571g Characterization of Dendrimer-Derived Supported Metal Nanoparticles

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We have been investigating the utilization of dendrimer-metal nanocomposites for the preparation of heterogeneous catalysts with controlled particle sizes. Poly(amido)amine (PAMAM) Starburst dendrimers are hyperbranched polymers that contain amine and amide functionalities arranged in regular patterns, and have the ability to complex metal cations into their branches. Commercially available PAMAM dendrimers were used to prepare and dendrimer-metal nanocomposites in solution, incorporating different noble metals (i.e., Pt, Pd, Rh, Au) and combinations thereof. These metal-dendrimer nanocomposites were then supported onto commercial and model supports via incipient wetness impregnation and/or spontaneous adsorption processes. The resulting materials were thermally treated under various oxidative and reductive protocols to remove the dendrimer “shell” and obtain supported mono- and bimetallic catalysts. The different steps of these processes both during the liquid phase synthesis and the subsequent impregnation and catalyst activation processes were monitored in situ via EXAFS and FTIR spectroscopies. The use of these techniques allows for monitoring of the changes in the nature and environment of the metal species involved (i.e., oxidation state and coordination number), as well as changes of the dendrimer's functional groups during metal complexation and dendrimer decomposition/desorption. The presence and type of incorporated metal, as well as the nature of support used, further influences the pathway and rate of these processes. Finally, the metal nanoparticles formed were imaged at different stages (i.e., in solution and on the support prior and following the removal of the dendrimer “shell”) via high resolution transmission electron microscopy. Overall, our results suggest that in most cases dendrimer stabilization allows for compositional and structural control greater than that of traditionally prepared mono- and bi-metallic catalysts.