# Fe-, N-doped Graphitic Mesoporous Carbon Materials as Oxygen Reduction Electrocatalysts for Alkaline Fuel Cells

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Nitrogen-doped graphitic carbon materials have been reported to exhibit electrocatalytic activities and possibly replace expensive precious metal catalysts (e.g., platinum in the cathodes of fuel cells).[1] N-doped graphitic carbon materials are typically prepared either by pyrolysis[2] or by chemical vapor deposition (CVD)[3] of N-containing precursors in presence of transition metal catalysts. The former usually yields relatively poor graphitic properties, while CVD methods often result in significant flow losses of precursors and therefore low yields. Recently, macrocyclic compounds[4] or ionic liquids[5] have been showEn to yield highly graphitic N-doped carbon materials after pyrolysis, though pyrolyzing such expensive precursor materials is generally not attractive.

We have developed a facile synthesis method to prepare N-containing mesoporous carbon materials with highly graphitic properties based on pyrolyzing inexpensive N-containing organic precursors in the presence of transition metal salts. The resultant materials show highly graphitic properties as established by X-ray diffraction and high-resolution transmission electron microscopy, along with high BET surface areas of ~800 $m^2/g$. Figure 1(a) shows a representative XRD pattern of N-doped mesoporous carbon materials that exhibits a strong reflection at 26.2°, which corresponds to a *d*-spacing of 0.34 nm associated with the inter-layer distance separating graphite planes that are clearly visible in the HR-TEM image of Figure 1(b).

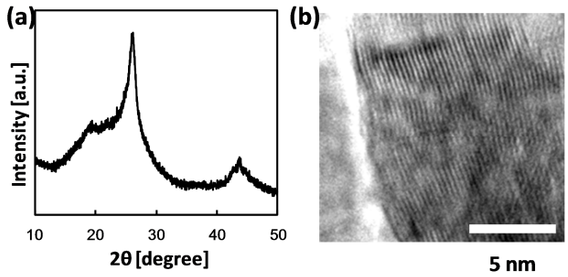


**Figure 1**. (a) X-ray diffraction pattern and (b) high-resolution TEM image of N-doped mesoporous carbon materials

These transition-metal-derived N-doped graphitic mesoporous carbon materials exhibit excellent oxygen reduction activities, especially under alkaline conditions. Oxygen reduction electrochemical activities were evaluated by using linear sweep voltammetry with 0.1 M NaOH solution as the electrolyte and subsequently compared with commercially available carbon-supported platinum catalysts (20 wt% Pt/C). Polarization curves measured with a rotating disk electrode exhibit a high onset potential of 1.046 V vs. RHE for N-doped graphitic mesoporous carbon catalysts, which is comparable to that measured for a Pt/C catalyst (1.04 V). In addition, N-doped mesoporous carbon catalysts exhibit a half-wave potential of 0.89 V vs. RHE, which is higher than Pt/C (0.85 V). N-doped graphitic mesoporous carbon catalysts also show superior stability and methanol tolerance compared to Pt/C catalysts. These results indicate that N-doped graphitic mesoporous carbon materials may be potentially cost-effective electrocatalysts for fuel cells and related devices such as metal-air batteries.

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## References

1. Gong, K.; Du, F.; Xia, Z.; Durstock, M.; Dai, L., *Science* **2009,** *323* (5915), 760-764.
2. Fulvio, P. F.; Jaroniec, M.; Liang, C.; Dai, S., *The Journal of Physical Chemistry C* **2008,** *112* (34), 13126-13133.
3. Xia, Y.; Mokaya, R., *Advanced Materials* **2004,** *16* (17), 1553-1558.
4. Liu, R.; Wu, D.; Feng, X.; Müllen, K., *Angewandte Chemie International Edition* **2010,** *49* (14), 2565-2569.
5. Paraknowitsch, J. P.; Zhang, J.; Su, D.; Thomas, A.; Antonietti, M., *Advanced Materials* **2010,** *22* (1), 87-92.