P57 : Dawson-type polyoxometalates in Liquid phase isobutane hydroxylation

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Abstract:

Dawson polyoxometalates (POMs) have been reported as efficient catalysts for gasphase oxidative dehydrogenation of isobutane to isobutene [1] and isobutyric acid to methacrylic acid with oxygen molecular as oxidant [2]. In this paper, we have studied the catalytic activity of a series of Dawson-type POMs, as potassium salts, namely K₆P₂W₁₂Mo₆O₆₂, K₇P₂Mo₅VW₁₂O₆₂, P₂W₁₀Mo₅V₃O₆₂ and P₂W₁₂V₆O₆₂, in the isobutane hydroxylation in acetonitrile solvent with hydrogen peroxide as oxidant. POMs were characterized by FTIR, ³¹P NMR and BET. Terbutyl alcohol (tert-BuOH), isobutyl alcohol (iso-BuOH), acetone, isopropyl alcohol (iso-PrOH) and isobutyric acid (isoBuOOH) are the products detected in isobutane hydroxylation at 60°C. The distribution of these products depends of POM composition and the framework symmetry. Thus, V free POM didn't catalyse liquid isobutane oxidation while V containing POMs promoted the formation of oxygenated products at low temperature. The first major product is isobutiric acid and the second one is tertiobutanol. The investigation of isobutanol and tertiobutanol transformation in equivalent reaction conditions (with or without catalyst) evidenced the formation of isobutyric acid and C_3 oxygenated products. This suggests that tertiobutanol and isobutanol would the primary products, while acetone, isopropanol and isobutyric acid would result from successive transformations of the C_4 alcohols. The results shown that the presence of vanadium atoms favoured the formation of oxygenated compounds in the isobutane oxidation. The distribution of reaction products depends on the polyanion composition.

References :

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