



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

**Andrei L. Tchougréeff**



## **Semi Empirical Methods Of Quantum Chemistry:**

## Adopting the Track of Phrase: An Emotional Symphony within **Semi Empirical Methods Of Quantum Chemistry**

In some sort of used by monitors and the ceaseless chatter of instantaneous interaction, the melodic beauty and emotional symphony developed by the prepared term frequently diminish in to the background, eclipsed by the constant sound and distractions that permeate our lives. However, nestled within the pages of **Semi Empirical Methods Of Quantum Chemistry** a stunning literary treasure filled with raw thoughts, lies an immersive symphony waiting to be embraced. Crafted by a masterful composer of language, that captivating masterpiece conducts viewers on a mental trip, well unraveling the hidden melodies and profound impact resonating within each cautiously constructed phrase. Within the depths with this moving assessment, we will explore the book is central harmonies, analyze its enthralling writing design, and submit ourselves to the profound resonance that echoes in the depths of readers souls.

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