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ACID-BASE CATALYZED ACTIVATION OF n-ALKANES
Isomerization of n-butane (SPP 1091)

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Sulfated zirconia catalysts show a high activity for the isomerization of hydrocarbons even at low temperatures. To elucidate the nature of active centers and the mechanism of alkane activation on sulfated zirconias a network of three working clusters with groups from heterogeneous catalysis, surface science and theory was built up. The combination of methods (DRIFTS, XRD, XPS, Temporal Analysis of Products (TAP), LEED, DFT catalytic experiments) provided the opportunity to bridge the materials and pressure gap for this important catalytic system. One of the main results was the identification of the pyrosulfate group as the active species on the surface to start the catalytic reaction by formation of butene. Theoretical calculations supported not only the interpretation of DRIFT spectra but were also able to characterize different sulfate species on the surface of tetragonal zirconia surfaces. Experiments under deactivating conditions (DRIFTS, TAP) showed a strong correlation of catalytic activity and sulfate structure. The kinetic experiments allowed to set up a model for the reaction mechanism, which includes the activation of n-butane on pyrosulfate groups via an oxidative dehydrogenation and the propagation via an intramolecular rearrangement. Moreover, a new preparation technique allowed gas phase sulfation and could be shown as a posibility to recover the catalytic activity of deactivated sulfated zirconias.