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ACID-BASE CATALYZED ACTIVATION OF n-ALKANES
n-Butane conversion on well-defined sulfated zirconia samples (SPP 1091)

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Epitaxial and nanocrystalline zirconia film samples have been developed as model systems to study the surface chemistry and surface reactions of sulfated zirconia (SZ) catalysts. Epitaxial c-ZrO_2 (111) films of the cubic (c) type are prepared on a Pt (111) substrate allowing STM, LEED and AES characterization of surface structure and composition. Exposure to SO_3 yields a $(\sqrt{3} \times \sqrt{3})$ R30° sulfation structure. DFT calculations explain the energetic preference of the $\sqrt{3}$ structure and demonstrate similarity of c-SZ to industrially used tetragonal SZ systems. During reaction with n-butane, the $(\sqrt{3} \times \sqrt{3})$ R30° sulfation structure is stable and its chemical activity is indicated by the formation of surface carbon. DFT calculations also show that sulfation of t-ZrO_2 (101) with H_2SO_4 pyrosulfate in agreement with IR-experiments. This pyrosulfate can easily provide a very reactive, 'labile' SO_3 species promoting conversion of n-alkanes. Nanocrystalline films allow application of TEM, SEM and surface science techniques as XPS while exhibiting powder characteristics such as multiple facets and potential for bulk and surface defects. The films exhibit properties corresponding to those of typical powder catalysts. Tetragonal nanocrystals, surface composition, weak n-butane, and reactive ammonia interaction indicate they are suitable SZ model catalysts. Isobar measurements show n-butane adsorption to be promoted on SZ thin films in comparison to oxidized silicon wafers.