

# Numerical study on soot removal in partial oxidation of methane to syngas reactors

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## Abstract

The serious carbon deposition existing in catalytic partial oxidation of methane (CPOM) to syngas process is one of the key problems that impede its industrialization. In this study, 3-dimensional unsteady numerical simulations of the soot formation and oxidation in oxidation section in a heat coupling reactor were carried out by computational fluid dynamics (CFD) approach incorporating the Moss-Brookes model for soot formation. The model has been validated and proven to be in good agreement with experiment results. Effects of nozzle type, nozzle convergence angle, channel spacing, number of channels, radius/height ratio, oxygen/carbon ratio, preheat temperature and additional introduction of steam on the soot formation were simulated. Results show that the soot formation in oxidation section of the heat coupling reactor depends on both nozzle structures and operation conditions, and the soot concentration can be greatly reduced by optimization with the maximum mass fraction of soot inside the oxidation reactor from 2.28% to 0.0501%, and so that the soot mass fraction at the exit reduces from 0.74% to 0.03%.

## Key words

carbon deposition; partial oxidation of methane; numerical simulation; soot formation; nozzle; optimization

## 1. Introduction

Due to the increasing difficulties and costs of petroleum recovery, conversion of methane to syngas will play an important role in future industries. The converted syngas can be used in processes like Fischer-Tropsch (F-T) synthesis, methanol synthesis, ammonia synthesis, hydrotreating processes and reduction in metallurgical operations [1]. Currently, the investment and operation of syngas production section account for 50%–75% of the total process costs. Therefore, the improvements and renovations of the syngas production process are of great importance [2]. Although other routes for methane conversion, such as steam reforming (SR), CO<sub>2</sub> reforming, or some combination routes [3–10], have been proposed, the CPOM process is still in primary consideration due to its advantages like: (1) The reaction is mildly exothermic that can be operated under lower temperatures and thus the energy consumption is lower; (2) The stoichiometric H<sub>2</sub>/CO ratio of the produced syngas is 2, which is the optimum value for processes like methanol and F-T syntheses [11–21].

In previous studies, fixed bed reactors were most commonly adopted in methane conversion [11,22], and different modifications of the reactor configurations have been reported

to improve their performances [23–25]. The current research group has proposed a heat coupling reactor, as shown in Figure 1, in which two coaxial tubes were installed in the reactor [26]. Oxidation section is located inside the inner tube and reforming section is located in the annulus. The methane was first partially oxidized in the oxidation section and the effluent was then introduced into the reforming section. In the reactor, the heat released by the oxidation reactions thus can be directly absorbed by the reforming reactions. However, one defect occurred in the heat coupling reactor. The serious soot formation has caused a series of problems such as lowering the conversion rate, deactivation of the downstream reforming catalyst and reducing heat transfer efficiency.

The process of soot formation and oxidation is so extremely complicated that researchers have not completely unveiled its mechanisms. In order to reduce the release of soot at the exit of reactor, it is necessary to measure the soot distributions inside the reactor accurately, which helps the deeper understanding of mechanisms of soot formation and oxidation, and therefore, more accurate soot formation model can be established. Researchers have developed methods like sub-micron particle polarization intensify differential scattering (PIDS) [27] and laser-induced incandescence imaging [28,29]

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to measure soot distributions in diesel engines. But these experimental measurements are time and cost consuming and also have deviations due to their limitations [30].

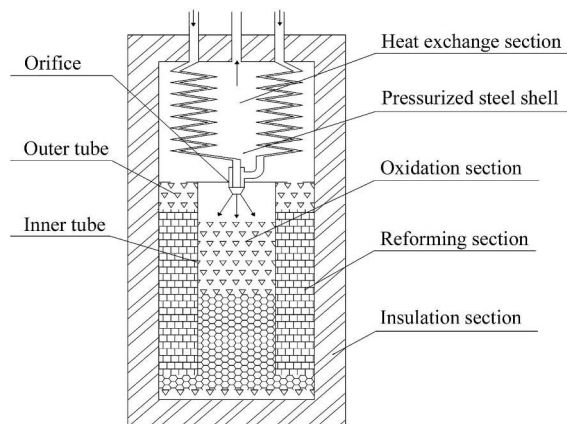


Figure 1. Schematic diagram of heat coupling reaction system

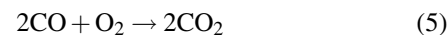
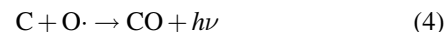
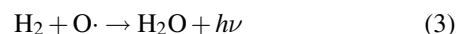
With the development of computational power, researchers begin to investigate soot formations through numerical methods. Most of previous soot models were developed for diesel engines [31]. In recent years, these models were mainly multi-stage ones which took into account particle growth and soot dynamics. For example, for particle nucleation, Tesner et al. [32] introduced a three-stage model. Following this framework, several models were proposed in which the intermediate species for particle nucleation were linked to acetylene [33,34] or a generic soot precursor radical [35]. With considerations of soot inception (methane cracking, formation and growth of aromatic rings, polymerization and oxidation of aromatics), surface growth, coagulation and oxidation, Frenlach and co-workers [36,37] proposed a detailed dynamics model of soot formation, which was validated by measurements of laminar premixed flames. D'Errico et al. [38] and Hong et al. [39] also applied the four-step Moss-Brookes soot model in their diesel engine simulations. Lindstedt and Louloudi [40] also proposed a joint-scalar transported PDF (probability density function) modeling on soot formation. The detailed kinetic soot model can be divided into two components: one for gas phase chemistry and the other for particle growth and destruction.

As can be seen, the multi-stage soot model can well describe the soot formation mechanisms in diesel engine and thus can be expanded the applications in the CPOM reactions. Since the heat coupling reactor proposed by current research group has a distinctive structure which is far from that of diesel engines, it is still necessary to investigate the soot formation in such reactor. And the soot formation, which is generated in the oxygen concentrated region due to the poor mixing of reactant gases, is one of the key problems that affect the efficiency of total reactor. Since it is difficult to have detailed analysis of soot formation in oxidation section of the reactor due to its really complex mechanisms, in this study, a four-step CFD method was employed to simulate the soot formation and the reduction of soot formation by optimization of reactor structure and operation conditions was discussed.

## 2. CFD modeling with premixed combustion and soot formation

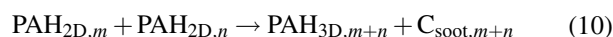
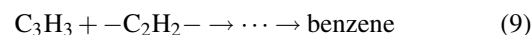
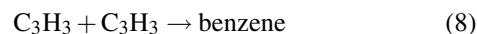
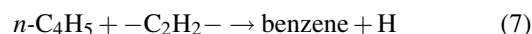
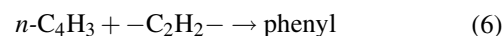
### 2.1. Soot formation mechanisms

The elementary reactions in the gas phase include:



It can be seen that the carbon deposition is mainly caused by soot formation from Reaction 2, thus the inhibition of soot formation is of the first importance.

The mechanisms of carbon formation without oxygen include initial formation of soot and coagulation of soot particles. The initial formation of soot can be divided into following steps: methane cracking (Equation 6); formation of the first aromatic ring (Equations 7–9); formation of polycyclic aromatic hydrocarbons (PAH); growth and polymerization of PAH and nucleation of soot (Equation 10). The process is presented in Figure 2.



The coagulation of soot particles can be presented by:



where,  $i$ ,  $j$  and  $n$  represent soot particles and carbon numbers, and 2D indicates that the PAH molecular is in 2-dimensional form.

The particle concentration decreases with:

$$\frac{d(N_i^{\text{soot}})}{dt} = -k(N_i^{\text{soot}})^2 \quad (13)$$

where,  $k$  is the reaction rate constant,  $\text{kg}\cdot\text{m}^3\cdot\text{s}^{-1}$ .

If oxygen exists, oxidation of soot occurs in addition to the previous reactions. Thus the reaction should include the following steps: adsorption of PAH on the soot surface, surface growth and oxidation of soot.

According to the mechanisms of soot formation, the oxidation reactor can be divided into three zones: main

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