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Introduction

There is continuing interest in catalytic combustion for numerous applications. Current research efforts intend to clarify the heteroneneous kinetics, as well as the different combustion modes that appear as the chemistry interacts with the outer premixed flow, where homogeneous reactions may also occur simultaneously if the existing temperature is sufficiently high. The stagnation-point flow considered here, formed when a premixed jet of fuel and air impinges at a right angle on a catalytic plate, is often selected in experimental and theoretical investigations (see, e.g., [1]–[8] and the early work cited in [1]).

In many applications of catalytic combustion, including for instance the Passive Auto-catalytic Recombiners (PAR) presently used as a mitigation tool in nuclear power plants, chemical reactions occur solely on the catalyst surface, while the outer premixed flow remains chemically frozen. This is typically the case for low and moderate values of the peak temperature, e.g., $T_{\text{PEAK}} \lesssim 1000$ K for hydrogen-oxygen combustion at atmospheric pressure [2, 3, 6]. In the resulting problem, the outer frozen flow and the heterogeneous combustion process are coupled through the fluxes of heat and species at the catalyst surface, so that, in principle, one has to solve simultaneously a set of algebraic equations for the heterogeneous chemistry together with a set of differential equations for the outer flow. It is shown below that, by rescaling appropriately the mass fractions and temperature of the gas stream, the outer flow can be integrated independently, providing simple expressions for the fluxes of species and heat at the plate in terms of the plate temperature. These expressions replace the integration of the conservation equations in the gas phase, thereby reducing the problem to the solution of a set of nonlinear algebraic equations.

Simplified Formulation

The heterogeneous combustion process occurs according to the overall reaction $F + \nu_{O_2}O_2 \rightarrow \nu_P P + q$, where q , ν_{O_2} and ν_P are, respectively, the amount of heat released, the number of moles of oxygen consumed and the number of moles of products produced per mole of fuel consumed. Hence, the effect of the catalytic surface on the outer flow is that of a sink for reactants and a source for heat and combustion products. The rate ω of the above reaction depends in general on the detailed surface kinetics, different for different fuels and for different catalysts and typically involving a number of surface reactions along with adsorption and desorption reactions. The resulting value of ω is a function of the relative surface coverages of reactants, products and intermediates, which must be determined as part of the solution.

The surface kinetics depends also on the plate temperature T_p as well as on the mass fractions of fuel, oxygen and products found in the gas phase at the plate Y_{F_p} , $Y_{O_2_p}$, and Y_{P_p} . To compute these values, one needs to solve the conduction-reaction balance equation at the plate surface

$$\frac{\mu c_p}{Pr} \left(\frac{\partial T}{\partial y} \right)_p = q\omega + P \quad (1)$$

together with the associated diffusion-reaction balance equations

$$\frac{\mu}{W_F L_F Pr} \left(\frac{\partial Y_F}{\partial y} \right)_p = \frac{\mu}{\nu_{O_2} W_{O_2} L_{O_2} Pr} \left(\frac{\partial Y_{O_2}}{\partial y} \right)_p = -\frac{\mu}{\nu_P W_P L_P Pr} \left(\frac{\partial Y_P}{\partial y} \right)_p = \omega. \quad (2)$$

In the formulation, y represents the distance to the plate, with the subscript p denoting conditions at $y = 0$, Pr is the Prandtl number, W_i and L_i are the molecular weight and the Lewis number of species i , and μ is the viscosity. For generality, a source term P has been included in (1). This additional term is negative if heat losses are accounted for, is positive in configurations with external heating and is identically zero for an adiabatic plate.

Since the surface gradients $(\partial T/\partial y)_p$ and $(\partial Y_i/\partial y)_p$ depend on the competition of diffusion and convection in the approaching stream, the problem requires in general to solve the steady boundary-layer conservation equations for the stagnation-point flow. These equations can be conveniently written by introducing the mass-weighted coordinate $\eta = (2^j a \rho_\infty / \mu_\infty)^{1/2} \int_0^y (\rho / \rho_\infty) dy$, and stream function $f(\eta)$ defined such that $u = arf'$ and $v = -(2^j \rho_\infty \mu_\infty a)^{1/2} f / \rho$ are the radial and axial velocity components, with r denoting the radial coordinate. Here, ρ represents the density of the gas mixture and $2^j a$ is the rate of strain imposed on the flow. Both planar ($j = 0$) and axisymmetric ($j = 1$) configurations are considered in the analysis for completeness. Introducing the rescaled variables

$\theta = (T - T_\infty)/(T_p - T_\infty)$ and $\theta_i = (Y_i - Y_{i\infty})/(Y_{i_p} - Y_{i\infty})$ reduces the problem to that of integrating [9, 10]

$$f''' + ff'' + (\rho_\infty/\rho - f'^2)/2^j = 0 \quad (3)$$

$$(c_p\theta')' + Pr c_p f\theta' + \sum c_{p_i}(Y_{i_p} - Y_{i\infty})\theta'_i\theta'/L_i = 0, \quad (4)$$

$$\theta''_i + L_i Pr f\theta'_i = 0, \quad (5)$$

with boundary conditions

$$\begin{cases} \eta = \infty : & f' - 1 = \theta_i = \theta = 0, \\ \eta = 0 : & f = f' = \theta_i - 1 = \theta - 1 = 0 \end{cases} \quad (6)$$

In writing the above equations, a constant value $\rho\mu = \rho_\infty\mu_\infty$ has been assumed, and changes in the transport numbers Pr and L_i have been neglected [2]. In general, the specific heat at constant pressure must be computed in the form $c_p = \sum_i c_{p_i}Y_i$, where $c_{p_i}(T)$ is the specific heat of species i .

In principle, the values of Y_{i_p} and T_p must be specified for the numerical integration of (3)–(6). The solution can be simplified by neglecting changes in c_p along with changes in mean molecular weight. With these additional assumptions, which are often used in the literature [1], Eq. (4) reduces to $\theta'' + Pr f\theta' = 0$, while the density ratio appearing in Eq. (3) becomes $\rho_\infty/\rho = 1 + \gamma\theta$, where

$$\gamma = (T_p - T_\infty)/T_\infty \quad (7)$$

is the dimensionless temperature increase. The numerical integration of the resulting gas-phase conservation equations for different values of γ , the only unknown parameter left in the formulation, provides the normalized surface gradients $\Theta = -(\theta')_p$ and $\Theta_i = -(\theta'_i)_p$ needed in (1) and (2). Sample values of the resulting functions $\Theta_i(\gamma, L_i)$ obtained with $Pr = 0.7$ and $j = 1$ are given in Fig. 1.

It is worth noting that the simplifying assumptions of constant c_p and constant molecular weight used here were checked to introduce negligibly small errors in surface gradients. For instance, for a mixture of hydrogen and air ($\phi = 0.5$) with $T_p = 1000$ K, $T_\infty = 298$ K and complete fuel consumption at $\eta = 0$ the surface gradients obtained from the complete boundary-layer equations (3)–(5) differ by less than 1.5 % from those computed with the aforementioned simplifications. Furthermore, it was seen that, in the ranges of practical applicability $0 \leq \gamma \leq 5$ and $0.3 \leq L_i \leq 2.0$, the dependence of the surface gradients on γ

and L_i can be expressed with errors smaller than 3 % through the simple expressions

$$\Theta_i = 0.08 + 0.42L_i^{1/2} - (0.005 - 0.0045L_i^{1/2})\gamma - 0.0024L_i^{1/2}\gamma^2, \quad (j = 0) \quad (8)$$

$$\Theta_i = 0.08 + 0.39L_i^{1/2} - (0.004 - 0.0035L_i^{1/2})\gamma - 0.0017L_i^{1/2}\gamma^2, \quad (j = 1)$$

where a value $L_i = 1$ must be used to compute the temperature gradient Θ . The degree of accuracy is illustrated in Fig. 1. Use of (8) allows us to write (1) in the form

$$\gamma\Theta(\gamma) = \frac{Pr(q\omega + P)}{c_p T_\infty (2^j \rho_\infty \mu_\infty a)^{1/2}}, \quad (9)$$

while the diffusion-reaction balance equations become

$$\begin{aligned} \frac{(Y_{F_\infty} - Y_{F_p})}{W_F L_F} \Theta_F(\gamma, L_F) &= \frac{(Y_{O_{2\infty}} - Y_{O_{2p}})}{\nu_{O_2} W_{O_2} L_{O_2}} \Theta_{O_2}(\gamma, L_{O_2}) \\ &= \frac{(Y_{F_p} - Y_{F_\infty})}{\nu_P W_P L_P} \Theta_P(\gamma, L_P) = \frac{Pr \omega}{(2^j \rho_\infty \mu_\infty a)^{1/2}}. \end{aligned} \quad (10)$$

Conclusions

Equations (9) and (10), supplemented with (8) and with a set of algebraic equations for the surface kinetics, determine the solution for Y_{i_p} , T_p and ω , giving a formulation that is attractive for numerical and analytical studies of heterogeneous combustion. The resulting problem is particularly simple in computations where the catalytic rate ω is determined from an explicit expression in terms of Y_{i_p} and T_p [1, 2]. As an illustrative example, let us consider catalytic combustion of propane, whose bifurcation behavior was analyzed in [1] by a shooting method with a stable integration algorithm suited for stiff problems. Using γ as a control parameter and the rate ω employed in [1], Eq. (10) provides a simple cubic equation for Y_{F_p} , whose solution can be substituted in (9) to yield the source term P . The heterogeneous ignition and extinction temperatures then follow from the solution to $dP/d\gamma = 0$. In this way, the complete bifurcation curves given in [1], whose computation required several minutes on a CRAY II supercomputer, can be determined in a few seconds on a personal computer.

The proposed method is also attractive for bifurcation analyses with detailed surface kinetics [4]–[8]. In that case, Eqs. (9) and (10) must be supplemented with a set of equations for the chemical rates associated with the heterogeneous scheme, in which the surface

coverages enter as additional variables. Since the solution procedure does not require integration of any differential equation, the set of nonlinear algebraic equations that emerge can be solved with a standard numerical routine, thereby reducing considerably the required computation time. The formulation is also advantageous for investigations of surface kinetics [3], readily providing accurate computational results to be used in interpreting experimental data.

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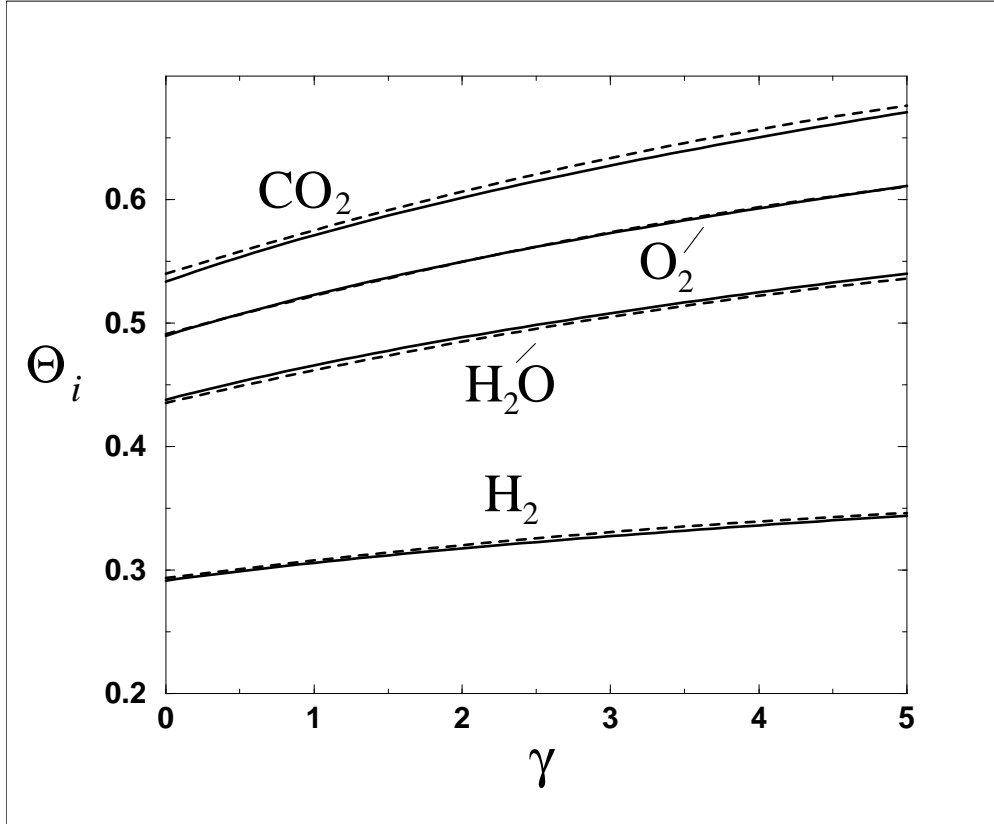


Figure 1: The variation with γ of the functions Θ_i for CO₂ ($L_{\text{CO}_2} = 1.39$), O₂ ($L_{\text{O}_2} = 1.11$), H₂O ($L_{\text{H}_2\text{O}} = 0.83$), and H₂ ($L_{\text{H}_2} = 0.30$) evaluated from numerical integrations (solid lines) and from Eq. (8) (dashed lines).