

C–H bond activation by the excited zinc atom: gas-phase formation of methylzinc hydride (HZnCH₃) based on multireference second-order perturbation theory and coupled cluster calculations.

Autorzy

Jerzy Moc

Rok wydania

2021

Czasopismo

ACS Omega

Numer woluminu

6

Strony

24280-24288

DOI

10.1021/acsomega.1c04531

Kolekcja

Naukowa

Język

Angielski

Typ publikacji

Artykuł

Streszczenie

The pioneering spectroscopic observations of the methylzinc hydride [HZnCH₃(X1A1)] molecule were reported previously by the Ziurys group [J. Am. Chem. Soc. 2010, 132, 17186–17192], and the possible formation mechanisms were suggested therein, including those with the participation of excited zinc atoms in reaction with methane. Herein, the ground singlet state and the lowest excited triplet state potential energy surfaces of the Zn+CH₄ reaction have been explored using high-level electronic structure calculations with multireference second-order perturbation theory and coupled cluster singles and doubles with perturbative triples (CCSD(T)) methods in conjunction with all-electron basis sets (up to aug-cc-pV5Z) and scalar relativistic effects incorporated via the second-order Douglas–Kroll–Hess (DK) method. Based on the ab initio results, a plausible scenario for the formation of HZnCH₃(X1A1) is proposed involving the activation of the C–H bond of methane by the lowest excited 3P state atomic zinc. Calculations also highlight the importance of an agostic-like Zn...H–C interactions in the pre-activation complex and good agreement between the structure of the HZnCH₃(X1A1) molecule predicted at the DK-CCSD(T)/aug-cc-pVQZ-DK level of theory and that derived from rotational spectroscopy, as well as the discrepancies between the ab initio and density functional theory predictions.

Słowa kluczowe

Zinc, Hydrocarbons, Chemical reactions, Quantum mechanics, Potential energy

Licencja otwartego dostępu

CC-BY

Licencja na prawach której można swobodnie kopiować, rozprowadzać, zmieniać i remiksować objęty prawem autorskim utwór (Utwór-przedmiot prawa autorskiego) pod warunkiem podania imienia i nazwiska autora utworu pierwotnego oraz źródła pochodzenia utworu.

Pełny tekst licencji:

<https://creativecommons.org/licenses/by/3.0/pl/legalcode>

Adres publiczny

<http://dx.doi.org/10.1021/acsomega.1c04531>

Strona internetowa wydawcy

<https://www.acs.org/content/acs/en.html>