Predefinition and Activation of Nanographitic Domains in Polymer Derived Carbons for Obtaining Active Dehydrogenation Catalysts

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Abstract

The field of carbon catalysed oxidative dehydrogenations has been developed within the context of the exploration of carbon nanomaterials (e.g. carbon nanotubes, carbon nanofibers, nanodiamond etc.). The high redox activity of carbon nanomaterials is based predominantly on the presence of large conjugated (graphitic) domains in combination with a high density of defect sites (e.g. edges, in-plane defects) which host oxygen surface groups as active centers. However, because of intrinsic drawbacks such as the high pressure drop of fixed nanocarbon catalyst beds combined with high porosities and thus low space time yields, alternatives to carbon nanomaterials as catalysts are of high interest.

In this context, catalytic graphitization of a polymer precursor with transition metals (e.g. Ni, Co, Fe) was employed to prepare hybrid amorphous/graphitic carbons. Catalytic graphitization offers excellent control of the crystallite size by carefully choosing the graphitization catalyst, catalyst loading and graphitization temperature. The active carbon dehydrogenation catalyst is prepared by removing the amorphous parts of the hybrid carbon materials via selective oxidation. This method produces macro-/mesoporous carbonaceous materials that exhibit size controlled nanographitic domains, while still showing a macroscoping shape (approx. 100 µm). These materials display a high density of structural defects combined with graphitic domains and can host redox-active sites. The materials were tested as catalysts in the oxidative dehydrogenation of ethanol to acetaldehyde and showed good activity and selectivity.

Topics

Carbons for Catalysis