Upcycling Spent Catalysts into Novel Metal-doped Carbon Catalysts via Electrospray Deposition

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Renewable hydrogen production is gaining traction as a key solution in the shift to a low-carbon energy system, with alternatives like biomass reforming and water electrolysis gradually replacing fossil-based hydrogen sources. Fuel cells, which efficiently convert hydrogen into electricity, are also emerging as critical clean energy technologies. In this context, carbon-based materials are considered promising catalyst supports due to their low cost, high surface area, and porous structure, which provides abundant active sites. Functionalization with noble metals, such as platinum, further enhances their catalytic performance. Aligned with circular economy principles, urban mining – particularly the recovery of platinum from spent automotive catalysts – offers a sustainable and cost-effective alternative to traditional extraction methods. This work, developed within the PRIN PNRR Project DURABLE, presents a novel approach to fabricating metal-doped carbon films by integrating noble metal recycling from waste materials with scalable electrospray deposition techniques.

Precious metals are extracted from spent catalysts using mild hydrometallurgical leaching (*e.g.*, HCl, H_2O_2, $CuCl_2$/NaCl) and subsequently adsorbed onto activated carbon. These metal-loaded materials are used directly, without further purification, to formulate inks for electrospray deposition.

Electrospray deposition enables homogeneous and precise thin-film fabrication by atomizing liquid suspensions under high electric fields. In this study, various catalytic ink formulations are investigated, including both commercial metal precursors and noble metals recovered through urban mining. Process parameters such as needle-to-collector distance, liquid flow rate, and applied voltage are systematically varied to assess their impact on the stability of the cone-jet mode, which is essential for consistent and reproducible deposition.

The resulting films are characterized in terms of morphology and surface properties, with a focus on their potential application in liquid-phase catalytic reactors. The porous structure and functionalized surfaces exhibit promising features in terms of sensitivity, selectivity, and reusability. Comparative analyses with commercially available doped carbons are also carried out to evaluate the advantages and limitations of the recovery-based approach. Overall, the study outlines a sustainable pathway for converting secondary resources into high-value thin-film materials, with potential applications in hydrogen generation and conversion systems.

Keywords: *electrospray deposition, activated carbon, noble metal recovery, urban mining, catalysis*