

Adsorption of multiple H₂ molecules on Pd₃ and Pd₄ clusters. A density functional study.

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The reaction/adsorption of multiple H₂ molecules on Pd_{*n*} clusters with *n* = 3 and 4 were studied using the density functional theory. It has been shown that the activation of the first H₂ molecule by Pd₃ and Pd₄ takes place without or with a small energetic barrier (based on Δ*H* (298.15 K)) and leads to the formation of Pd₃(H)₂ and Pd₄(H)₂ complexes, respectively, where the H–H bond is broken. For the Pd₄(H)₂ complex, various isomeric structures were found in the singlet and triplet states, among which the singlet **Pd4_1_c(e,e')**, with the two H ligands bridging the Pd–Pd edges not sharing the Pd atom, is found to be the most favorable. Unlike the first H₂ addition reaction, dissociative adsorption of the second H₂ molecule, reactions Pd₃(H)₂ + H₂ → Pd₃(H)₄ and Pd₄(H)₂ + H₂ → Pd₄(H)₄, appeared to be thermodynamically and kinetically unfavorable. Instead, molecular adsorption of additional *m*H₂ molecules onto the “naked” Pd centers of the Pd₃(H)₂ and Pd₄(H)₂ complexes was shown to be feasible, which is in good agreement with the experimental Pd_{*n*} + D₂ saturation studies. The thermodynamic stabilities of the resulting Pd₃(H)₂(H₂)_{*m*} (*m* = 1–3) and Pd₄(H)₂(H₂)_{*m*} (*m* = 1–4) species were discussed in terms of the Δ*H* and Δ*G* values estimated at *T* = 298.15 and 70 K.

Słowa kluczowe

Molecular structure, Palladium, Cluster chemistry, Chemical structure

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