Title

Exploring the catalytic potential of AuxPt4-x clusters on TiC and ZrC (001) surfaces for hydrogen dissociation

Abstract

The adsorption and dissociation of hydrogen on bimetallic clusters of AuxPt4-x supported on TiC (0 0 1) and ZrC (0 0 1) surfaces, has been studied using periodic boundary density functional theory (DFT). Simulations reveal that H2 exhibits moderate adsorption energies on AuxPt4-x/TMC (TM = Ti and Zr) systems and dissociates with a tiny barrier comparable to archetypal catalyst such as Pt (0 0 1). The incorporation of two different metal atoms (Au and Pt) in the cluster results in a noticeable enhancement of catalytic activity compared to clusters of equivalent size composed of pure metals like Pd, Cu, and Pt when deposited on TiC (0 0 1). Furthermore, our calculations reveal that the adsorbed H atom on the AuPt3 cluster is prone to spill over the C sites on both surfaces, and the migration of hydrogen atoms on both supports is thermodynamically favorable. In essence, our results provide compelling evidence that when AuxPt4-x clusters are supported on surfaces with a significant degree of polarity, as TMCs, the complete system H2/AuxPt4-x/TMC can efficiently activate and dissociate H2 concurrently, highlighting the potential for enhanced catalytic efficiency in hydrogenation reactions. © 2024

Authors

Gomez T.; Calatayud M.; Arratia-Perez R.; Muñoz F.; Cárdenas C.

Author full names

Gomez, Tatiana (24399021200); Calatayud, Monica (7004728004); Arratia-Perez, Ramiro (6603574282); Muñoz, Francisco (56372188500); Cárdenas, Carlos (55666781200)

Author(s) ID

24399021200; 7004728004; 6603574282; 56372188500; 55666781200

Year

2024

Source title

Applied Surface Science

Volume

657.0

Art. No.

159815

DOI

10.1016/j.apsusc.2024.159815

Link

https://www.scopus.com/inward/record.uri?eid=2-s2.0-85186699653&doi=10.1016%2fj.apsusc.2024.159815&partnerID=40&md5=a31236ba6baf0f7567795116630b016a

Affiliations

Theoretical and Computational Chemistry Center, Institute of Applied Sciences, Faculty of Engineering, Universidad Autonoma de Chile, Avenida Pedro de Valdivia 425, Santiago, Chile; Sorbonne Université, CNRS, Laboratoire de Chimie Théorique, LCT, 4 Place Jussieu, Paris, F-75005, France; Doctorado En Fisicoquímica Molecular, Facultad de Ciencias Exactas, Universidad Andres Bello, Republica 275, Santiago, Chile; Departamento de Física, Facultad de Ciencias, Universidad de Chile, 653-Santiago, Chile; Centro Para El Desarrollo de las Nanociencias Y Nanotecnología, CEDENNA, Av. Ecuador, Santiago, 3493, Chile

Authors with affiliations

Gomez T., Theoretical and Computational Chemistry Center, Institute of Applied Sciences, Faculty of Engineering, Universidad Autonoma de Chile, Avenida Pedro de Valdivia 425, Santiago, Chile; Calatayud M., Sorbonne Université, CNRS, Laboratoire de Chimie Théorique, LCT, 4 Place Jussieu, Paris, F-75005, France; Arratia-Perez R., Doctorado En Fisicoquímica Molecular, Facultad de Ciencias Exactas, Universidad

Andres Bello, Republica 275, Santiago, Chile; Muñoz F., Departamento de Física, Facultad de Ciencias, Universidad de Chile, 653-Santiago, Chile, Centro Para El Desarrollo de Ias Nanociencias Y Nanotecnología, CEDENNA, Av. Ecuador, Santiago, 3493, Chile; Cárdenas C., Departamento de Física, Facultad de Ciencias, Universidad de Chile, 653-Santiago, Chile, Centro Para El Desarrollo de Ias Nanociencias Y Nanotecnología, CEDENNA, Av. Ecuador, Santiago, 3493, Chile

Index Keywords

Atoms; Catalyst activity; Density functional theory; Dissociation; Hydrogen; Hydrogenation; Titanium carbide; Zirconium compounds; Adsorption energies; Bimetallic clusters; Catalytic potential; Density-functional-theory; Equivalent size; Hydrogen dissociation; Metal atoms; Periodic boundaries; Pure metals;]+ catalyst; Binary alloys

Funding Details

Center for the Development of Nanosciences and Nanotechnology; Fondo Nacional de Desarrollo Científico y Tecnológico, FONDECYT, (1220366, 1220715, 1231487, ANID-REDES 190102); Centro para el Desarrollo de la Nanociencia y la Nanotecnología, CEDENNA, (AFB 220001)

Funding Texts

This work was financed by i) FONDECYT through projects N°1220366, 1231487 and 1220715 ii) ANID-REDES 190102 iii) Center for the Development of Nanosciences and Nanotechnology, CEDENNA AFB 220001. Powered@NLHPC: This research was partially supported by the supercomputing infrastructure of the NLHPC (ECM-02).

References

Anand S., Pinheiro D., Sunaja Devi K.R., Recent advances in hydrogenation reactions using bimetallic nanocatalysts: a review, asian, J. Org. Chem., 10, (2021); Okolie J.A., Patra B.R., Mukherjee A., Nanda S., Dalai A.K., Kozinski J.A., Futuristic applications of hydrogen in energy, biorefining, aerospace, pharmaceuticals and metallurgy, Int. J. Hydrogen Energy., 46, (2021); (2023); Shanmugam S., Mathimani T., Rajendran K., Sekar M., Rene E.R., Chi N.T.L., Ngo H.H., Pugazhendhi A., Perspective on the strategies and challenges in hydrogen production from food and food processing wastes, Fuel, 338, (2023); Lende A.B., Bhattacharjee S., Tan C.S., Hydrogenation of polyethylene terephthalate to environmentally friendly polyester over Vulcan XC-72 carbon supported rh-pt bimetallic catalyst, Catal. Today., 388-389, (2022); Huang Z., Yu D., Makuza B., Tian Q., Guo X., Zhang K., Hydrogen reduction of spent lithium-ion battery cathode material for metal recovery: mechanism and kinetics, Front. Chem., 10, (2022); Galashev A.Y., Ivanichkina K.A., Vorob'ev A.S., Rakhmanova O.R., Katin K.P., Maslov M.M., Improved lithium-ion batteries and their communication with hydrogen power, Int. J. Hydrogen Energy., (2021); Pacchioni G., Hydrogen bonding makes organic solar cells stretchy, Nat. Rev. Mater., 7, (2022); Shi Z., Li N., Lu H.K., Chen X., Zheng H., Yuan Y., Ye K.Y., Recent advances in the electrochemical hydrogenation of unsaturated hydrocarbons, Curr. Opin. Electrochem., 28, (2021); Raman R., Nair V.K., Prakash V., Patwardhan A., Nedungadi P., Green-hydrogen research: what have we achieved, and where are we going? bibliometrics analysis, Energy Rep., 8, (2022); Khan M.A., Al-Shankiti I., Ziani A., Idriss H., Demonstration of green hydrogen production using solar energy at 28% efficiency and evaluation of its economic viability, Sustain. Energy Fuels., 5, (2021); Yilanci A., Dincer I., Ozturk H.K., A review on solar-hydrogen/fuel cell hybrid energy systems for stationary applications, Prog. Energy Combust. Sci., 35, (2009); Jeong H., Kim J., Methanol dehydrogenation reaction at AU@PT catalysts: insight into the

methanol electrooxidation, Electrochim. Acta., 283, (2018); Zhang W., Wang H., Jiang J., Sui Z., Zhu Y., Chen D., Zhou X., Size dependence of pt catalysts for propane dehydrogenation: from atomically dispersed to nanoparticles, ACS Catal., 10, (2020); Shan Y., Hu H., Fan X., Zhao Z., Recent progress in catalytic dehydrogenation of propane over pt-based catalysts, Phys. Chem. Chem. Phys., 25, (2023); Al-Nayili A., Majdi H.S., Albayati T.M., Cata Saady N.M., Formic acid dehydrogenation using Noble-metal nanoheterogeneous catalysts: towards sustainable hydrogen-based energy, Catalysts, 12, (2022); Silveri F., Quesne M.G., Vines F., Illas F., Catlow C.R.A., De Leeuw N.H., Catalytic reduction of carbon dioxide on the (001), (011), and (111) surfaces of TiC and ZrC: a computational study, J. Phys. Chem. C., 126, (2022); Hwu H.H., Chen J.G., Surface chemistry of transition metal carbides, Chem. Rev., 105, (2005); Levy R.B., Boudart M.; Rodriguez J.A., Illas F., Activation of noble metals on metal-carbide surfaces: novel catalysts for CO oxidation, desulfurization and hydrogenation reactions, Phys. Chem. Chem. Phys., 14, (2012); Asara G.G., Ricart J.M., Rodriguez J.A., Illas F., Exploring the activity of a novel Au/TiC(001) model catalyst towards CO and CO2 hydrogenation, Surf. Sci., 640, (2015); Rodriguez-Gomez A., Holgado J.P., Caballero A., Cobalt carbide identified as catalytic site for the dehydrogenation of ethanol to acetaldehyde, ACS Catal., 7, (2017); Liu W., Chen B., Duan X., Wu K.H., Qi W., Guo X., Zhang B., Su D., Molybdenum carbide modified nanocarbon catalysts for alkane dehydrogenation reactions, ACS Catal., 7, (2017); Porosoff M.D., Kattel S., Li W., Liu P., Chen J.G., Identifying trends and descriptors for selective CO2 conversion to CO over transition metal carbides, Chem. Commun., 51, (2015); Schweitzer N.M., Schaidle J.A., Ezekoye O.K., Pan X., Linic S., Thompson L.T., High activity carbide supported catalysts for water gas shift, J. Am. Chem. Soc., 133, (2011); Urzhuntsev G.A., Toktarev A.V., Echevskii G.V., Delii I.V., Vlasova E.N., Bukhtiyarova G.A., Prospects for using mo- and W-containing catalysts in hydroisomerization. a patent review. part 1: catalysts based on molybdenum and tungsten phosphides, Catal. Ind., 8,

(2016); Zhang S.B.X.Y., Pessemesse Q., Latsch L., Engel K.M., Stark W.J., van Bavel A.P., Horton A.D., Payard P.A., Coperet C., Role and dynamics of transition metal carbides in methane coupling, Chem. Sci., 14, (2023); Rodriguez J.A., Liu P., Takahashi Y., Nakamura K., Vines F., Illas F., Desulfurization of thiophene on Au/TiC(001): au-C interactions and charge polarization, J. Am. Chem. Soc., 131, (2009); Meyer S., Nikiforov A.V., Petrushina I.M., Kohler K., Christensen E., Jensen J.O., Bjerrum N.J., Transition metal carbides (WC, Mo2C, TaC, NbC) as potential electrocatalysts for the hydrogen evolution reaction (HER) at medium temperatures, Int. J. Hydrogen Energy., 40, (2015); Jun H., Kim S., Lee J., Development strategies in transition metal carbide for hydrogen evolution reaction: a review, Korean J. Chem. Eng., 37, (2020); Prats H., Pinero J.J., Vines F., Bromley S.T., Sayos R., Illas F., Assessing the usefulness of transition metal carbides for hydrogenation reactions, Chem. Commun., 55, (2019); Silveri F., Quesne M.G., Roldan A., De Leeuw N.H., Catlow C.R.A., Hydrogen adsorption on transition metal carbides: a DFT study, Phys. Chem. Chem. Phys., 21, (2019); Posada-Perez S., Vines F., Valero R., Rodriguez J.A., Illas F., Adsorption and dissociation of molecular hydrogen on orthorhombic β-Mo2C and cubic δ-MoC (001) surfaces, Surf. Sci., 656, (2017); Hoang T.K.A., Antonelli D.M., Exploiting the kubas interaction in the design of hydrogen storage materials, Adv. Mater., 21, (2009); Florez E., Gomez T., Liu P., Rodriguez J.A., Illas F., Hydrogenation reactions on Au/TiC(001): effects of au-C interactions on the dissociation of H2, ChemCatChem, 2, (2010); Gomez T., Florez E., Rodriguez J.A., Illas F., Reactivity of transition metals (pd, pt, cu, ag, au) toward molecular hydrogen dissociation: extended surfaces versus particles supported on TiC(001) or small is not always better and large is not always bad, J. Phys. Chem. C., 115, (2011); Toshima N., Yonezawa T., Bimetallic nanoparticles - novel materials for chemical and physical applications, New J. Chem., 22, (1998); Liu L., Corma A., Metal catalysts for heterogeneous catalysis: from single atoms to nanoclusters and nanoparticles, Chem. Rev., 118, (2018); He W., Han X., Jia H., Cai J., Zhou Y., Zheng Z., AuPt alloy

nanostructures with tunable composition and enzyme-like activities for colorimetric detection of bisulfide, Sci. Rep., 7, (2017); Singh A.K., Xu Q., Synergistic catalysis over bimetallic alloy nanoparticles, ChemCatChem., 5, (2013); Mustieles Marin I., Asensio J.M., Chaudret B., Bimetallic nanoparticles associating Noble metals and first-row transition metals in catalysis, ACS Nano., 15, (2021); Perdew J.P., Burke K., Ernzerhof M., Generalized gradient approximation made simple, Phys. Rev. Lett., 77, (1996); Grimme S., Semiempirical GGA-type density functional constructed with a long-range dispersion correction, J. Comput. Chem., 27, (2006); Blochl P.E., Projector augmented-wave method, Phys. Rev. B., 50, (1994); Kresse G., Furthmuller J., Efficiency of ab-initio total energy calculations for metals and semiconductors using a plane-wave basis set, Comput. Mater. Sci., 6, (1996); Kresse G., Furthmuller J., Efficient iterative schemes for ab initio total-energy calculations using a plane-wave basis set, Phys. Rev. B - Condens. Matter Mater. Phys., 54, (1996); Kresse G., Hafner J., Ab initio molecular dynamics for liquid metals, Phys. Rev. B., 47, (1993); Kresse G., Hafner J., Ab initio molecular-dynamics simulation of the liquid-metalamorphous-semiconductor transition in germanium, Phys. Rev. B., 49, (1994); Kresse D., Joubert G.A., (1999); Monkhorst H.J., Pack J.D., Special points for brillouin-zone integrations, Phys. Rev. B., 13, (1976); Echeverri A., Cardenas C., Calatayud M., Hadad C.Z., Gomez T., Theoretical analysis of the adsorption of ammonia–borane and their dehydrogenation products on the (001) surface of TiC and ZrC, Surf. Sci., 680, (2019); Vines F., Sousa C., Illas F., Liu P., Rodriguez J.A., A systematic density functional study of molecular oxygen adsorption and dissociation on the (001) surface of group IV-VI transition metal carbides, J. Phys. Chem. C., 111, (2007); Vines F., Sousa C., Liu P., Rodriguez J.A., Illas F., A systematic density functional theory study of the electronic structure of bulk and (001) surface of transition-metals carbides, J. Chem. Phys., 122, (2005); Florez E., Gomez T., Rodriguez J.A., Illas F., On the dissociation of molecular hydrogen by au supported on transition metal carbides: choice of the most active support, Phys. Chem. Chem.

Phys., 13, (2011); Manz T.A., Introducing DDEC6 atomic population analysis: part 3, Comprehensive Method to Compute Bond Orders, RSC Adv., 7, pp. 45552-45581, (2017); Van Vleet M.J., Misquitta A.J., Stone A.J., Schmidt J.R., Beyond born-Mayer: improved models for short-range repulsion in ab initio force fields, J. Chem. Theory Comput., 12, pp. 3851-3870, (2016); Song C., Ge Q., Wang L., DFT studies of Pt/Au bimetallic clusters and their interactions with the CO molecule, J. Phys. Chem. B., 109, (2005); Moreno N., Ferraro F., Florez E., Hadad C.Z., Restrepo A., Spin-orbit coupling effects in au m pt n clusters (m + n = 4), J. Phys. Chem. A., 120, (2016); Rodriguez J.A., Liu P., Vines F., Illas F., Takahashi Y., Nakamura K., Dissociation of SO2 on Au/TiC(001): effects of au-C interactions and charge polarization, Angew. Chemie - Int. Ed., 47, (2008); Florez E., Feria L., Vines F., Rodriguez J.A., Illas F., Effect of the support on the electronic structure of au nanoparticles supported on transition metal carbides: choice of the best substrate for au activation, J. Phys. Chem. C., 113, (2009); Mao J., Li S., Chu X., Yang Z., Interactions of small platinum clusters with the TiC(001) surface, J. Appl. Phys., 118, (2015); Rodriguez J.A., Feria L., Jirsak T., Takahashi Y., Nakamura K., Illas F., Role of au-C interactions on the catalytic activity of au nanoparticles supported on TiC(001) toward molecular oxygen dissociation, J. Am. Chem. Soc., 132, (2010); Fujitani T., Nakamura I., Akita T., Okumura M., Haruta M., Hydrogen dissociation by gold clusters, Angew. Chemie - Int. Ed., 48, (2009); Kubas G.J., Metal-dihydrogen and σ-bond coordination: the consummate extension of the dewar-chatt-duncanson model for metal-olefin π bonding, J. Organomet. Chem., 635, pp. 37-68, (2001); Lozano-Reis P., Prats H., Sayos R., Rodriguez J.A., Illas F., Assessing the activity of ni clusters supported on TiC(001) toward CO2and H2Dissociation, J. Phys. Chem. C., 125, (2021); Henkelman G., Uberuaga B.P., Jonsson H., Climbing image nudged elastic band method for finding saddle points and minimum energy paths, J. Chem. Phys., 113, (2000); Henkelman G., Jonsson H., Improved tangent estimate in the nudged elastic band method for finding minimum energy paths and saddle points, J. Chem. Phys., 113,

(2000); Wilke S., Scheffler M., Potential-energy surface for dissociation over Pd(100), Phys. Rev. B - Condens. Matter Mater. Phys., 53, (1996); Kroes G.J., Gross A., Baerends E.J., Scheffler M., McCormack D.A., Quantum theory of dissociative chemisorption on metal surfaces, Acc. Chem. Res., 35, (2002); Dong W., Kresse G., Hafner J., Dissociative adsorption of H2 on the Pd(111) surface, J. Mol. Catal. A Chem., 119, (1997); Pasteur A.T., Dixon-Warren S.J., Ge Q., King D.A., Dynamics of hydrogen dissociation on Pt{100}: steering, screening and thermal roughening effects, J. Chem. Phys., 106, (1997); Olsen R.A., Kroes G.J., Baerends E.J., Atomic and molecular hydrogen interacting with Pt(111), J. Chem. Phys., 111, (1999); Arboleda N.B., Kasai H., Dino W.A., Nakanishi H., (2007); Wiesenekker G., Kroes G.J., Baerends E.J., Mowrey R.C., Erratum: dissociation of H2 on Cu(100): dynamics on a new two-dimensional potential energy surface (journal of chemical physics (1995) 102 (3873)), J. Chem. Phys., 103, (1995); White J.A., Bird D.M., Payne M.C., Stich I., Surface corrugation in the dissociative adsorption of H2 on Cu(100), Phys. Rev. Lett., 73, (1994); Alvarez-Falcon L., Vines F., Notario-Estevez A., Illas F., On the hydrogen adsorption and dissociation on cu surfaces and nanorows, Surf. Sci., 646, (2016); Gross A., Hammer B., Scheffler M., Brenig W., High-dimensional quantum dynamics of adsorption and desorption of H2 at Cu(111), Phys. Rev. Lett., 73, (1994); Gross A., Scheffler M., Trends in the chemical reactivity of surfaces studied by ab initio quantum-dynamics calculations, Phys. Rev. B - Condens. Matter Mater. Phys., 59, (1999); Montoya A., Schlunke A., Haynes B.S., Reaction of hydrogen with Ag(111): binding states, minimum energy paths, and kinetics, J. Phys. Chem. B., 110, (2006); Xu Y., Greeley J., Mavrikakis M., Effect of subsurface oxygen on the reactivity of the Ag(111) surface, J. Am. Chem. Soc., 127, (2005); Hammer B., Norskov J.K., Why gold is the noblest of all the metals, Nature, 376, (1995); Lucci F.R., Darby M.T., Mattera M.F.G., Ivimey C.J., Therrien A.J., Michaelides A., Stamatakis M., Sykes E.C.H., Controlling hydrogen activation, spillover, and desorption with pd-au single-atom alloys, J. Phys. Chem. Lett., 7, (2016); Figueras M., Gutierrez R.A., Vines F., Ramirez

P.J., Rodriguez J.A., Illas F., Supported molybdenum carbide nanoparticles as hot hydrogen reservoirs for catalytic applications, J. Phys. Chem. Lett., 11, (2020)

Correspondence Address

T. Gomez; Theoretical and Computational Chemistry Center, Institute of Applied Sciences, Faculty of Engineering, Universidad Autonoma de Chile, Santiago, Avenida Pedro de Valdivia 425, Chile; email: tatiana.gomez@uautonoma.cl

Publisher

Elsevier B.V.

ISSN

01694332

CODEN

ASUSE

Language of Original Document

English

Abbreviated Source Title

Appl Surf Sci

Document Type

Article

Publication Stage

Final

Source

Scopus

EID

2-s2.0-85186699653