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Transport effects on pattern formation and maximum temperature in homogeneous–heterogeneous combustion

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HIGHLIGHTS

- Maximum temperature rise in coupled homogeneous-catalytic combustion is analyzed.
- Impact of transport parameters such as Lewis and Peclet numbers is studied.
- Turing patterns as well as transport limited patterns are shown to be possible.
- Parameter regions for which 3-D solutions can exist are identified.

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ABSTRACT

We study the impact of the Lewis number, Le_f (thermal diffusivity of the reaction mixture to the molecular diffusivity of the limiting reactant) and the Peclet numbers on the maximum temperature attained for coupled homogeneous–heterogeneous combustion process in a parallel plate reactor using one, two and three-dimensional models. For the case of 1-D models, we find that the maximum temperature never exceeds the adiabatic value for physically consistent boundary conditions. For 2-D models, we find that for $Le_f < 1$, the hot spot temperature can exceed the adiabatic value, it is always located on the wall and its distance from the inlet and magnitude increase with increasing radial Peclet number. However, for $Le_f > 1$, contrary to some literature claims (Zheng and Mantzaras, 2014), the peak temperature never exceeds the adiabatic value, though the temperature can be non-monotontic across the channel. We show that 3-D solutions can bifurcate either from 1-D or 2-D solutions irrespective of the value of the Lewis number. It is also shown that an infinite number of solutions that are discontinuous in the axial coordinate can exist for the common case of large axial heat Peclet number. The implications of these observations for catalyst and process design in systems in which both homogeneous and catalytic reactions occur are discussed.

1. Introduction and literature review

Catalytic combustion technologies have been widely studied due to their promise of meeting future energy demands and production of intermediate chemicals. The models describing catalytic combustion and catalytic partial oxidations typically involve both catalytic and homogeneous reactions (see for example, references [2–6]). Homogeneous ignition in catalytic combustion has been investigated in various settings such as stagnation point flows, external boundary layer flows and two-dimensional channel flows (for a good overview of the subject see Hayes and Kolaczkowski [7]). However, most of these studies have been numerical, mostly relying on CFD packages. Multiple ignitions and extinctions are possible for such thermally coupled systems, and a direct numerical study can obscure the essential features as multiple reactions and space dimensions may detract from the underlying physics. Our previous work [8] using 0-D two-mode model showed that in typical hydrocarbon oxidations, the first ignition-extinction pair is due to the catalytic reaction alone, while the thermally coupled ignition and extinction only come on the scene when the catalytic reaction is in the mass transfer controlled regime. Further, when the inlet temperature is sufficiently high, the onset of a second ignition and extinction for a thermally coupled system (with a finite wall reaction rate) and a system with infinitely fast catalytic reaction is practically indistinguishable. The case of homogeneous

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Notation

Roman letters

$\langle c \rangle$	cross-section averaged concentration
C_{pf}	fluid phase heat capacity
D_m	diffusion coefficient in the fluid phase
Da	Damköhler number for the homogeneous reaction
Da_s	Damköhler number for the catalytic reaction
E_a	activation energy for the homogeneous reaction
E'_a	activation energy for the catalytic reaction
k	wavenumber
k_v	first order rate constant for the homogeneous reaction
k_s	first order rate constant for the catalytic reaction
L	length of the channel
a	spacing along the first transverse direction
b	spacing along the second transverse direction
Le_f	fluid Lewis number
P	transverse Peclet number
Pe	axial Peclet number
Pe_r	radial Peclet number
T	fluid temperature
T_{in}	inlet fluid temperature
$\overline{u}$	average fluid velocity in the channel
x'	coordinate along the length of the channel (dimensional)
y'	first transverse coordinate (dimensional)
z'	second transverse coordinate (dimensional)
x	coordinate along the length of the channel (dimensionless)
y	first transverse coordinate (dimensionless)
z	second transverse coordinate (dimensionless)
X	dimensionless coordinate along the length of the channel normalized with the transverse Peclet number

Greek letters

ρ	density
α	aspect ratio
β	dimensionless adiabatic temperature rise
γ	dimensionless activation energy for the homogeneous reaction
γ_c	dimensionless activation energy for the catalytic reaction
ζ	dimensionless coordinate along the length of the channel normalized with the axial Peclet number
θ	dimensionless temperature
$\langle \theta \rangle$	cross-section averaged temperature
θ_m	dimensionless mixing-cup temperature
θ_s	dimensionless temperature at the surface
ϕ	Thiele modulus for the homogeneous reaction
ϕ_s	Thiele modulus for the catalytic reaction
Λ	ratio of $(Da + Da_s)$ and Pe

Subscripts and superscripts

f	fluid phase
s	solid phase
m	cup-mixing
$*$	potential
E	excess

reaction in the fluid phase with an infinitely fast wall reaction has not been analyzed in detail in literature and is ripe for a theoretical analysis. The present article focuses on this limiting case (as well as other limiting cases) with an emphasis on the location of the hot spot, which is crucial for determining the reactor thermal stability. The Lewis number of the limiting reactant is an important parameter that dictates the trends in the maximum temperature. Other parameters such as the Thiele moduli and Peclet numbers (axial, transverse and radial) also play a significant role. The effect of Lewis number (Le_f) on the surface temperature has been studied by various investigators (e.g. Satterfield et. al. [9], Hegedus [10], Pfefferle and Pfefferle [11,12]) when only catalytic reaction is present and in the combustion literature when only the homogeneous reaction is present. When Lewis number is less than unity, the catalyst surface receives the limiting reactant faster than it can expel the heat produced due to the reaction. This situation leads to superadiabatic surface temperatures, the existence of which has been reported and verified in the literature [9,10]. Even though numerical simulations have been done demonstrating the effects of Lewis numbers greater than and less than unity on the maximum temperature in the solid phase [9–11,13], there have not been many studies which also study the effects on the gas phase temperatures. The recent work of Zheng and Mantzaras [1] reports that in systems with fast catalytic and potential homogeneous reaction, the temperature within the reactor may exceed adiabatic values for Lewis numbers greater than 1. This has been presented as a novel and significant result. However, the analysis presented in this work demonstrates that these claims are questionable. We study a hierarchy of models asking the question: what is the maximum temperature attainable for the model and when is it achieved? We find that physically consistent 1-D and 2-D models do not lead to gas phase temperatures exceeding the adiabatic for $Le_f > 1$. We also examine the stability of 1-D/2-D solutions to 3-D perturbations and show that 3-D solutions can exist in coupled homogeneous–heterogeneous reactions, though under extreme conditions (high adiabatic temperature rise and short contact times).

In the next section, we formulate a 3-D mathematical model that describes the thermally coupled homogeneous–heterogeneous combustion in a parallel plate channel. We also formulate various limiting cases of this model that are amenable to analysis. In the following sections, the limiting models are analyzed to provide insight on the impact of various transport parameters representing the heat and mass transfer phenomena on coupled homogeneous–heterogeneous combustion. We provide the theory and analytical expressions for concentration and temperature fields in two-dimensional domains, and in limiting one dimensional models. Finally, we examine the stability of 1-D/2-D solutions to 3-D perturbations and discuss the various types of solutions that could exist or coexist in the system. The implications of 3-D solutions and the maximum temperature for catalyst and process design are discussed in the last section.

2. Mathematical models

We consider a system consisting of flow between parallel plate reactors in which homogeneous reaction occurs in the fluid phase and catalytic reaction on the wall. For simplicity, the following assumption are made: (i) the velocity profile is invariant with axial position and the pressure drop is negligible so that the flow field can be decoupled from the species and energy balances (ii) one reactant (either fuel or oxygen) is in excess and the reaction rate is linear in the limiting reactant (iii) the physical properties can be assumed constant or taken to be average values without impacting the qualitative features (iv) The system is adiabatic.