

Highly efficient hydrogenation of furfural to valuable products catalysed by Rh (III), Pd (II), Ni (II), Fe (II) and Ru (II) half-sandwich picolinamide and the SBA-15 supported molecular complexes as pre-catalysts

Pamela S. Moyo Gift Mehlana Leah C. Matsinha Banothile C. E. Makhubela

Abstract

Novel palladium, rhodium, iron, ruthenium and nickel half-sandwich picolinamide complexes have been prepared and characterised in detail using high-resolution electrospray ionisation mass spectrometry (HR-ESI-MS), multi-dimensional nuclear magnetic resonance spectroscopy, elemental analysis (EA), Fourier transform-infrared spectroscopy (FT-IR), and single crystal X-ray diffraction (SCXRD). All the systems proved to be active with good catalytic performance for the hydrogenation of furfural. Among the pre-catalyst, C2 (Rh metal centre) displayed complete conversion (100%). Pre-catalyst C5 (Ru metal centre) also resulted in excellent selectivity to EL (51%) and FA (49%). All the pre-catalysts with early transition metal centres (Ni, Fe) gave selectivity towards FA (100%) under optimal conditions. Possible pathways for the formation of catalytically active have been proposed based on in situ NMR studies obtained using C2. Successful modification of molecular complexes (C2 and C3) on SBA-15 resulted in supported molecular complexes (C2@SBA-15 and C3@SBA-15). Furthermore, the supported molecular catalysts were characterised using inductively coupled plasma-optical emission spectroscopy (ICP-OES), Fourier-transform infrared spectroscopy (FT-IR), powder X-ray diffraction (PXRD), BET surface area, selected area electron diffraction (SAED), X-ray photoelectron spectroscopy (XPS) and high-resolution transmission electron microscopy (HR-TEM). The supported molecular complex C2@SBA-15 was recycled five times (with consistent activity and selectivity).

KEYWORDS

ethyl levulinate, furfural, furfuryl alcohol, supported molecular rhodium catalysts