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Steps in hydrogen production from methanol on sub-nanometer palladium clusters FAISAL MEHMOOD[1], JEFFREY P. GREELEY[2], PETER ZAPOL[1], LARRY A. CURTISS[1,2], [1]Material Science Division and [2]Center of Nanoscale Materials, Argonne National Laboratory — Extensive experimental and theoretical work has been done to understand the decomposition of methanol on various metal and metal oxide nanoparticles for hydrogen production. The activity of sub-nanometer sized particles $<$ 1nm however is not very well known, primarily because of technical challenges involved in preparation and stabilization of the clusters. To explore the properties of the Pd clusters computationally, we have carried out density functional calculations for the methanol decomposition reaction on Pd_4 and Pd_8 clusters. The thermodynamics and kinetics of three decomposition routes involving C–O, C–H and O–H scission were investigated; activation energy barriers were determined with the nudged elastic band method. A detailed analysis of the PES for methanol decomposition shows C–O activation to be the least favorable step. In addition, all possible reaction paths for the Pd_4 cluster are much lower in comparison to single crystal surface and large nanoparticles. To understand how particle size affects the elementary reaction steps, we also present a comparison of methanol decomposition on Pd_4 with Pd_8 clusters. Finally, we will discuss the implication of a linear correlation between the transition state and final state energies that is followed for all elementary reaction steps on Pd_4 and Pd_8 clusters.

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