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Computational fluid dynamics modeling of the combustion and emissions characteristics in high-temperature catalytic micro-combustors

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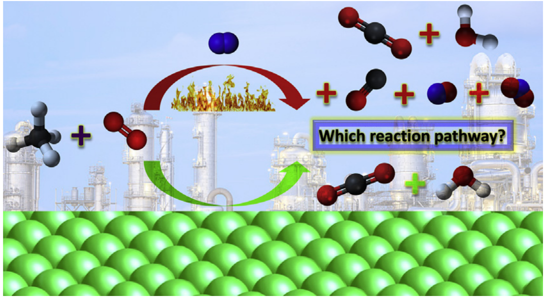
HIGHLIGHTS

- The effect of operating conditions on the products distribution was studied.
- The reaction pathways involved in the combustion process were identified.
- The mechanism responsible for the formation of pollutants was clarified.
- Catalytic combustion can effectively reduce the emission of pollutants.
- Products distribution diagrams were constructed and design guidance was provided.

GRAPHICAL ABSTRACT



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ABSTRACT

The combustion and emissions characteristics of methane-air mixtures in catalytic micro-combustors were studied in high-temperature environment where both heterogeneous and homogeneous reactions can occur simultaneously. Special emphasis was placed on understanding the role of each reaction mechanism in determining the distribution of combustion products. Computational fluid dynamics simulations were performed under various operating conditions to clarify the mechanism responsible for the formation of pollutants during the catalytic combustion process. Comparisons were made between the results obtained for different reaction pathways. Distribution diagrams of the combustion products were constructed and design recommendations were made. It was shown that catalytic combustion can effectively reduce the emission of pollutants, even though the system is operated within the homogeneous flammability limits. There is a strong interplay between heterogeneous and homogeneous chemistry during the combustion process, and their individual contributions depend critically on the operating conditions. Heterogeneous chemistry can significantly inhibit homogeneous chemistry, thus reducing the formation of pollutants and improving the distribution of products. The distribution of products can also be controlled by adjusting the design parameters such as pressure, temperature, feed composition, and combustor dimension. Nitric oxide is the main nitrogen pollutant formed in small-scale combustion systems under lean-burn conditions.

1. Introduction

Micro-combustors, typical dimensions in the sub-millimeter range, are emerging as a powerful tool for portable production of energy [1,2]. Due to the high energy density of hydrocarbons, micro-combustors may eventually replace expensive lithium-ion batteries in portable energy devices [3,4]. Furthermore, due to their inherently higher heat-transfer coefficients, micro-combustors can be efficient heat sources for the

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Nomenclature

A	pre-exponential factor, [mol, cm, s], Table 2
A'	surface area, m^2, Eq. (10)
c_p	heat capacity at constant pressure, J/(kg·K), Eq. (7)
C	molar concentration, mol/m^2, Eq. (12)
d	gap distance between the plates, m, Fig. 1 and Table 1
d_{pore}	average pore diameter, m, Eq. (15) and Table 1
D	diffusivity, m^2/s, Eq. (6)
D_{eff}	effective diffusivity, m^2/s, Eq. (12)
D_m	mixture-averaged diffusivity, m^2/s, Eq. (6)
D^T	thermal diffusivity, m^2/s, Eq. (6)
Ea	activation energy, kJ/mol, Table 2
F	area ratio, m^2/m^2, Eq. (10)
h	specific enthalpy, J/kg, Eq. (7)
$k_{adsorption}$	adsorption rate constant, Eq. (16)
k_{f_r}	rate coefficient of the forward reaction k, Table 2
K	number of species, Eq. (4)
l	length, Table 1
m	total number of species, Eq. (8)
p	pressure, Pa, Eq. (2)
R	ideal gas constant, J/(mol·K), Eq. (7)
s	sticking coefficient, Table 2
$\dot{s}$	rate of appearance of a heterogeneous product, mol/(m^2·s), Eq. (8)
T, T_o	absolute temperature, reference temperature, K, (4) and (7)
u, v	streamwise velocity component, transverse velocity component, m/s, Eq. (1)
V,$\vec{V}$	diffusion velocity, diffusion velocity vector, m/s, Eqs. (5) and (6)
W,$\bar{W}$	relative molecular mass, relative molecular mass of the gas mixture, dimensionless, Eqs. (5) and (6)
x, y	streamwise coordinate, transverse coordinate, Eq. (1) and Fig. 1
Y	mass fraction, Eq. (4)

Greek variables

β	temperature exponent, Table 2
Γ	surface site density, mol/m^2, Eq. (8)
γ	specific surface area of the washcoat, m^2/m^3, Eq. (12)
δ	thickness, m, Eq. (12)
ε_i	dependence of the rate coefficients on the surface coverage of species i, kJ/mol, Table 2
ε_p	porosity, dimensionless, Eq. (14) and Table 1
η	effectiveness factor, Eq. (9)
θ_{free}	free-surface site fraction, Eq. (16)
Θ	surface coverage, Table 2
λ	thermal conductivity, W/(m·K), Eq. (4)
μ	dynamic viscosity, kg/(m·s), Eq. (2)
μ_i	dependence of the rate coefficients on the surface coverage of species i, Table 2
ρ	density, kg/m^3, Eq. (1)
σ	site occupancy, Eq. (8)
τ_p	tortuosity factor, dimensionless, Eq. (14) and Table 1
Φ	Thiele modulus, dimensionless, Eq. (11)
$\dot{\omega}$	rate of appearance of a homogeneous product, mol/(m^3·s), Eq. (5)

Subscripts

eff	effective, Eq. (11)
g	gas, Eq. (4)
i	species index, Eq. (11)
in	inlet, Table 1
k, m	gaseous species index, surface species index, Eqs. (4) and (8)
x, y	streamwise component, transverse component, Eq. (4)

production of hydrogen by steam reforming [5–7] and ammonia decomposition [8,9] in integrated micro-chemical systems for fuel cell applications.

Recent efforts have attempted to utilize homogeneous combustion of hydrocarbon fuels in portable energy devices to directly produce heat or power [1–4]. Unfortunately, the high temperatures generated through homogeneous combustion present significant challenges for many practical applications [10–13]. Furthermore, homogeneous flames are typically quenched when confined within spaces with sub-millimeter dimensions due to thermal and radical quenching [14]. In contrast, catalytic combustion appears to be a promising alternative to conventional homogeneous combustion [15–18]. The lower temperatures generated through catalytic combustion can significantly widen the operating window of portable energy devices [19,20]. Additionally, catalytic combustion can be a very effective way to reduce the emission of pollutants. Furthermore, catalytic combustion can simplify the design [19–22] and greatly improve the performance of the system [23,24].

Catalytic micro-combustors show great promise in many applications ranging from the production of syngas [25,26] to combustion [27–30], since complete conversion can be achieved at residence times as short as several milliseconds [31–34]. The basic principles of catalytic combustion have been addressed in past reviews [16,17]. At higher temperatures, there is a significant contribution from homogeneous reactions to the overall reaction-rate in parallel to heterogeneous reactions. In most cases, the occurrence of homogeneous reactions is an undesired feature in catalytic combustion, because it complicates the fundamental understanding of reaction mechanisms and results in selectivity losses.

The presence of a catalyst can expand the flammability limits and operation windows for hydrocarbon fuels [31,32]. It can also significantly reduce the emission of pollutants during the combustion process [31,32], which is of great importance due to the potential applications of this technology in both gas turbine and internal combustion engine systems [35,36]. However, the effect of catalysts is still poorly understood in the operating regime, in which heterogeneous and homogeneous reactions can occur simultaneously. It is therefore of great interest to clarify the role of heterogeneous and homogeneous reaction pathways under these conditions in order to better understand the mechanism responsible for the formation of pollutants.

Modeling these catalytic systems is needed both to better understand the factors affecting the distribution of products produced during the combustion process. To provide an accurate description of the operation of these systems, a two-dimensional model with detailed heat and mass transfer is necessary [37–40]. The importance of heat transfer in small scale catalytic combustion systems has been highlighted in the literature [41,42]. Computational fluid dynamics modeling and simulation also play an important role in the design and operation of these catalytic systems [43,44], and thereby the interplay between kinetics and transport in combustion characteristics can be illustrated. Since the small dimensions encountered in catalytic micro-combustors render spatially resolved measurements within a system difficult, detailed modeling and simulation are invaluable in elucidating the mechanism underlying the combustion process and assisting in the design of these catalytic systems.

Due to the complexity involved in a computational fluid dynamics