

Abstract Submitted
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Low-temperature dehydrogenation catalysis of isophorone ($C_9H_{14}O$) on Pd(111) XINGUO REN, WEI LIU, ADITYA SAVARA, WIEBKE LUDWIG, KARLHEINZ DOSTERT, SWETLANA SCHAUERMANN, ALEXANDRE TKATCHENKO, HANS-JOACHIM FREUND, MATTHIAS SCHEFFLER, Fritz-Haber-Institut der MPG, Berlin, Germany — Selective hydrogenation and dehydrogenation of hydrocarbons is one of the most extensively used processes in catalysis. In particular, it is important to design catalysis which can occur at or below room temperature. Here, we investigate the adsorption and dehydrogenation of a model organic/metal catalyst: isophorone ($C_9H_{14}O$) on the Pd(111) surface. Density-functional theory (DFT) calculations including van der Waals (vdW) interactions are carried out to elucidate the adsorbate geometry and reaction dynamics. The vdW interactions dramatically modify the potential-energy surface and enhance the binding energy by about 1 eV. The combination of our theoretical results with the analysis of infrared (IR) spectra and temperature-programmed desorption (TPD) experiments leads us to propose a dehydrogenation pathway from the weakly chemisorbed reactant ($C_9H_{14}O$) to the strongly chemisorbed product ($C_9H_{10}O$), which occurs after four C-H bond cleavages. The obtained low dehydrogenation temperature (130-150 K) from PBE+vdW barriers, including zero-point energy vibrations, is consistent with the TPD analysis.

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