



Article

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

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† Electronic Supplementary Information available.

**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  $C_3H_3$  and  $C_2H_2$  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 ( $CH_2CCH$ ) and

# Semi Empirical Methods Of Quantum Chemistry

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### **Table of Contents Semi Empirical Methods Of Quantum Chemistry**

1. Understanding the eBook Semi Empirical Methods Of Quantum Chemistry
  - The Rise of Digital Reading Semi Empirical Methods Of Quantum Chemistry
  - Advantages of eBooks Over Traditional Books
2. Identifying Semi Empirical Methods Of Quantum Chemistry
  - Exploring Different Genres
  - Considering Fiction vs. Non-Fiction
  - Determining Your Reading Goals
3. Choosing the Right eBook Platform
  - Popular eBook Platforms
  - Features to Look for in an Semi Empirical Methods Of Quantum Chemistry
  - User-Friendly Interface
4. Exploring eBook Recommendations from Semi Empirical Methods Of Quantum Chemistry
  - Personalized Recommendations
  - Semi Empirical Methods Of Quantum Chemistry User Reviews and Ratings
  - Semi Empirical Methods Of Quantum Chemistry and Bestseller Lists

5. Accessing Semi Empirical Methods Of Quantum Chemistry Free and Paid eBooks
  - Semi Empirical Methods Of Quantum Chemistry Public Domain eBooks
  - Semi Empirical Methods Of Quantum Chemistry eBook Subscription Services
  - Semi Empirical Methods Of Quantum Chemistry Budget-Friendly Options
6. Navigating Semi Empirical Methods Of Quantum Chemistry eBook Formats
  - ePub, PDF, MOBI, and More
  - Semi Empirical Methods Of Quantum Chemistry Compatibility with Devices
  - Semi Empirical Methods Of Quantum Chemistry Enhanced eBook Features
7. Enhancing Your Reading Experience
  - Adjustable Fonts and Text Sizes of Semi Empirical Methods Of Quantum Chemistry
  - Highlighting and Note-Taking Semi Empirical Methods Of Quantum Chemistry
  - Interactive Elements Semi Empirical Methods Of Quantum Chemistry
8. Staying Engaged with Semi Empirical Methods Of Quantum Chemistry
  - Joining Online Reading Communities
  - Participating in Virtual Book Clubs
  - Following Authors and Publishers Semi Empirical Methods Of Quantum Chemistry
9. Balancing eBooks and Physical Books Semi Empirical Methods Of Quantum Chemistry
  - Benefits of a Digital Library
  - Creating a Diverse Reading Collection Semi Empirical Methods Of Quantum Chemistry
10. Overcoming Reading Challenges
  - Dealing with Digital Eye Strain
  - Minimizing Distractions
  - Managing Screen Time
11. Cultivating a Reading Routine Semi Empirical Methods Of Quantum Chemistry
  - Setting Reading Goals Semi Empirical Methods Of Quantum Chemistry
  - Carving Out Dedicated Reading Time
12. Sourcing Reliable Information of Semi Empirical Methods Of Quantum Chemistry
  - Fact-Checking eBook Content of Semi Empirical Methods Of Quantum Chemistry
  - Distinguishing Credible Sources
13. Promoting Lifelong Learning

- Utilizing eBooks for Skill Development
- Exploring Educational eBooks

### 14. Embracing eBook Trends

- Integration of Multimedia Elements
- Interactive and Gamified eBooks

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