Study on soot formation characteristics in the diesel combustion process based on an improved detailed soot model

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ARTICLE INFO

ABSTRACT

Although much research has been done on soot formation in engines, the mechanisms involved in the process are poorly understood and models of the process are overly simplistic. An improved detailed soot model is developed that couples a reduced diesel surrogate fuel chemical reaction mechanism of n-heptane/toluene, implemented into KIVA-3V2 code, for the numerical investigation of soot formation, mass concentration, and size distribution in diesel engines. This detailed soot model incorporates the effects of soot precursors, including isomers of acetylene and polycyclic aromatic hydrocarbons (PAHs), and the physical processes of PAH deposition on the particle surface, soot formation, and particle surface growth. Compared with experiment results in an optical engine and a single-cylinder diesel engine, the improved detailed soot model was effective: the simulated in-cylinder combustion pressure, heat release rate, and ignition timing were in excellent agreement with the experimental results. The simulated two-dimensional, transient distribution of soot concentration was in good agreement with that obtained by using the two-color method, and the simulated changing trend of soot emission was consistent with the experimental results. Therefore, the detailed soot model can be used to accurately simulate and predict soot emission at different conditions in diesel engines. Furthermore, at the initial stage of combustion, large amounts of small-size soot particles were produced by the pyrolysis reactions and polymerization of the hydrocarbon fuel, and the particle size was in the range of 5–40 nm. At the middle stage of combustion, soot particles continued to grow by particle coagulation, surface growth, and the deposition of PAHs, which greatly expanded the particle size range and caused many large-size particles to be produced in the engine cylinder. At the late stage of combustion, the range of particle size distribution stabilized under the influence of further oxidation reactions, and the particle size ranged from 5 to 20 nm. The details of soot-relevant quantities (e.g., particle size, number concentration, and mass) provide valuable insights into soot formation and oxidation processes in diesel engines.

1. Introduction

Soot formation and oxidation mechanisms are two of the most important questions remaining about diesel engine emission control, in spite of the extensive research on these topics [1]. A full understanding of the detailed mechanism of soot formation and oxidation processes is still lacking. The primary component of diesel fuel is quite complex, including straight-chain alkanes, branched-chain alkanes, naphthene, alkanes, and aromatics. Therefore, the direct numerical simulation of diesel fuel is very difficult. N-heptane is similar to conventional diesel in cetane number, which has an important effect on diesel combustion, and the combustion process in diesel engines has been simulated using n-heptane as a surrogate diesel fuel [2].

Conventional diesel fuel contains 30–35% polycyclic aromatic hydrocarbons (PAHs), and numerous studies have demonstrated that aromatics play a key role in the soot formation process in diesel engines [1,3–5]. Therefore, the chemical kinetic mechanisms of the mixture of n-heptane and aromatics should be developed to simulate the diesel combustion process, the soot formation process, and the oxidation process more accurately and effectively in diesel engines. Golovitchev et al. [6] used 70 mol% n-heptane and 30 mol% toluene mixture as the diesel surrogate fuel, which could effectively represent the cetane number and the other properties of diesel fuels; they coupled the chemical kinetic mechanism with 3D computational fluid dynamics (CFD) to model diesel combustion and soot formation processes in diesel engines. This chemical kinetic mechanism contains 70 species and 313 reactions. $C_{14}H_{28}$ is assumed to be the chemical expression of the mixture to replace real diesel fuel in this mechanism, and it is assumed that $C_{14}H_{28}$

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Nomenclature

A_1	benzene (C_6H_6)	N	particle number concentration (m^{-3})
A_2	naphthalene ($C_{10}H_8$)	N_A	Avogadro number (6.023×10^{23} mol^{-1})
A_{soot}	total surface area of all soot particles in a unit volume	n_{OH}	number concentration of OH radical (m^{-3})
BTDC	before top dead center	p_{C2H2}	partial pressure of acetylene (Pa)
C_α	constant coefficient (6.55)	PAHs	polycyclic aromatic hydrocarbons
C_{min}	minimum number of carbon atoms	R	molar gas constant (8.3145 J mol^{-1} K^{-1})
$C(s)$	soot mass density (mol m^{-3})	T	average temperature (K)
C_{soot}	initial carbon molecule	TDC	top dead center
C_{soot}^*	radicals of the carbon molecule	α	steric factor
DI	direct injection	ρ_{soot}	particle density (1800 kg m^{-3})
f_v	particle volume fraction	γ_{OH}	collision efficiency between OH radical and soot particles (0.13)
k	reaction rate coefficients (s^{-1})		
k_B	Boltzmann constant (1.38×10^{-23} J K^{-1})	x_{soot}^*	number density of C_{soot}^* (m^{-3})
M_{OH}	relative molecular mass of OH radical	ω	reaction rate (mol m^{-3} s^{-1})
M_{soot}	molecular weight of soot (12 g mol^{-1})		

decomposes into n-heptane and toluene through two global reactions as shown below to maintain a constant molar proportion of n-heptane to toluene (7:3, respectively),

$$2C_{14}H_{28} \Rightarrow 3C_7H_{16} + C_7H_8$$

$$3C_{14}H_{28} + O_2 \Rightarrow 4C_7H_{16} + 2C_7H_8 + 2H_2O$$
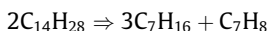
The traditional empirical soot model, such as Hiroyasu's two-step soot model [7], provided basic knowledge on bulk soot distribution and transport in the high-temperature combustion environments of conventional diesel engines. However, Hiroyasu's model oversimplified the diesel soot formation processes, and the particle size and other information in the process of soot formation could not be calculated. To overcome some limitations of the two-step soot model, Fusco et al. [8] proposed an eight-step soot model that added two intermediate products and included particle nucleation, coagulation, surface growth, and oxidation. Fuel is assumed to decompose into soot precursors and other components through one global reaction in this multi-step soot model. Kazakov et al. [9] improved upon the Fusco model, developing a nine-step semi-empirical soot model that could make the particle size distribution more homogenized.

Detailed soot models have been proposed by researchers to simulate a more realistic formation and oxidation process of soot particles from two aspects: gas-phase chemical reaction kinetics and particle dynamics [10–12]. In gas-phase chemical reaction kinetics, detailed reaction mechanisms are applied; particle dynamics uses advanced numerical methods.

The chemical reaction pathways to soot were investigated by experimenting with detailed kinetic models of soot formation under the conditions used in shock-tube pyrolysis experiments by Frenklach et al. [13]. The formation of the first aromatic ring is based on the cyclization reaction of ethenyl and acetylene. The stabilizing species, such as acenaphthylene and coronene, are produced by the polymerization of PAHs through the H-abstraction/C_2H_2-addition reaction mechanism. In addition, the formation of benzene plays a particularly important role in the polymerization process of PAHs. Frenklach and Wang [14] suggested that the main bottleneck of soot formation appeared at the formation of the first aromatic ring in the combustion process of acetylene. In Frenklach's soot model, soot concentration and size distribution moments are changed with the further growth of larger PAHs through the process of linear aggregation.

The rapid polymerization of PAHs continues until the planar aromatic structure with 12–14 benzene rings is formed. Through the coagulation of PAHs, two polymeric PAH molecules are com-

bined into an initial soot particle. With the decrease of the PAHs number concentration, the soot number concentration decreases. In this process, since the total mass and the volume of soot particles remain nearly unchanged, the soot particles with three-dimensional structure are produced.

Tao et al. [15] proposed a soot model that was more applicable to the high-temperature and high-pressure conditions of diffusion combustion in diesel engines. Tao's soot model included two parts: a detailed chemical reaction mechanism and a phenomenological semi-empirical soot model.

In this paper, an improved detailed soot model is described, which is composed of the detailed chemical reaction mechanism and the phenomenological soot model; this soot model is used to analyze and study the formation characteristics of soot particles in the combustion process. The effectiveness and correctness of the improved detailed soot model is experimentally verified in an optical engine and a single-cylinder diesel engine. The reduced chemical reaction mechanism is applied as the gas-phase chemical kinetics part of the detailed soot model. To incorporate particle dynamics, the effects of PAH deposition on particle size distribution and on the soot formation process are included to provide a reliable scientific basis for understanding the formation and oxidation process of soot particles, yielding valuable insights into controlling particle emissions in diesel engines.

2. Computing models

2.1. Chemical reaction mechanism of diesel fuel

The reduced kinetic mechanism of the n-heptane/toluene diesel surrogate fuel was validated through diesel engine combustion modeling by researchers [6,16,17], but the computation cost for evaluating 70 species and 313 reactions is too large when coupled with three-dimensional CFD. Hence, the chemical reaction kinetic mechanism of the n-heptane/toluene diesel surrogate fuel is further reduced in this paper.

2.1.1. The pyrolysis reaction mechanism of diesel surrogate fuel $C_{14}H_{28}$

$C_{14}H_{28}$ was decomposed into n-heptane and toluene through two global reactions to keep mole proportion of n-heptane and toluene at 7:3 [6]. However, reaction rates of the two decomposing global reactions were so low that a long $C_{14}H_{28}$ decomposing duration occurred and heat release was suppressed. Thus, to advance ignition timing of the diesel surrogate fuel and keep the mole ratio