Gold-catalyzed aerobic oxidation of dibenzylamine: Homogeneous or heterogeneous catalysis?

Au(OAc)(3) is applied as an effective catalyst of the selective oxidation of dibenzylamine to dibenzylimine using molecular oxygen as the only oxidant. When Au(OAc)(3) was preadsorbed onto CeO2, the supported catalyst was more active than any homogeneous or heterogeneous catalyst known for this reaction. Although, some fascinating color changes in the early stage of the reaction indicated the formation of an amine complex, conventional filtration experiments proved the heterogeneity of the system. The fate of the active gold component was studied by in situ X-ray absorption spectroscopy (XANES) using a specially designed cell. These investigations revealed that in the early stage of the reaction Au(OAc)(3) is dissolved and subsequently reduced by the amine and the in situ formed gold nanoparticles are the real active species of the reaction. Formation of gold nanoparticles during dibenzylamine oxidation was proved independently by transmission electron microscopy. Our findings lead to a simple synthetic procedure using a commercially available gold salt, which upon interaction with the amine forms highly active and selective gold nanoparticles. (C) 2008 Elsevier B.V. All rights reserved.