A novel synthesis approach towards Fe-based non-noble metal oxygen reduction catalysts with finely tunable composition

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Due to their high power-to-weight ratio, their emission-free operation and their fast start up time, polymer electrolyte fuel cells are excellently suited energy conversion devices for the electric-automotive sector [1]. However, the sluggish kinetics of the oxygen reduction reaction at the cells' cathode limit their overall efficiency. Currently, platinum is used to catalyze said reaction, but its high price motivates the search for inexpensive alternatives. Non-noble metal catalysts (NNMCs) represent a route towards greatly reducing the catalyst cost and therefore the price of the overall stack and have recently been shown to be a viable replacement in terms of initial activity [2,3].

Despite these advancements, NNMC widespread application is hindered by the catalysts' instability. This is believed to be partially caused by the materials' inhomogeneous composition, which not only consists of active sites (typically pictures as an N-coordinated Fe^{II}-ion), but also of inactive side phases (e.g. iron carbides or oxides). The latter could catalyze side reactions and possibly contribute to the depletion of ORR-active sites and membrane poisoning [4].

Looking for approaches to minimize the content of these instability-inducing side phases, in this work we present a new pyrolysis-based synthesis pathway for the preparation of Fe-based NNMCs in which polyacrylonitrile is used as a C- and N-precursor, a Fe-phenanthroline complex as a metal source and a thermally decomposable compound as a pore inducing agent. The latter is chosen as to offer porosity control while going hand in hand with the need for inexpensive catalyst production (as compared to approaches based on the use of costly metalorganic frameworks [3]). The influence of different synthesis parameters is studied by the means of electrochemical activity measurements as well as surface- and bulk-property characterization techniques. We show how the catalytic activity as well as the catalysts' composition are not only determined by the chosen content of pore inducing agent and the final iron content, but also by the choice of pyrolysis temperature and atmosphere enabling improvement and fine-tuning of these crucial Features.

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