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CARBON-BASED GAS DIFFUSION ELECTRODES IN UVC/e-H_2O_2 TECHNOLOGY FOR THE REMOVAL OF ANTIDEPRESSANT RESIDUES (Oral)

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Amitriptyline (AMTP) is a tricyclic compound used for the treatment of depression, anxiety, and pain management. Despite the availability of newer substitute drugs in Brazil, its low cost and effectiveness maintain its popularity. AMTP can enter water systems through incomplete metabolism or improper disposal, impacting aquatic life even at low concentrations. Since conventional water treatments face difficulties in eliminating these types of recalcitrant compounds, developing new methods like UVC/e-H_2O_2 is essential.

Electrogenerated H_2O_2 (e-H_2O_2) using gas diffusion electrodes (GDE) is a promising technology for AMTP degradation, offering a simple, sustainable, and cost-effective system through the oxygen reduction reaction (ORR), which enables the in-situ production of H_2O_2 (a precursor of the hydroxyl radical, a stronger oxidizing species) and reduces the use of reagents. In this regard, the search for stable electrocatalysts with high activity and selectivity for the two-electron pathway has become a topic of interest in recent years (Wang et al., 2021; Xie et al., 2022). This study explores the use of La_2O_3 in GDE to improve oxygen reduction selectivity and activity in the two-electron pathway for AMTP degradation.

Electrochemical characterization

ORR was evaluated using the rotating disk electrode technique with La/C ratios (1.0-3.0%) supported on Printex L6 carbon. The electrolyte used was a 0.05 M K_2SO_4 solution (pH 9) saturated with O_2 during measurements. Experiments were conducted with an Autolab PGSTAT 302N potentiostat/galvanostat at a constant scan rate of 5.0 mV s^{-1} and rotation speeds ranging from 300 to 1,500 rpm. Figure 1 shows CV and LSV curves in basic media for ORR with 1.0-3.0% La on PCL6. The La/C ratios of 1.0% and 2.0% show a more positive onset potential for ORR and a higher ring current compared to PCL6, indicating selective reduction of O_2 through the $2e^-$ pathway. In contrast, the 3.0% La/C modifier exhibits a more negative onset potential for ORR and a lower ring current compared to PCL6, suggesting a decrease in H_2O_2 production, favouring the $4e^-$ pathway for water production.

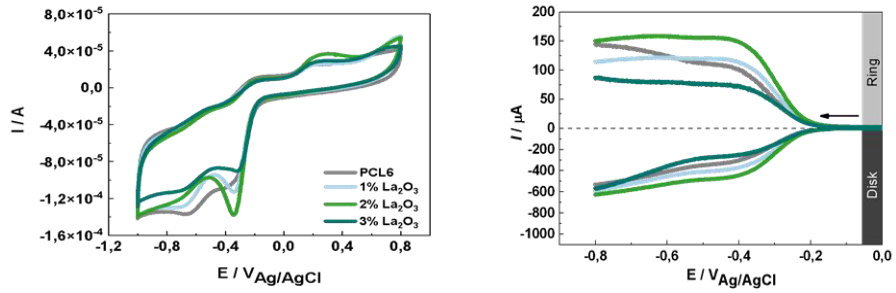

Figure 1. (A) CV and **(B)** LSV curves for the ORR at 1.0 – 3.0% content of La/C in a 0.05 M K_2SO_4 solution, saturated with $O_{2(g)}$. Electrode rotation speed: 900 rpm.

H_2O_2 quantification using La/C-GDE

The optimal metal/carbon ratio (2.0%) determined by the RRDE was utilized to prepare the gas diffusion electrodes (GDEs). Both modified and unmodified GDEs were tested in a non-divided cell to measure H_2O_2 production at various current densities. An Ag/AgCl (3 M KCl) electrode served as the reference, while a DSA® electrode was used as the counter electrode. A 250 mL 0.05 M K_2SO_4 solution acted as the supporting electrolyte, with O_2(g) supplied to the GDEs at a rate of 80 mL min^{-1}. H_2O_2 concentration was quantified using the peroxymolybdate complex method and analyzed via UV-visible spectrophotometry at 350 nm using a UV-1900 spectrophotometer. Fig. 2A shows electrogenerated H_2O_2 in 0.05 M K_2SO_4 at current densities of 10, 20, and 30 mA cm^{-2}. In all cases, the final accumulation of H_2O_2 is greater for the La-modified electrode, achieving optimal results at the highest current density studied, with a maximum concentration of H_2O_2 of 600 mg L^{-1} compared to 540 mg L^{-1}.

AMTP electrochemical degradation

The optimal current density, identified from H_2O_2 production studies, was applied to assess the degradation of 250 mL of 25 mg L^{-1} amitriptyline (AMTP) in 0.05 M K_2SO_4 (pH 9). The same non-divided cell used for H_2O_2 electrogeneration was utilized for this purpose. AMTP degradation and mineralization were conducted using the e-H_2O_2/UVC method, with a UV-C light source (NUCHONG 9W lamp) serving as the irradiation source. Fig. 2B shows the degradation of 25 mg L^{-1} of AMTP at 30 mA cm^{-2} by e-H_2O_2, e-H_2O_2/UVC, anodic oxidation (AO), and the AO/UVC process. The e-H_2O_2/UVC treatment shows the removal of 92.44% of the contaminant in 25 minutes of electrolysis and 49.58% mineralization after 1 hour.

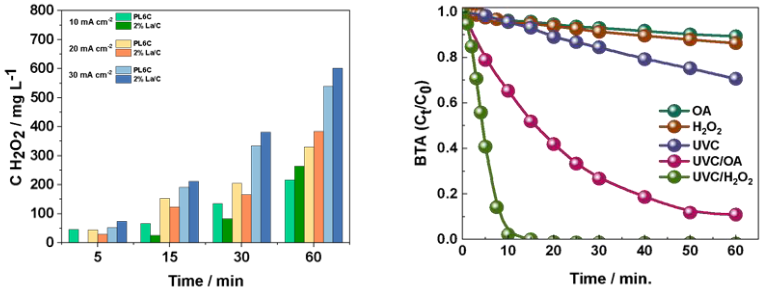

Figure 2. (A) Concentration of H_2O_2 electrogenerated in a 0.05 M K_2SO_4 as a function of the applied current density, and **(B)** AMTP degradation by e-H_2O_2, e-H_2O_2/UVC, AO and AO/UVC process at 30 mA cm^{-2}.

Conclusions

The La_2O_3 used as a modifier in low proportions demonstrated high performance for the ORR via 2-e- pathway, showing enhanced activity at lower potentials compared to PCL6, at basic condition.

Acknowledgments

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