



C. Wang contributed to data collection, analysis, and manuscript review. S. Song organized data and reviewed the manuscript. M. Tang assisted with data organization. X. Wang managed the project, reviewed and edited the manuscript. M. Wu reviewed the manuscript.

Data availability

The authors confirm that the data supporting the findings of this study are available within the article and its ESI.†

Conflicts of interest

There are no conflicts of interest to declare.

Acknowledgements

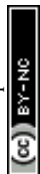
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