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Large-Scale Synthesis of High-Quality Zeolite-Templated Carbons without Depositing External Carbon Layers

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Abstract

Zeolite-templated carbons (ZTCs) possess extra-large surface areas ($\geq 2000 \text{ m}^2 \text{ g}^{-1}$) and micropore volumes ($\geq 1.0 \text{ cm}^3 \text{ g}^{-1}$), which allow superior electrochemical performances and gas adsorption properties compared with conventional activated carbons. Unfortunately, ZTCs have been mostly synthesized at a small laboratory-scale due to the limited reproducibility of carbon structures at a large-scale synthesis. The biggest challenge in the carbon replication of zeolites is the deposition of dense carbon layers at the external surface of zeolite crystallites before the complete zeolite micropore filling with carbon. The formation of external carbon layers hinders a faithful carbon replication of zeolite structure, and also significantly limits the adsorption properties of the resultant template-free ZTCs. Here, we demonstrate that acetylene chemical vapor deposition (CVD) at a relatively mild temperature (823 K), followed by thermal treatment at 1073 K, can selectively deposit carbons in the micropores of zeolites, regardless of the synthesis batch size. By combining the mild CVD condition and the use of a rotary tubular furnace, which allows the continuous agitation of zeolites during the synthesis, 50 g NaX and BEA zeolite could be faithfully replicated to ZTCs with a high BET surface area ($> 2700 \text{ m}^2 \text{ g}^{-1}$) and micropore volume ($> 1.10 \text{ cm}^3 \text{ g}^{-1}$) without depositing nonporous external carbon layers that significantly hinder the bulky molecular diffusion.

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