

Quantum chemical molecular dynamics simulations of fullerene self-assembly from benzene

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Abstract

Fullerenes were discovered as products of the evaporation of carbon and these can also be formed in low-pressure fuel-rich flames of certain hydrocarbons. Fullerenes formation in diffusion flames of hydrocarbons (such as benzene) diluted with argon and burning in oxygen offers advantages for practical operation. Experimental results show that fullerene formation is sensitive to changes in operating conditions, such as fuel/oxygen ratio, chamber pressure, and inert gas dilution. With lower fuel/oxygen ratios the amount of fullerenes formed decreases to small amounts near the critical conditions for soot formation. Despite the advances in refining the fullerene formation conditions in combustion processes, in flame the chemical formation mechanisms involved have still not been elucidated at the atomic level, preventing a rational approach towards systematic improvement of reaction conditions. In this work, we present high-temperature quantum chemical molecular dynamics simulations on model hydrocarbon systems initially consisting of benzene molecules to shed light on their atomistic self-assembly mechanism. The simulations were carried out using density functionally tight binding (DFTB) method. The effect of hydrogen, carbon to hydrogen ratio, carbon density and temperature on the fullerene molecule formation is discussed in detail.