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## Aerogel-Derived Fe-N-C Catalysts for Oxygen Electro-Reduction. Linking Their Pore Structure and PEMFC Performance

Frédéric Jaouen, Hongxin Ge, Kavita Kumar, Nicolas Bibent, Frédéric Maillard, Laetitia Dubau Sandrine Berthon-Fabry,

### Abstract :

Iron-based catalysts are intensely investigated for application at the cathode of proton exchange membrane (PEM) and, more recently, of anion exchange membrane fuel cells (AEMFC). Besides optimising their kinetic activity toward the oxygen reduction reaction (ORR), the design and control of their pore structure plays a key role in achieving the highest possible performance in fuel cell. Fe-N-C catalysts derived from the metal-organic framework ZIF-8 are highly active but also mainly microporous, which can lead to mass-transport issues. The silica templating approach typically leads to slightly less active Fe-N-C catalysts, but featuring a combination of micropores and mesopores, which can facilitate mass-transport. Aerogel methods for the synthesis of Fe-N-C catalysts with a hierarchical pore size distribution has been hitherto under-investigated. In contrast to the silica templating method, it is more environmental friendly since no dissolution of a template is needed (no HF). However, highly active Fe-N-C catalysts prepared by aerogel method have not yet been reported in PEM system (even if promising results were recently obtained in rotating disk electrode [1-2]), and it is therefore also unclear if the hierarchical pore size distribution derived from this method is beneficial for improving the mass-transport properties of Fe-N-C cathodes.

This presentation will first report the synthesis method and the key parameters investigated, such as the ratio of melamine to resorcinol, the nature and amount of the iron precursor, the addition of an additional N-rich ligand, on their ORR activity. The materials were characterized for their texture (N<sub>2</sub> sorption, Hg porosimetry), Fe coordination (X-ray absorption and Mössbauer spectroscopy) and surface chemistry (XPS) and morphology. The most promising materials were investigated in PEMFC, for their initial power performance and durability. The results show that, by playing with these synthesis parameters, the aerogel method can yield highly active ORR catalysts (Figure 1), and can give a control on their hierarchical porosity, especially in tuning the mesoporous volume and the average pore width of mesopores. Most catalysts display an atomic dispersion of Fe, explaining their high ORR activity. The presentation will particularly discuss the link between the pore size distribution in this set of materials (micro, meso and macropores), and the mass-transport properties in PEMFC, under pure O<sub>2</sub> or air at the cathode. Their durability will also be discussed, from accelerated stress tests performed in rotating disk electrode and/or PEMFC.

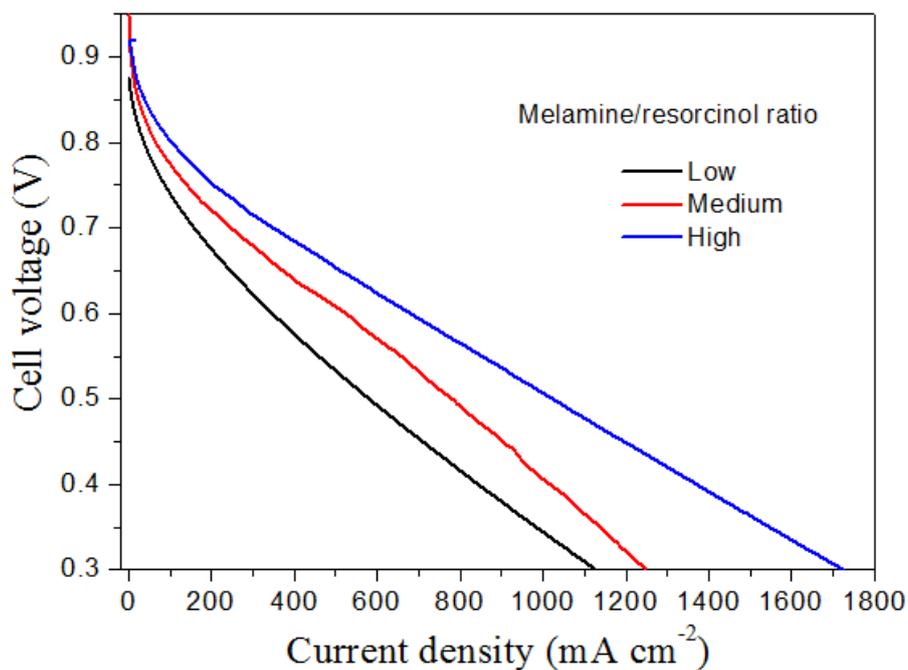


Figure 1. Effect of melamine/resorcinol ratio on fuel cell performance of Fe-N-C catalysts prepared by aerogel method.

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#### References:

- [1] Youling Wang, Sandrine Berthon-Fabry, One-pot synthesis of Fe-N-containing carbon aerogel for oxygen reduction reaction, *Electrocatalysis* 12 (2021) 78-90
- [2] Youling Wang, Mikkel J. Larsen, Sergio Rojas, Moulay-Tahar Sougrati, Frédéric Jaouen, Pilar Ferrer, Diego Gianolio, Sandrine Berthon-Fabry, Influence of the synthesis parameters on the Proton Exchange Membrane Fuel Cells performance of Fe-N-C aerogel catalysts, *Journal of Power Sources* 514 (2021) 230561