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## Carbon from waste biomass to synthesize Fe-N-C electrocatalysts for the oxygen reduction reaction

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Fuel cells are clean and efficient energy devices able to harvest electric energy from the chemical reaction of a fuel and oxygen without any byproduct. Up to now the most widely used electrocatalysts for the cathodic oxygen reduction reaction (ORR) in polymer electrolyte fuel cells (PEFC) make use of platinum or other noble metals to favor this sluggish reaction, but they face problems, such as expensive price, scarcity and geopolitical concerns correlated with the location of the main producers. Great efforts are being made in the design and development of low cost and stable ORR electrocatalysts not containing platinum group metals (PGM) but still retaining a good activity in alkaline or acidic environment.

In the last decades the usage of transition metals electrocatalysts such as Fe-N-C have been proposed. They can reach electrocatalytic activities similar to those of PGM exploiting a process of heteroatom doping (mainly nitrogen-doped catalyst), although the mechanism of reaction and active centers are still object of debate. The study of N-doped porous carbon materials has become an interesting topic because of their low cost, non-toxicity and renewability, displaying a promising performance as ORR electrocatalysts. A specific niche is being taken by biomass-derived materials, especially from waste, that have been considered for supercapacitors, metal-air battery, and fuel cells applications. The already low cost of PGM-free materials is even lowered if the starting precursors are a common and abundant waste. Moreover, this pathway of valorizing waste into valuable resources and products fits very well in the view of circular economy.

Recently reported biomass derived materials with good activity were obtained from coconuts shells, eggplants, soybeans, or other biomass sources, but they are only the tip of the iceberg, as a multitude of materials have been synthesized, sometimes with more provocative value than real scientific interest. Purpose of this study is the engineering of the biomass carbon structure and incorporation of iron active sites in the material with ball milling method. The starting biomass material, pyrolyzed spent tea leaves and coffee grounds, presents a macro-porous structure. Hence, the porous network is increased by activation with CO<sub>2</sub> or urea in order to artificially create and tune the pores. Tea and coffee are very common wastes commonly used by many families as a drink for breakfast or afternoon break. This implies that there is abundance of spent product that normally is thrown away without being recycled or reutilized. Thus, waste biomass could find a second life as a carbon source to be a precursor for ORR electrocatalysts.

The best results have been obtained by ball milling of activated biochar and Fe(II) phthalocyanine. The biochar used as a carbon support was produced from pyrolysis of waste tea leaves at 1500 °C in argon

atmosphere, then activated with CO<sub>2</sub> or urea. FE-SEM, HR-TEM, XPS, and Raman analyses were performed to investigate the morphology and the physicochemical properties of the electrocatalysts. The ORR activity and methanol tolerance of the Fe-N-C electrocatalysts were tested in rotating ring disk electrode (RRDE), showing promising results in terms of mass activity, onset and half-wave potential in an alkaline environment. Two different short potential cycling protocols demonstrated the high stability of these Fe-N-C electrocatalysts, especially when compared with a 20 wt. % commercial Pt/C electrocatalyst.