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Towards environmentally friendly catalysts for H_2O_2 electrogeneration: obtaining and using amorphous carbon from waste as support for Pd-single atoms

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Hydrogen peroxide (H_2O_2) is one of the most important chemical compounds in modern society, being used in diverse applications such as a bleaching agent, medical disinfection, and environmental oxidant.[1] The prevailing method for its production involves anthraquinone reduction, constituting over 90% of the global production.[1] Due to challenges in storage and the need for limited quantities in certain applications, the in situ electrogeneration of H_2O_2 has garnered interest.[2] Typically, carbonaceous materials are employed in electrochemical H_2O_2 generation, either in their pristine state or as a support for metal catalysts. Commercially, the carbon used is derived through the unsustainable and environmentally unfriendly incomplete combustion of heavy petroleum products at temperatures as high as 2000 ºC. In this study, we propose a sustainable approach by utilizing carbon derived from renewable sources as a support for Pd single-atom dispersion. Three diverse sources were chosen: sewage sludge, a by-product of biological wastewater treatment plants with limited reuse due to high impurity levels; sugarcane bagasse, a residue generated in significant quantities (180 Mt in Brazil in the 2022/2023 crop); and tannin/cellulose, low-cost biomasses with potential for transformation into high-value functional materials. The methodology for obtaining carbon materials from sewage sludge and sugarcane bagasse involved impregnation followed by pyrolysis, varying temperature, holding time, and activator type/concentration. For tannin-cellulose, a xerogel route using HNO_3 was employed. Subsequently, Pd single-atom modification was achieved through a wet impregnation process using $Pd(acac)_2$ precursor. Results revealed distinct physical morphologies in the obtained amorphous carbon, particularly in structure, total surface area, and functional groups. Nonetheless,

the modification using Pd single-atoms successfully presented Pd dispersion in the sub-nanometer scale. Electrochemical results showed that the onset potential taken at 0.1 mA cm^{-2} variated from 0.527 V vs. RHE when the material was supported on the carbon obtained from sewage sludge until 0.700 V vs. RHE for the single atoms dispersed over the carbon obtained from tannin-cellulose biomasses. Selectivity across all materials ranged from 80-90%, slightly lower than Pd over Vulcan carbon.[3] In conclusion, carbon materials obtained from waste resources exhibit promise as support for metals in H_2O_2 electrogeneration, enhancing the sustainability and environmental friendliness of such applications. While the materials in this study demonstrated overall positive results, the synthesis route for this application is in its early stages, necessitating further optimization. The use of lower temperatures (400-900 ºC, holding time max 2h), lower concentration of activating agents (0-20 %m/V), and utilizing near-zero-cost residues make this synthesis route highly attractive, holding potential for significant attention in the coming years.

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