

Carbon nanotubes for chemical energy conversion and storage

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Carbon nanotubes (CNTs) are promising materials for numerous applications because of their unique mechanical, chemical, and electrochemical properties. Various carbon materials such as carbon fibers or carbon cloth can be used as support for the growth of CNTs. As highly effective surface treatments, the localized etching by water vapor and the exposure to nitric acid vapor are used to enhance the number of surface defects. The iron catalyst for the nanotube growth was synthesized by wetting with iron nitrate, by chemical vapor deposition (CVD) of ferrocene, or by electrochemical iron deposition. Carbon nanotubes were subsequently grown on the iron-loaded fibers by pyrolysis of methane or cyclohexane. A highly dispersed, hierarchically structured catalyst support was obtained in this way.

Nitrogen-doped carbon materials have been used as metal-free electrocatalysts in the oxygen reduction reaction (ORR). Some of the N-doped carbon catalysts show remarkable activities in the ORR especially under alkaline conditions, where the activities are often assigned to certain nitrogen groups and surface defects on the carbon surface. To elucidate the electrocatalytic mechanisms, a major challenge is the controlled synthesis of specific nitrogen groups on carbon surfaces. Different methods were employed for the synthesis of nitrogen-doped carbon materials: (a) Catalytic growth of N-doped CNTs from N-containing organic precursors; (b) Post-treatment of oxygen-functionalized CNTs under ammonia atmosphere at elevated temperatures; (c) Post-treatment of oxygen-functionalized CNTs by pyrolyzing N-containing organic precursors at elevated temperatures; (d) Pyrolysis of N-containing polymers. The effect of these methods on the electrocatalytic properties will be discussed.

Fischer-Tropsch synthesis (FTS) is a well-established industrial process for converting synthesis gas derived from coal, natural gas or biomass over iron or cobalt catalysts into mainly linear hydrocarbons exhibiting a broad chain-length distribution. In particular, high synthesis temperatures and Fe-based catalysts are essential for short-chain α -olefins. Fe catalysts supported on active carbon were reported to have a higher activity per unit volume and higher olefin selectivity compared to unsupported catalysts. In my talk, I will report on Fe nanoparticles deposited on oxygen-functionalized CNTs (OCNTs) and on nitrogen-functionalized CNTs (NCNTs). Catalytic testing under industrially relevant conditions was applied to demonstrate the unique properties of OCNTs and NCNTs for the Fe-catalyzed high-temperature FTS. The obtained catalysts showed excellent olefin selectivities, moderate methanation tendency, low growth probabilities and good stabilities. In addition, the Fe/CNT catalysts were applied in CO₂ hydrogenation.

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