



Article

The Accuracy of Semi-Empirical Quantum Chemistry Methods on Soot Formation Simulation †

Yang Cong ¹ , Yu Zhai ¹ , Xin Chen ^{2,*} and Hui Li ^{1,*}

¹ Institute of Theoretical Chemistry, College of Chemistry, Jilin University, 2519 Jiefang Road, Changchun 130023, China

² Institute of Systems and Physical Biology, Shenzhen Bay Laboratory, Shenzhen 518055, China

* Correspondence: chenxin@szbl.ac.cn (X.C.); prof_lihui@jlu.edu.cn (H.L.)

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Abstract: Soot molecules are hazardous compounds threatening human health. Computational chemistry provides efficient tools for studying them. However, accurate quantum chemistry calculation is costly for the simulation of large-size soot molecules and high-throughput calculations. Semi-empirical (SE) quantum chemistry methods are optional choices for balancing computational costs. In this work, we validated the performances of several widely used SE methods in the description of soot formation. Our benchmark study focuses on, but is not limited to, the validation of the performances of SE methods on reactive and non-reactive MD trajectory calculations. We also examined the accuracy of SE methods of predicting soot precursor structures and energy profiles along intrinsic reaction coordinate(s) (IRC). Finally, we discussed the spin density predicted by SE methods. The SE methods validated include AM1, PM6, PM7, GFN2-xTB, DFTB2, with or without spin-polarization, and DFTB3. We found that the shape of MD trajectory profiles, the relative energy, and molecular structures predicted by SE methods are qualitatively correct. We suggest that SE methods can be used in massive reaction soot formation event sampling and primary reaction mechanism generation. Yet, they cannot be used to provide quantitatively accurate data, such as thermodynamic and reaction kinetics ones.

Keywords: soot; soot precursors; PAHs; molecular dynamics simulation; semi-empirical; DFT tight-binding; benchmark



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1. Introduction

Soot, a mass of carbonaceous nanoparticles, is a byproduct from incomplete combustion. The deposition of soot particles in a combustion chamber affects the combustion efficiency and even reduces the life of engines. Atmospheric soot also plays an important role in the global climate system [1]. Moreover, epidemiological evidence has already shown that black carbon, mature soot particles emitted into the atmosphere, is associated with health damage [2,3]. Soot precursor formation processes depend on fuel composition and flame conditions. Some parts of the complicated soot inception mechanisms are still unclear and waiting for more research.

Soot particles are generated from gas-phase species, which are called soot precursors. Although numerous experimental and simulation research studies support the theory that polycyclic aromatic hydrocarbon (PAH) species are major precursors [4,5], the genesis and growth mechanisms of aromatic compounds are still under debate. Various pathways are proposed for the formation of soot precursor, some research studies deem benzene as the first aromatic ring and soot-growing seed [5,6]. Another class of theoretical works suggested that stable cyclopentadienyl radicals formed by C₃H₃ and C₂H₂ are crucial initial structures to form other aromatic rings [7]. Johansson et al. [8] proposed a soot inception routine, which started from a cyclopentadienyl radical without the direct participation of benzene. Jin et al. [9] also suggested a propargyl radical (CH₂CCH) and

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Quantum Chemistry in the Age of Machine Learning Pavlo O. Dral,2022-09-16 Quantum chemistry is simulating atomistic systems according to the laws of quantum mechanics and such simulations are essential for our understanding of the world and for technological progress Machine learning revolutionizes quantum chemistry by increasing simulation speed and accuracy and obtaining new insights However for nonspecialists learning about this vast field is a formidable challenge Quantum Chemistry in the Age of Machine Learning covers this exciting field in detail ranging from basic concepts to comprehensive methodological details to providing detailed codes and hands on tutorials Such an approach helps readers get a quick overview of existing techniques and provides an opportunity to learn the intricacies and inner workings of state of the art methods The book describes the underlying concepts of machine learning and quantum chemistry machine learning potentials and learning of other quantum chemical properties machine learning improved quantum chemical methods analysis of Big Data from simulations and materials design with machine learning Drawing on the expertise of a team of specialist contributors this book serves as a valuable guide for both aspiring beginners and specialists in this exciting field Compiles advances of machine learning in quantum chemistry across different areas into a single resource Provides insights

into the underlying concepts of machine learning techniques that are relevant to quantum chemistry Describes in detail the current state of the art machine learning based methods in quantum chemistry *Hybrid Methods of Molecular Modeling* Andrei L. Tchougréeff, 2008-08-01 Hybrid Methods of Molecular Modeling is a self contained advanced review offering step by step derivation of the consistent theoretical picture of hybrid modeling methods and the thorough analysis of the concepts and current practical methods of hybrid modeling based on this theory Hybrid Methods of Molecular Modeling presents its material in a sequential way paying attention both to the physical soundness of the approximations used and to the mathematical rigor necessary for practical developing of the robust modeling code Historical remarks are given when it is necessary to put the current presentation in a more general context and to establish relation with other areas of computational chemistry The reader should have experience with basic concepts of computational chemistry and or molecular modeling Basic knowledge of operators wave functions electron densities is necessary

Encyclopedia of Physical Organic Chemistry, 6 Volume Set Zerong Wang, Uta Wille, Eusebio Juaristi, 2017-04-17 Winner of 2018 PROSE Award for MULTIVOLUME REFERENCE SCIENCE This encyclopedia offers a comprehensive and easy reference to physical organic chemistry POC methodology and techniques It puts POC a classical and fundamental discipline of chemistry into the context of modern and dynamic fields like biochemical processes materials science and molecular electronics Covers basic terms and theories into organic reactions and mechanisms molecular designs and syntheses tools and experimental techniques and applications and future directions Includes coverage of green chemistry and polymerization reactions Reviews different strategies for molecular design and synthesis of functional molecules Discusses computational methods software packages and more than 34 kinds of spectroscopies and techniques for studying structures and mechanisms Explores applications in areas from biology to materials science The Encyclopedia of Physical Organic Chemistry has won the 2018 PROSE Award for MULTIVOLUME REFERENCE SCIENCE The PROSE Awards recognize the best books journals and digital content produced by professional and scholarly publishers Submissions are reviewed by a panel of 18 judges that includes editors academics publishers and research librarians who evaluate each work for its contribution to professional and scholarly publishing You can find out more at proseawards.com Also available as an online edition for your library for more details visit Wiley Online Library

Computational Advances in Organic Chemistry: Molecular Structure and Reactivity Cemil Ögretir, Imre G. Csizmadia, 2012-12-06 The lecturers as well as the participants came from varied scientific backgrounds for the NATO Advanced Study Institute ASD held at Altinoluk Edremit Turkey during the period of July 31 August 12 1989 The lecturers were University Professors from the USA Canada England Germany France and Spain and they covered a broad spectrum of specialties from methodology to applications On the other hand students coming from the various NATO countries arrived with an inhomogeneous background to absorb the broad spectrum of material covered by the lecturers However by the end of the two week period of the ASI that initial difference in scientific background had been

reduced substantially The lecturers had covered subject matters from the most fundamental to the most applied aspects of theoretical and computational organic chemistry The lectures were argnmented with tutorial sessions and computational laboratory led by a small group of carefnlly selected tutors Overall this NATO ASI was a at success and the Editors are hopeful that the present volume will communicate the scientific success and will radiate the intellectual spirit of the meeting

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Computational Chemistry David Young, 2004-04-07 A practical easily accessible guide for bench top chemists thisbook focuses on accurately applying computational chemistrytechniques to everyday chemistry problems Provides nonmathematical explanations of advanced topics incomputational chemistry Focuses on when and how to apply different computationaltechniques Addresses computational chemistry connections to biochemicalsystems and polymers Provides a prioritized list of methods for attacking difficultcomputational chemistry problems and compares advantages anddisadvantages of various approximation techniques Describes how the choice of methods of software affectsrequirements for computer memory and processing time

Lectures On Chemical Bonding And Quantum Chemistry Sambhu N Datta, 2019-02-25 The concept of a chemical bond evolved from a variety of experimental observations It became useful to understand at times even predict the molecular structure reactivity and mechanism of chemical reactions Every aspect of the concept of bonding received a quantitative interpretation from the advent of quantum mechanics and its application to chemistry In Lectures on Chemical Bonding and Quantum Chemistry the reader will find a comprehensive discourse on the basic interpretation of the chemical bond as well as current understanding in terms of a dancing molecule that not only travels rotates and pulsates around an equilibrium molecular structure but also interacts and collides with other molecules thereby transferring linear and angular momentum characteristics and adjusting total energies One will also find a thorough survey of quantum mechanical methodologies for calculation of molecular characteristics in specific states and their changes under spectroscopic transitions tunneling electron and proton transfer

phenomena and so on Guides to more advanced levels of theory are also provided

Modern Aspects of Electrochemistry Costas G. Vayenas, Brian E. Conway, Ralph E. White, 2005-12-28 This volume of Modern Aspects contains a remarkable spread of topics covered in an authoritative manner by some internationally renowned specialists In a seminal chapter Drs Babu Oldfield and Wieckowski demonstrate eloquently the strength of electrochemical nuclear magnetic resonance EC NMR to study in situ both sides of the electrochemical interface via the simultaneous use of and This powerful non invasive technique brings new insights to both fundamental and practical key aspects of electrocatalysis including the design of better anodes for PEM fuel cells The recent impressive advances in the use of rigorous ab initio quantum chemical calculations in electrochemistry are described in a remarkable chapter by Marc Koper one of the leading protagonists in this fascinating area This lucid chapter is addressed to all electrochemists including those with very little prior exposure to quantum chemistry and demonstrates the usefulness of ab initio calculations including density functional theory DFT methods to understand several key aspects of fuel cell electrocatalysis at the molecular level The most important macroscopic and statistical thermodynamic models developed to describe adsorption phenomena on electrodes are presented critically in a concise and authoritative chapter by Panos Nikitas The reader is guided through the seminal contributions of Frumkin Butler Bockris Guidelli and others to the current state of the art adsorption isotherms which are both rigorous and in good agreement with experiment

Organic and Bio-molecular Chemistry - Volume II Francesco Nicotra, 2009-04-14 Organic And Bio Molecular Chemistry is the component of Encyclopedia of Chemical Sciences Engineering and Technology Resources in the global Encyclopedia of Life Support Systems EOLSS which is an integrated compendium of twenty one Encyclopedias The Theme on Organic And Bio Molecular Chemistry in the Encyclopedia of Chemical Sciences Engineering and Technology Resources deal with the discipline that studies the molecules of life which are made by carbon atoms and includes also all the synthetic compounds the skeletons of which contain carbon atoms The first chapter describes in general terms for not expert readers what Organic and Bio molecular chemistry is the nature and behavior of organic compounds in living organisms the importance of organic compounds in the market and in our every day life The subsequent chapters are organized in order to provide the reader with information on the structure reactivity analysis and different applications of Organic Compounds These two volumes are aimed at the following five major target audiences University and College students Educators Professional practitioners Research personnel and Policy analysts managers and decision makers and NGOs

The Quantumchemical Calculation of Fluorine-containing Oxidizers of the Differential Fuels V. A. Babkin, 2007 The calculation of traditional fluorine containing F₂ OF₂ N₂F₄ ClO₃F ClF₅ ClF₃ and oxygen containing OF₂ O₂ H₂O₂ N₂O₄ HNO₃ ClO₃F oxidisers of differential fuels has been performed by the different classical semi empirical quantum chemical methods CNDO CNDO 2 MNDO AM1 PM3 and B INITIO in the many principal basis sets optimising the all geometric parameters It is shown the high correlative dependencies between the burn parameters of the differential fuels H₂ N₂H₄

H₂N₂ CH₃ 2 CH₂ AlH₃ B₅H₉ BeH₂ and calculated values of quantum chemical parameters of the fluorine containing oxygencontaining oxidisers exist in the form of I_p is specific impulse of pressure P_1 is specific traction in atmosphere P_i is specific traction in vacuum depending on Q_{fmin} is minimum electronic charge on fluorine atom Q_{fmin} is the minimum electronic charge on oxygen atom The authors performed comparative analysis of results of the quantum chemical semi empirical and ab initio calculations for different fuels The simple interpretation and illustration of the physical nature of these correlative dependencies are offered The authors established the technique of theoretical estimation of the burn parameters of oxidisers of the differential fuels that may be used to look for new more efficient non pollution oxidisers

Polyamic Acids and Polyimides Michael I. Bessonov, Vladimir A. Zubkov, 2020-11-25 Polyamic Acids and Polyimides surveys significant developments in basic research in the chemistry and physics of polyamic acids and polyimides over the last several years Traditional and new topics are discussed including catalytical imidization chemical reactions at thermal treatment quantum chemical study of synthesis and structure properties of isolated molecules and supermolecular and crystalline structures The book will be an excellent reference for researchers practitioners and graduate students working with polyimides and related heat resistant polymers and materials

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