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Towards state-of-the-art environmentally friendly catalysts for H_2O_2 electrogeneration: feasibility of using biomass-derived activated carbons as support for Pd-single atoms

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Hydrogen peroxide (H_2O_2) is a versatile compound with a wide range of industrial, environmental, and medical applications, including water treatment, disinfection, bleaching, and as an oxidant in chemical synthesis. Traditionally, H_2O_2 is produced through the anthraquinone process, which involves complex chemical reactions and significant energy consumption. However, this centralized production method poses challenges for transportation and storage, especially in applications requiring small quantities of H_2O_2, such as environmental remediation.[1] While in situ electrogeneration of H_2O_2 presents an attractive alternative for decentralized applications, current electrodes rely on commercially available amorphous carbons (e.g., Printex 6L and Vulcan XC 72R) derived from petroleum products, requiring high-temperature production (>2000°C) and generating significant pollutants. This study aims to develop and evaluate sustainable alternatives to petroleum-based carbon supports by investigating biomass-derived activated carbons for single-atom catalyst modifications in H_2O_2 electrogeneration. Our objectives include: (1) comparing the performance of different biomass-derived carbons as catalyst supports, (2) optimizing single-atom palladium incorporation, and (3) assessing the technical viability of these materials as replacements for commercial carbons. We investigated three distinct biomass feedstocks, each offering unique advantages: i) sewage sludge: abundant waste stream with high mineral content; ii) sugarcane bagasse: readily available agricultural residue with high carbon content; and iii) tannin-cellulose: naturally functionalized precursor with controlled composition. The materials were synthesized using optimized low-temperature activation protocols (<800°C) and modified according to Choi et al.'s[2] wet impregnation method with $Pd(acac)_2$ to achieve palladium single-atom dispersion, followed by characterization through HAADF-STEM imaging, XPS, and electrochemical techniques; key findings indicate successful Pd single-atom incorporation across all carbon supports, with sub-nanometer atomic dispersion (0.5-0.8 nm), onset potentials of 0.53-0.70 V vs. RHE (depending on feedstock type), H_2O_2 selectivity ranging from 80-90%, close to commercial benchmark materials (92%), and stability showing over 80% activity retention after 24 hours, with performance variations attributed to inherent feedstock properties—sewage sludge exhibited the lowest onset potential due to high mineral content, sugarcane bagasse achieved the highest stability due to optimal porosity, and tannin-cellulose provided the best Pd dispersion due to its surface functionality—together demonstrating that these biomass-derived activated carbons offer promising potential as sustainable alternatives to commercial amorphous carbons in electrochemical H_2O_2 production, with future work focusing on scaling up production while maintaining performance metrics and exploring additional biomass sources for improved sustainability.

Keywords*: biomass-derived carbon, single-atom catalysts, hydrogen peroxide, electrogeneration, sustainable materials*

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