Combustion and Flame 161 (2014) 1911–1922

Contents lists available at ScienceDirect

Combustion and Flame

journal homepage: www.elsevier.com/locate/combustflame

An analytical and numerical investigation of hetero-/homogeneous combustion with deficient reactants having larger than unity Lewis numbers

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A R T I C L E   I N F O

Article history:
Received 27 September 2013
Received in revised form 17 December 2013
Accepted 19 December 2013
Available online 11 January 2014

Keywords:
Hetero-/homogeneous combustion
Effects of larger than unity Lewis number of deficient reactant on hetero-/homogeneous combustion
Gas-phase superadiabaticity
Reactor thermal management

A B S T R A C T

The heterogeneous combustion and the combined hetero-/homogeneous combustion of deficient reactants with Lewis numbers (*Le*) larger than unity was investigated analytically and numerically in two geometrical configurations, the flat plate and the planar channel, under the condition of infinitely fast catalytic chemistry. Analytical results based on similarity solutions for the catalytic flat plate and on heat and mass transfer solutions for the channel were complemented by detailed 2-D numerical simulations. The larger than unity Lewis number led to the underadiabatic surface temperatures, which in turn gave rise to gas-phase regions with local energy excess. For the flat plate case, the maximum gas-phase energy excess was a non-monotonic function of the Lewis number. The peak occurred at $Le = 6.5$ with a corresponding energy excess 6.4% above the total energy of the fresh reactants. In channel-flow combustion, the maximum gas-phase energy excess was a monotonically increasing function of Lewis number, approaching asymptotically the considerably higher value of 20.8% as $Le \rightarrow \infty$. For current catalytic combustion methodologies, which include the fuel-lean hydrocarbon/air combustion (Lewis numbers of deficient hydrocarbon fuels up to ~3.2) and the fuel-rich hydrogen/air combustion (Lewis number of deficient oxygen ~2.3), the energy excess in the gas was significant and could reach up to 14%. Hetero-/homogeneous combustion simulations have shown that, upon homogeneous ignition, the gas-phase energy excess manifested itself with superadiabatic flame temperatures. However, the superadiabaticity in the gas was confined to the channel core, such that the surface temperature did not exceed the adiabatic equilibrium temperature. This behavior had key implications for the reactor thermal management and catalyst stability. Moreover, the gas-phase superadiabaticity led to peak prompt NO_x values 30% higher than those achieved by a diffusionally neutral deficient reactant ($Le = 1$).

1. Introduction

Catalytic combustion methodologies have attracted increased attention in large-scale power generation as a means to reduce NO_x emissions and improve combustion stability [1–3]. In the conventional fuel-lean catalytically stabilized thermal combustion (CST), fractional conversion of hydrocarbon fuels is achieved in a heterogeneous (catalytic) reactor, while the remaining fuel is combusted in a post-catalyst homogeneous (gas-phase) burnout zone, again at fuel-lean stoichiometries [4,5]. Alternative to the fuel-lean CST is the more recent catalytic-rich/gaseous-lean combustion approach [6,7]. Therein, a fraction of the air and all of the hydrocarbon fuel react at fuel-rich stoichiometries in a catalytic partial oxidation (CPO) reactor [8], producing syngas (a mixture comprising mostly CO and H_2). Syngas is afterwards mixed with the remaining (bypass) air, forming an overall fuel-lean homogeneous combustion zone. This methodology has several advantages compared to the conventional fuel-lean CST, which include extended extinction limits [6,7,9] and enhanced stability of the follow-up flame due to the highly-reactive hydrogen contained in the syngas mixture [10].

The aforementioned fuel-lean and fuel-rich catalytic combustion methodologies are currently investigated not only for hydrocarbons but also for syngas or pure hydrogen fuels. This is driven by new pre-combustion CO_2 capture strategies relevant to integrated gasification combined cycle (IGCC) power plants. Therein, hydrogen-rich syngases are produced via solid or liquid fossil fuel decarbonization, leading to pre-combustion capture of CO_2 [11]. Apart from solid and liquid fuels, decarbonization can also be economically applied to natural-gas-fired power plants [12]. The fuel-lean CST methodology for hydrogen or syngas is essentially the same as the corresponding one for hydrocarbons. However, in the fuel-rich approach with hydrogen and syngas, the catalyst does

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http://dx.doi.org/10.1016/j.combustflame.2013.12.018

Nomenclature

b	channel half-height, Fig. 6
$c_{p,k}$	specific heat at constant pressure of k-th species, Eq. (A.6)
D_F	diffusivity of fuel
d_h	channel hydraulic diameter, $d_h = 4b$
f	normalized stream function, Eq. (6)
h, h_k^o	total enthalpy, chemical enthalpy of k-th species, Eqs. (A.4) and (A.6)
K_g	total number of gaseous species, Eqs. (A.4) and (A.5)
Le	Lewis number (= Sc/Pr)
p	pressure
Pr	Prandtl number
Q	heat release per unit mass of fuel, Eq. (18)
R	universal gas constant
Re	channel Reynolds number based on d_h.
s	scaled streamwise distance, Eq. (4)
$\dot{s}_k$	catalytic molar production rate of k-th species, Eqs. (A.7) and (A.8)
Sc	Schmidt number
T, T_o	temperature and reference temperature, Eq. (A.6)
T^*	normalized gas temperature for flat plate, Eq. (5)
$\tilde{T}$	normalized gas temperature for channel, Eq. (17)
T_{ad}	adiabatic equilibrium temperature of fresh mixture
T_{eq}	local equilibrium temperature, Eq. (14)
$\tilde{T}_{eq}$	local energy excess for flat plate, Eq. (15), and for channel Eq. (22)
u, U_{IN}	streamwise velocity component, inlet streamwise velocity
v	transverse velocity component
$\vec{V}_k$	species diffusion velocity vector, Eqs. (A.4) and (A.5)
$W_k, \overline{W}$	molecular weight of k-th species, average molecular weight
Y_k	mass fraction of k-th species
Y^*	normalized mass fraction for flat plate, Eq. (5)
$\tilde{Y}$	normalized mass fraction for channel, Eq. (17)
x, y	streamwise and transverse coordinates, Figs. 1 and 6
x^+	non-dimensional streamwise distance, $x/(d_h Re Sc)$

Greek symbols

ΔT_c	adiabatic combustion temperature rise, Eqs. (12) and (18)
η	similarity variable for flat plate, Eq. (4)
λ	thermal conductivity of gas, Eq. (A.4)
μ	dynamic viscosity, Eq. (A.2)
ν'_k, ν''_k	stoichiometric coefficients of k-th species in reactants and products
ρ	density
Ψ	stream function
$\dot{\omega}_k$	homogeneous molar production rate of k-th species, Eq. (A.5)

Subscripts

ad	adiabatic equilibrium
c	channel center
F	fuel
IN	inlet of channel
m	mean value in channel (average over channel height)
W	wall
x, y	streamwise and transverse components
∞	free-stream in flat plate

not have a prime CPO function (at least for syngas mixtures with a low hydrocarbon content) but rather acts as a preheater and stabilizer for the subsequent gas-phase combustion zone [6,13,14]. Finally, apart from large-scale power generation, catalytic combustion of low hydrocarbons and hydrogen is also of interest in microreactors for portable power generation – albeit mostly within the fuel-lean combustion methodology [15–17].

Of fundamental importance for reactor thermal management and catalyst thermal stability is the Lewis number of the deficient reactant. Excursions of the catalyst surface temperature above or below the adiabatic equilibrium temperature (with the latter based on the incoming mixture stoichiometry and temperature) were reported in early catalytic combustion studies [18,19]. Such temperature excursions actually reflect the specific nature of catalytic reactions. In heterogeneous combustion, a degenerate diffusion reaction sheet is formed [20] with this terminology reflecting the fixed location of the reaction zone (catalyst surface). The inability of the catalytic reaction zone to adjust its position (in contrast to a gaseous diffusion flame that freely adjusts its position) leads in turn to temperatures markedly different form the adiabatic equilibrium temperature [21–23]. For diffusionally imbalanced deficient reactants with Lewis numbers less than unity (notably hydrogen, whereby at lean stoichiometries in air $Le_{H2} \sim 0.3$), CST is compounded by the attainment of surface temperatures exceeding the adiabatic equilibrium temperature by hundreds of degrees [22–27]. In these cases, fuel is transported more efficiently to the catalyst than heat away from it, leading to the observed superadiabatic surface temperatures. While in practical catalytic reactors other factors can also contribute to surface temperature superadiabaticity (e.g. heat recirculation in the solid reactor structure), for deficient reactants with sufficiently low Lewis numbers the dominant contributor of surface superadiabaticity is still the diffusional imbalance. Conversely, for limiting reactants with Lewis numbers larger than unity, i.e. fuel-lean catalytic combustion (CST) of C2+ hydrocarbons or fuel-rich catalytic combustion of hydrogen in air (whereby the limiting reactant is oxygen with $Le_{O2} \sim 2.3$), underadiabatic surface temperatures are attained over much of the reactor length [14,28,29].

The impact of varying Lewis number of the deficient reactant on the catalyst surface temperature has been reported in past studies. Satterfield et al. [18] analyzed the effect of different mass and heat transport rates on the catalyst surface temperature of tubular channels, while Hegedus [19] presented solutions for underadiabatic and superadiabatic surface temperatures in monolithic structures as a function of lumped geometrical parameters and the diffusivity of the limiting reactant. Subsequently, numerical solutions of the surface temperatures in catalytic channels for deficient reactant Lewis numbers larger or smaller than unity were reported in [22,23,27,28,30]. Quantification of the attained surface temperatures as a function of Lewis number was of prime concern in earlier studies, since theoretical work could be facilitated with simplified 1-D lumped heat and mass transfer models. However, as will be shown in the present work, surface temperature excursions inherently lead to opposite excursions in the gas-phase. For example, underadiabatic surface temperatures lead to excess enthalpy in the gas-phase, which upon homogeneous ignition would result in superadiabatic flame temperatures. These effects have not been addressed in the literature, probably as the main concern was initially placed on the surface temperatures themselves due to reactor material and catalyst thermal stability considerations. An