

Assessing the Catalytic Functions of Hydrogen Spillover with Pt-Encapsulated Aluminosilicates Having Controlled Nanostructures

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Hydrogen spillover has been studied for several decades, but its nature, catalytic functions, and even its existence remain topics of vigorous debate. Here, we prepared Pt-encapsulating aluminosilicates with various surface areas and surface hydroxyls in a controlled manner to elucidate the catalytic consequences of them. Pt encapsulation in NaA micropore was attained by using a mercaptosilane-assisted metal encapsulation method. To control the zeolite crystallite size, varied amount of polyethyleneglycol was added in the zeolite synthesis gel. Samples were decationized by ion-exchange with NH_4^+ and subsequent heat treatment. Decationization led to a significant loss of zeolite crystallinity generating controlled amount of surface hydroxyl groups which was verified by ^{27}Al and ^{29}Si MAS NMR. Catalytic results and DFT calculations showed that surface hydroxyls, presumably Brsted acids, are crucial for catalytic use of spillover hydrogen. The spillover catalysts showed very high activities in hydro-/dehydrogenation, but virtually zero activities in hydrogenolysis.