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Research paper

Geometric parameters effect on the reaction zone of premixed CH_4 catalytic combustion in a fibrous porous medium

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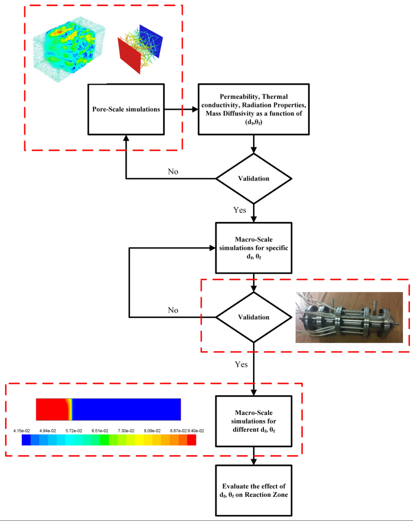
HIGHLIGHTS

- Experimental and numerical study of catalytic combustion in fibrous porous media.
- Study of reaction zone inside complex fibrous porous media based on pore-scale simulation.
- Effect of geometric parameters (fibers orientation, and fibers diameter) on the reaction zone.
- Temperature distribution in a centerline of a catalytic combustion reactor.

GRAPHICAL ABSTRACT



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ABSTRACT

Flameless catalytic heaters are thermal systems in which the chemical energy of gaseous fuel converts into heat with zero NO_x and low CO (≤ 10 ppm) emissions. Hence, they are called green heaters in the industry. These heaters benefit from a fibrous porous medium as support for catalyst nanoparticles. Pore structure has a significant effect on transport phenomena inside the porous medium. With the dramatic growth of computers, it is possible to study the impact of geometric detail on combustion phenomena. This study investigates the effect of fiber orientation and diameter on the temperature distribution and the reaction zone of CH_4 catalytic combustion in the fibrous medium. As the fiber axis angle increases from 10° to 90°, the reaction zone moves 24.1% toward the reactor inlet, the maximum temperature increases by 69.7 °C, and its distribution becomes more non-uniform. Moreover, as the diameter of the fibers increased from 5 μm to 10 μm, the reaction site moved 29.8% toward the end of the reactor, the maximum temperature increased by 34.9 °C, and its distribution became slightly more uneven. The results showed how the diameter and orientation of fibers influence the performance of porous catalytic reactors. This issue should be considered, especially to increase the life of catalytic burners.

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1. Introduction

Catalytic radiant heaters generate heat from natural gas combustion without the flame with zero NO_x and low CO (≤ 10 ppm) emissions [1-2]. This type of heater uses a fibrous porous medium on which the catalyst is coated.

The fibrous porous medium has increased the flammability limits and significantly reduced pollutants [3]. This type of porous medium is widespread in the application of thermal insulation [4]. However, due to the geometric complexity, analytical solutions for this problem have been limited and mainly solved with simplified assumptions [5-6]. Pore-scale simulation is a method for studying the effect of geometric parameters on flow, heat, and mass transfer properties in this type of porous medium that avoids time-consuming and costly manufacturing and testing processes [7-9]. In this method, the governing phenomena are studied by simulating a small part of the entire geometry representing the whole computational domain while considering all the details. For example, we can refer to the studies of Palma *et al.* [10], Zhang *et al.* [11], and Wang *et al.* [12], which have used this method in a variety of porous environments. In the mentioned studies, this method was used to calculate the properties of the porous medium. Banerjee and Paul [13] and Ghareghani *et al.* [14] reviewed both the applications of porous media in catalytic combustion and the challenges in this field. Yan *et al.* investigated the effect of material, thickness, and inlet conditions (equivalence ratio, inlet velocity) on temperature distribution and combustion products in a porous micro combustor [15]. To improve the thermal performance and efficiency of micro thermophotovoltaic systems, Wu *et al.* numerically investigated the effect of the material of the porous region and its wall in an air-hydrogen premixed combustion chamber and its effect on temperature distribution and entropy generation [16]. W. Liu *et al.* numerically investigated the effect of the porous washcoat thickness, porosity, average pore diameter, and tortuosity on combustion temperature and emission performance in a micro-channel porous reactor with methane fuel [17].

In our previous studies, the effect of the diameter and orientation of the fibers in the porous medium on the flow properties, heat transfer, and equivalent mass transfer was identified [18-23]. However, after reviewing the literature, it was found that the effect of changing these geometric properties on the thermal performance of premixed combustion in this type of porous medium has not been studied. Therefore, the main innovation in this article is to investigate this issue and its effect on the reaction zone of premixed CH_4 catalytic combustion.

The Pore-scale simulation process for this study is shown schematically in Fig. 1. Equivalent coefficients of permeability (κ), heat conduction $K_{eff.}$, heat radiation (α, β, ω), and mass diffusion $D_{eff.}$ are the main results of the pore-scale simulations. These equivalent coefficients can be used in macro-scale numerical studies to predict temperature distribution and reaction rate.

In our previous works [18-19], summarized in Table 1, the structure of the porous medium was studied.

Table 1. Geometric characteristics of the porous medium.

Character	Amount
Porosity	98.7%
Diameter of the fibers	5 μm
Angle between fibers and flow drection (θ)	≈ 90 °
Fiber material	Al_2O_3
Coated catalyst	Pt

Moreover, using this technique, we obtained the permeability, conduction, and radiation heat transfer and mass transfer coefficients as a function of the fiber's axis angle with the flow direction (θ) reported in [20-23]. The model's calculation process was validated with experimental results.

In this study, the effect of fibers orientation for five different angles of 10°, 30°, 45°, 60°, and 90° on the temperature distribution and methane combustion zone is addressed using calculated thermophysical properties. In other words, in our previous studies, the effect of geometric properties (orientation and diameter of fibers)

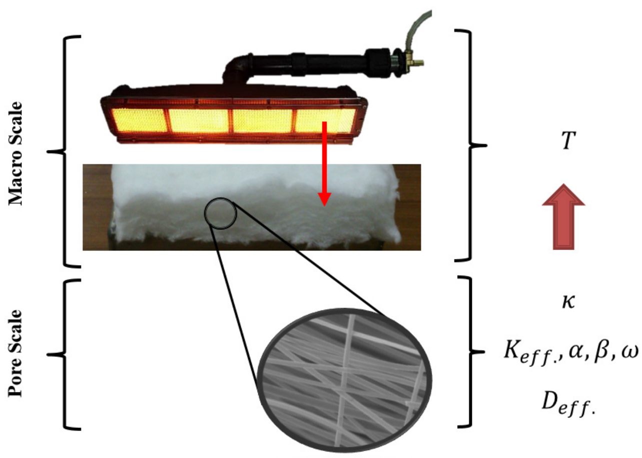

Fig. 1. Pore-scale and macro-scale simulation steps.

on flow properties, heat transfer, and mass transfer in the fibrous porous medium was obtained using pore-scale simulation. The mentioned coefficients were presented as a function according to the geometrical properties and validated. In this paper, thermophysical properties are obtained from the results of our previous studies and used in the simulation for different combinations of fiber orientation and diameter. In this way, the effect of these cases is evaluated without entering the geometric details into a full-scale simulation. The obtained temperature distribution was used to investigate the combustion reaction zone and the maximum temperature changes.

The fiber's orientation in the fibrous medium is shown schematically in Fig. 2 for three angles of 10°, 45°, and 90°. The inlet flow direction of the methane-air mixture is considered in the *z*-direction.

In addition, the effects of fiber diameter on temperature distribution and methane combustion area for four values of 5, 7, 8.5, and 10 μm are also discussed in this application. The method of changing the diameter of the fibers in the fibrous porous medium in this study is shown schematically in Fig. 3. Since this section investigates the effect of diameter, other geometric characteristics should remain constant. One of these characteristics is Solid volume fraction (SVF). For this purpose, the total solid volume occupied by the environment's fibers must remain constant. If, according to Fig. 3, each fiber in the first state is divided into m in the second state (diameter reduction), then to keep the volume of the

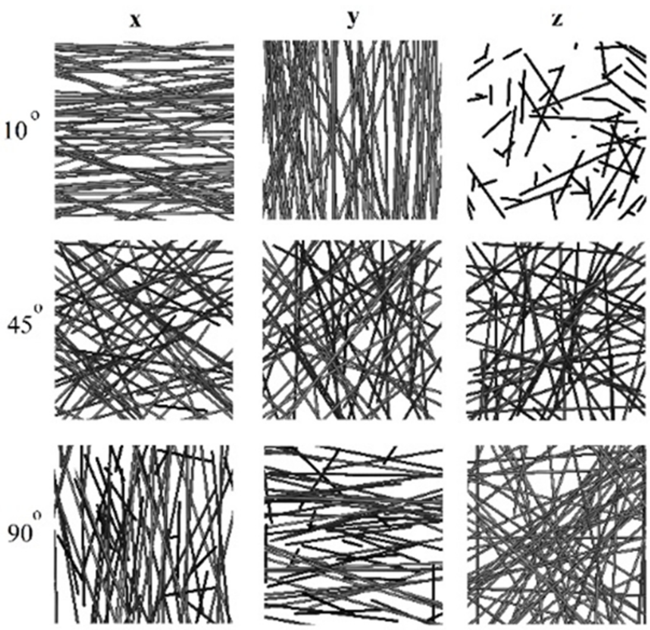

Fig. 2. The fiber's orientation in the fibrous medium for three different angles of 10°, 45°, and 90° [22].

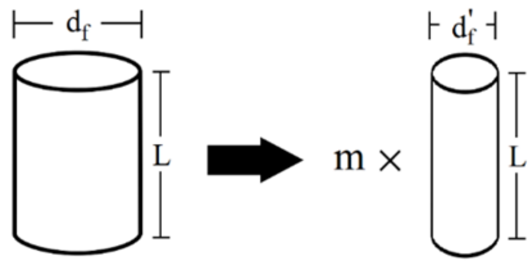

Fig. 3. Converting a fiber to *m* fibers of the same length at constant porosity [22].

solid constant, *m* is obtained from Eq. (1).

$$m = (d_f / d'_f)^2 \tag{1}$$

2. Numerical section

The pressure drop inside the medium was modified using the simulation's permeability coefficient for an inlet velocity of 0.005 m.s^{-1}, the expected flow rate in the heater under the operating conditions, and then calculated based on the experiments [20].

Due to the low velocity and low Reynolds number, the effect of inertia was ignored. The coefficient of equivalent thermal conductivity was previously obtained as a quadratic polynomial Eq. (2) in terms of temperature. The coefficients a, b, and c are presented in [22] for different orientation angles.

$$k_m = aT^2 + bT + c \tag{2}$$

Albedo, extinction, absorption, and scattering coefficients are necessary to calculate each diameter and orientation. These radiation coefficients for the desired geometry were calculated and reported in [21] based on pore-scale simulation and the two-flux model. For considering radiation in reactor simulations, the DO model was applied.

The equivalent mass diffusion coefficients for the gas species were also obtained in terms of temperature as a quadratic polynomial Eq. (3). The coefficients *d*, *e*, and *f* are reported in [20] for different orientation angles.

$$D_m = dT^2 + eT + f \tag{3}$$

The main reaction occurring in the reactor is presented in Eq. (4).

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O \tag{4}$$

However, if the combustion takes place with air in the presence of N_2, and in the absence of a catalyst, not only may NO_x be present in the combustion products, but CO is also found in significant amounts in the combustion products due to incomplete combustion.

While the literature has reported different Arrhenius coefficients to determine the catalytic combustion reaction of methane with air on Pt catalyst, Song *et al.*'s coefficients [24] showed the best agreement with experimental results [20] and were used in this study (Eq. (5)).

$$\dot{R} = A.exp\left(-\frac{E_a}{RT}\right)[CH_4]^n[O_2]^o \quad (5)$$

where $\dot{R}$ is the reaction rate, *n* and 0 are the reaction orders, and E_a is the activation energy from Song *et al.* [24]. The experimental and computational domains with assigned boundary conditions are shown in Figs. 4 and 5. This study used a cylindrical reactor with a length of 10 cm and a diameter of 6 cm for the porous medium. The fibrous porous medium in which combustion occurs was located from the position $x = 3$ cm to $x = 9$ cm, and the inlet and outlet of the reactor contained an empty space. The temperature was recorded at 9 points on the centerline of the reactor. Methane and oxygen entered the reactor after adjusting the equivalence percentage and passing through the mixing chamber. Electric elements provided the reactor's activation energy with 300 W power at the beginning of the start-up. After 30 minutes of stable operation, the elements were removed from the circuit, and the combustion reaction continued as self-progressing. The preparation and coating process of the catalyst on the fibrous porous support and more details about the experimental setup are presented in [20].

Velocity inlet and pressure outlet boundary conditions were considered at the inlet and outlet of the reactor, respectively. The mass fraction of methane and oxygen

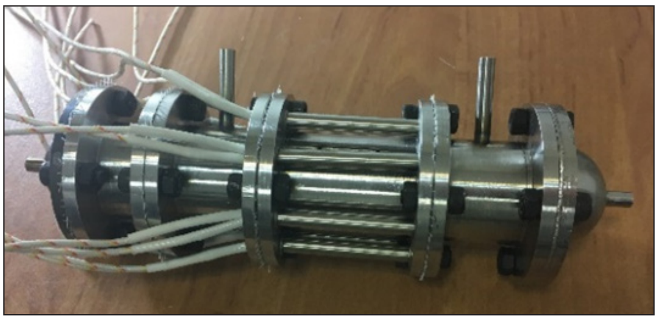

Fig. 4. The experimental reactor.

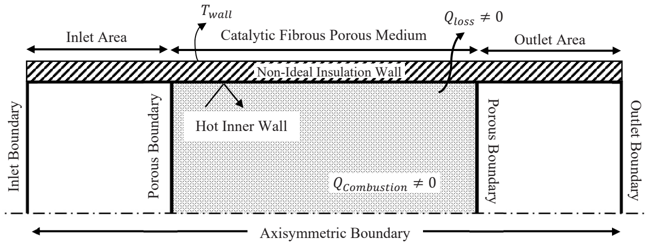

Fig. 5. The computational domain with assigned boundary conditions based on [20].

at the inlet was fixed according to the equivalence ratio. Although the walls were insulated, the boundary condition of natural convection heat transfer on the cylinder was considered due to the non-ideality of thermal insulation.

Only half of the cylinder was considered in two dimensions assuming angular and axial symmetry. A square grid was used for this simple geometry. In order to check the mesh independency, the simulation was repeated with several different meshes, including 30,000, 40,000, 90,000, and 200,000. In addition, the values of fluid velocity and methane mass fraction on the central reactor line were investigated. It was found that the difference in the results for the 40,000 and higher mesh numbers was less than 0.3%. However, the calculation time increased significantly. Hence, the solution domain with 40,000 cells was considered. The simulations were then performed using ANSYS FLUENT 17.1 software [25].

3. Results and discussion

The temperature distributions for different fibers orientation in the porous medium are shown in Fig. 6.

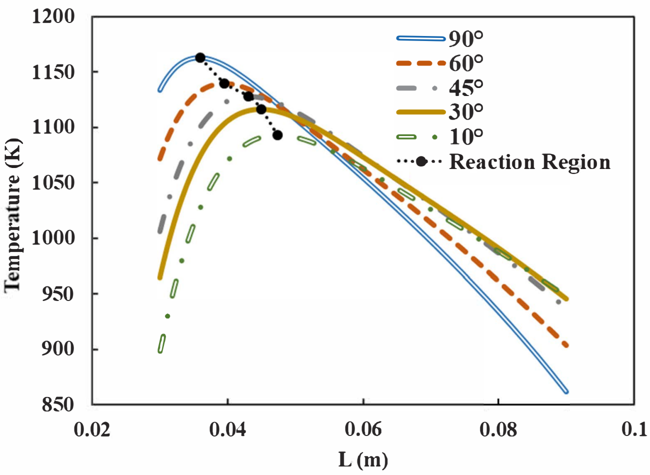

Fig. 6. Temperature distributions on the reactor's center line for five different fiber orientations at constant porosity.

As θ (the angle of fibers with the flow direction) increases, the temperature peak at which the reaction takes place moves toward the inlet of the reactor. Moreover, by changing the angle from 10° to 90°, the combustion reaction area shifts by 24.1%, a considerable change. This shift is due to the inverse correlation of θ with the permeability coefficient. By increasing this angle, the resistance to flow increases, and the flammable mixture contacts the catalytic surface closer to the beginning of the reactor. Since the nature of the combustion reaction in the fibrous porous medium is of the surface reaction type, the reaction starts wherever the reactants are placed on the catalytic surface. By increasing the angle θ and consequently increasing the fibrous obstacles in the flow direction, the reactants are placed on the surface earlier. As a result, the reaction is carried out at a shorter distance from the beginning of the reactor. This issue is shown schematically in Fig. 7.

On the other hand, a higher peak and a more non-uniform temperature distribution are observed with increasing θ. These are the main results of the inverse correlation of θ with conduction and radiation heat transfer coefficients in a fibrous porous medium.

As a result of the nature of conductive and radiative heat transfer, heat transfer will be weaker at higher angles of fibers. This issue is discussed in detail in [21,22]. The main factor of this phenomenon is the dominant influence of the thermal conductivity coefficient of alumina compared to the passing gas mixture, as well as the lower view factors of the fibers with each other at higher angles. In other words, due to the weakening of conduction and radiation heat transfer mechanisms, the longitudinal heat transfer decreases as the angle of the fiber's axis increases. Along with this change, the maximum temperature increased by 69.7 °C. This change in long-term operation leads to reduced catalyst life and increased maintenance costs.

The temperature distribution for the different diameters of the fibers in the porous medium is shown in Fig. 8. As the diameter of the fibers increase, the temperature peak moves to the end of the reactor. This movement is caused by the direct relationship between fiber diameter and permeability. As can be seen in Fig. 8, a displacement of 29.8% was obtained between the two cases d = 5 μm and d = 10 μm, which has almost the same effect as the change of orientation. This effect is assignable to the direct relationship between fiber diameter and permeability. As mentioned above, as the diameter of fibers increases in a fixed SVF, their number decreases (Eq. (1)). Therefore, by reducing the diameter of the fibers, there will be more obstacles in the way of the fluid as it passes through the porous medium. This issue leads to a decrease in the permeability of the porous medium. The reader is referred to [22] for more information about this topic.

On the other hand, a slightly higher temperature peak is observed when the diameter of the fibers is increased, and more non-uniform temperature distribution is observed. These are consequences of the inverse relationship of fiber diameter with heat transfer coefficients and radiation absorption and diffusion coefficients in a porous fibrous medium. Increasing the diameter of fibers in a fixed SVF does not change the contribution of solids in the porous medium, which has the dominant effect on heat transfer, but due to the reduction of the fibers' view factors relative to each other, the conduction and radiation heat transfer mechanisms are weakened.

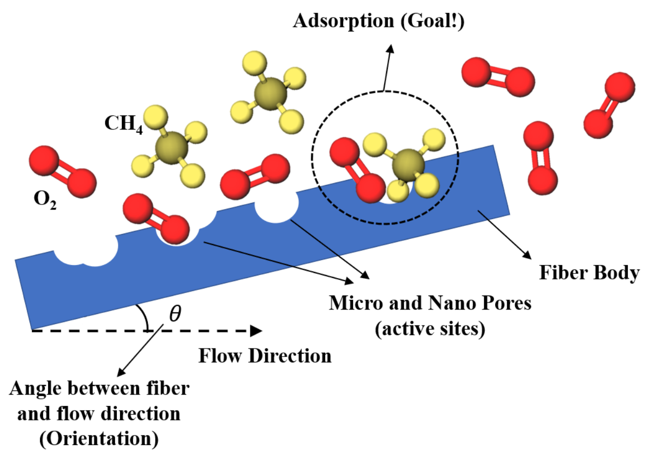

Fig. 7. Higher probability of adsorption at higher θ angles.

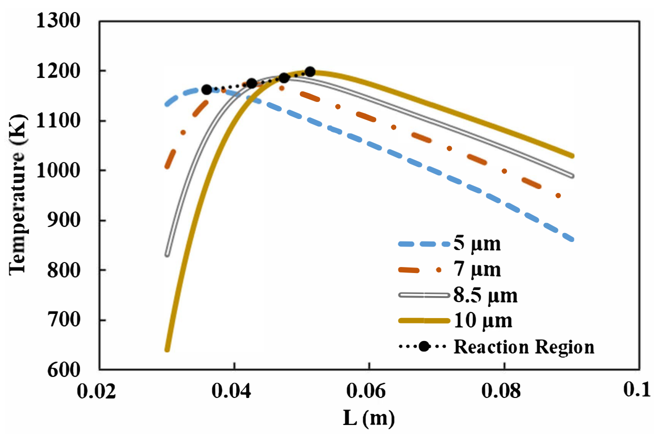

Fig. 8. Temperature distribution on the reactor's center line for four fiber diameters, 5, 7, 8.5, and 10 µm, in the constant porosity of the porous medium.

In other words, as a result of weakening the conduction and radiative heat transfer mechanism by increasing the diameter of the fibers, the heat transfer decreases in the longitudinal direction, which leads to an increase in temperature non-uniformity in the reactor and also increases the maximum temperature. The temperature change between cases d = 5 μm and d = 10 μm was equal to 34.9 °C, almost half of the effect of fibers orientation. However, this level of change affects the long-term performance of the heater and can lead to an increase in its maintenance costs.

4. Conclusions

This study investigated the effects of fiber diameter and orientation in the fibrous porous medium on the temperature distribution and the catalytic reaction zone. Flow, conduction, radiation heat transfer, and mass transfer coefficients were extracted from our previous studies using pore-scale simulations and used in a two-dimensional premixed reactor computational domain. The results showed that by increasing the fibers' angle from 10° to 90°, the maximum temperature in the reactor increases by 69.7 °C, and the reaction zone moves 24.1% toward the inlet. Moreover, more uniform temperature distributions were observed at the lower angles. In addition, the results showed that by increasing the diameter of the fibers from 5 to 10 μm, the maximum temperature in the reactor increases by 34.9 °C, and the reaction site moves 29.8% towards its end. Also, a more uniform distribution was observed in the reactor with smaller diameter fibers.

In the range of changes investigated for the diameter and orientation of the fibers, the effect of orientation on the temperature peak was twice the diameter. Also, the effect of changing both parameters on the reaction zone was almost the same. This issue is very significant for the production process. In other words, choosing a production process that results in a lower total cost is possible with the proper selection of these two parameters. In addition, the temperature peak in the reactor is critical in the design of catalytic heaters. The higher the maximum temperature, due to the formation of hot spots in the porous medium, the more the heater will require servicing and replacement of catalysts at shorter intervals. Also, a temperature sensor to measure the reactor center's temperature will be required to produce this type of heater. Due to the diameter and orientation of the fiber used in the heater, the temperature measurement accuracy depends entirely on the location of the sensor installation. The wrong choice of the measurement location leads to the wrong estimation of the service intervals and, consequently, an increase in the maintenance costs.

The results of this research can be used in optimizing the manufacturing procedures to produce a fibrous porous medium that suits specific application needs.

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