

An asymptotic theory for the propagation of a surface-catalysed flame in a tube

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Experiments have shown that when a mixture of fuel and oxygen is passed through a zirconia tube whose inner surface is coated with a catalyst, and then ignited at the end of the tube, a reaction front, or flame, propagates back along the tube towards the fuel inlet. The reaction front is visible as a red hot region moving at a speed of a few millimetres per second. In this paper we study a model of the flow, which takes into account diffusion, advection and chemical reaction at the inner surface of the tube. By assuming that the flame propagates at a constant speed without change of form, we can formulate a steady problem in a frame of reference moving with the reaction front. This is solved using the method of matched asymptotic expansions, assuming that the Reynolds and Damköhler numbers are large. We present numerical and, where possible, analytical results, first when the change in fluid density is small (a simplistic but informative limit) and secondly in the variable-density case. The speed of the travelling wave decreases as the critical temperature of the surface reaction increases and as the mass flow rate of fuel increases. We also make a comparison between our results and some preliminary experiments.

1. Introduction

In this paper we construct and analyse a mathematical model for a novel, surface-catalysed reaction front that has been observed in an experiment performed recently on a model fuel cell (Lefevre 2003). The experimental set-up is shown in figure 1. A mixture of fuel and oxygen is fed into one end of a long thin tube of zirconia (the inlet). The opposite end of the tube (the outlet) is heated by igniting the gas so as to form a premixed flame at the end of the tube. The increased temperature at the outlet starts a reaction on the inner surface of the zirconia, where a high-surface-area platinum catalyst is present, and the heat produced by the reaction causes the tube to glow red. When the premixed flame at the end of the tube is extinguished, the red glow remains, and then moves at a speed of a few millimetres per second along the tube towards the inlet. The motion of the reaction front is shown in figure 2;

† Professor King died in January 2005.

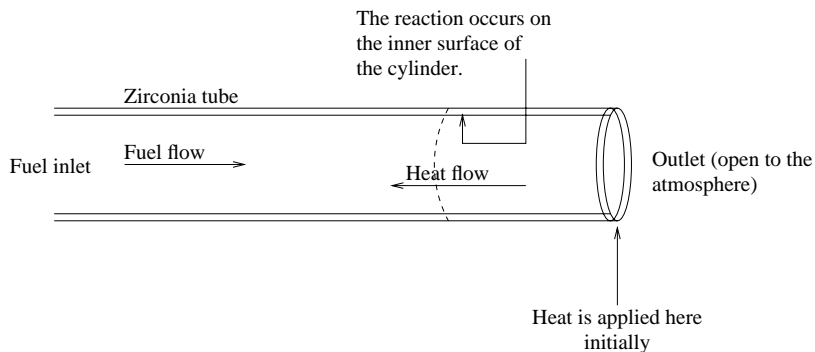


FIGURE 1. Schematic of the experiment.

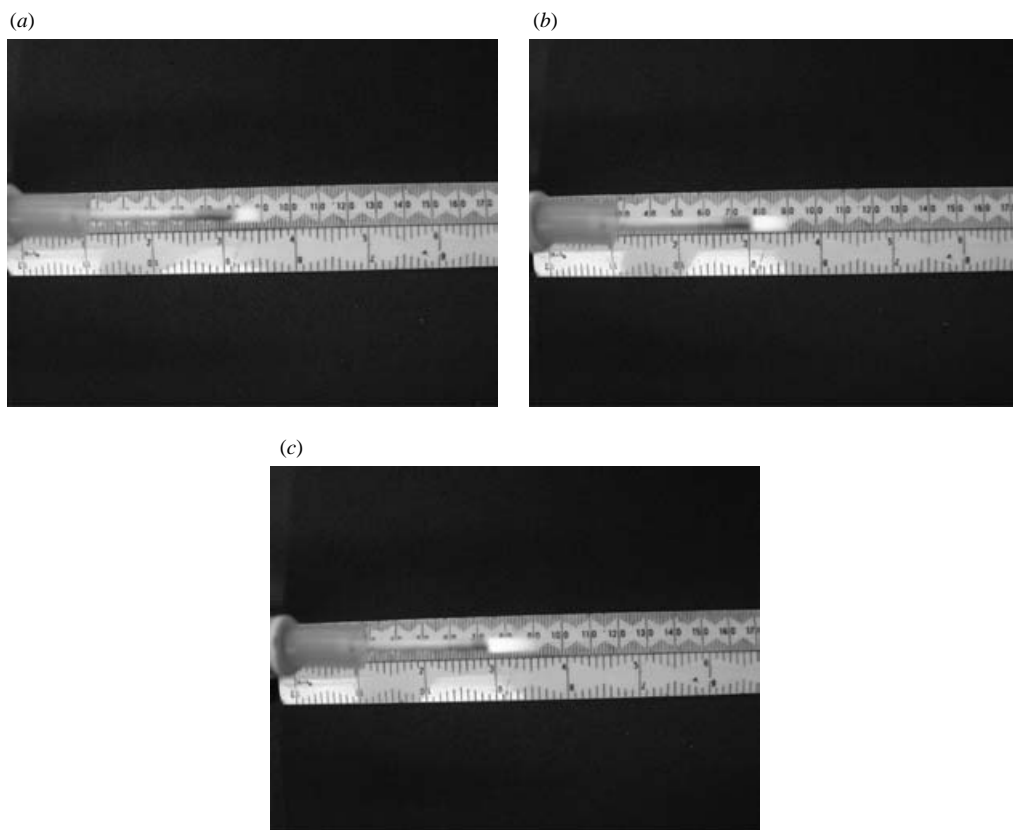


FIGURE 2. The observed propagation of the reaction front, or surface-catalysed flame. (a) At time $t = 0.0$ s the reaction front is at $d = 80$ mm. (b) At time $t = 6.5$ s the reaction front has progressed to $d = 77$ mm. (c) At time $t = 12.7$ s the reaction front has progressed to $d = 74$ mm.

fuel flows along the tube from the left and the outlet is at a distance shown by $d = 90$ mm on the ruler. The speed of the reaction front calculated from these pictures is about 0.46 mm s^{-1} . This speed decreases as the flow rate of fuel coming into the tube increases, and increases with the amount of catalyst on the inner surface of the tube. Note that this reaction front, or surface-catalysed flame, has some features of a

premixed flame and some of a diffusion flame, but is distinct from either: although the fuel and air are premixed, reaction only occurs at the wall of the tube, so the dynamics are controlled by the rate at which the mixture can be transported to the wall by diffusion.

There is an obvious analogy between this phenomenon and the classical experiment of Davy (1817). In his experiment, Davy heated a fine platinum wire and introduced it into a mixture of methane and air. He noted:

“...the oxygene (sic) and coal gas in contact with the hot wire combined without flame, and yet produced heat enough to preserve the wire ignited, and to keep up their own combustion.”

Apart from its intrinsic scientific interest, an understanding of surface-catalysed flames could improve our understanding of the way a fuel cell behaves as it is heated to its working temperature. A fuel cell is a device that converts chemical energy into electrical energy. It differs from a battery in that it is continuously supplied with fuel rather than simply using a finite amount of fuel stored within it. The fuel cell is one of the cleanest and most efficient forms of energy production available at the moment (Winkler 1994; Kumar, Krumpelt & Myles 1992). There are several types of fuel cell in development which have different applications. The solid oxide fuel cell (SOFC) consists of an electrolyte layer made from a solid ceramic, zirconia (ZrO_2), which at high temperatures (usually at least 700°C) can conduct oxygen ions (Singhal & Kendall 2003). Fuel reacts on one side of the zirconia, where a catalyst is present, reducing the oxygen concentration on this side. The other side, which is exposed to the air, remains at the atmospheric oxygen concentration. The fuel burning process therefore results in a gradient in oxygen concentration across the zirconia electrolyte. An electrical connection is placed across the zirconia, the cathode on the oxygen-rich side and the anode on the fuel side. At the cathode, electrons combine with oxygen molecules to form negatively charged oxygen ions, which travel through the zirconia down the concentration gradient. On reaching the anode, the oxygen ions release electrons and react with fuel. The electrons continue around the circuit back to the cathode.

An important design feature of the SOFC is that the oxygen-rich and fuel sides of the electrolyte must be kept separate. This ensures that the oxygen travels only through the electrolyte and produces an electrical current. Any leakage of oxygen between the two sides of the fuel cell reduces its efficiency. The tubular SOFC is made up of a tube of zirconia with tubular electrodes on either side. This arrangement helps to solve the problem of sealing one side from the other. The tubular design we study was developed by Kendall & Prica (1994) and Finnerty *et al.* (1998). Fuel flows through the zirconia tube and reacts with oxygen at the inner surface which is coated with a platinum catalyst. The tubes used in these cells are extruded from powders dispersed in polymer solutions which makes the cells more resistant to thermal and mechanical shocks. These cells can therefore be used for small-scale applications as they are not likely to break during rapid start-up.

Previous mathematical analysis of the tubular SOFC has considered the cell running under isothermal steady-state conditions (Cooper 2000; Cooper, Billingham & King 2000). The surface-catalysed reaction front that we study here is relevant to the initial heating and start-up of this type of fuel cell. We shall neglect electrochemical effects in our analysis, since the surface-catalysed reaction front is observed in the absence of electrochemical activity. In the following section we formulate a mathematical model that describes the flow field, temperature distribution and mass fraction distributions,

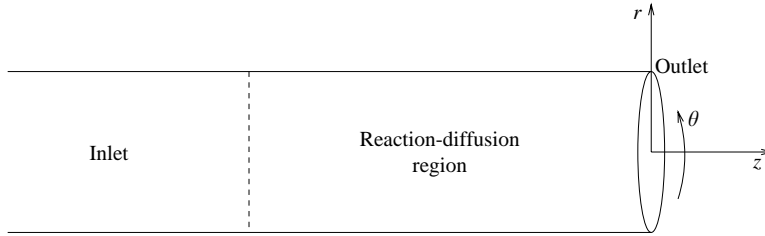


FIGURE 3. The coordinate system and asymptotic regions.

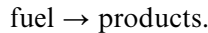
and the speed of the reaction front. We then describe the basic structure of the asymptotic solution, and some of the boundary value problems that we need to solve, in §3. We take two different approaches: in §4 we briefly discuss the case of small heat production, and hence constant density at leading order, whilst in §5 we present results for the variable-density model, which has a slightly more involved asymptotic structure. We then compare our theory with some preliminary experiments, and draw some conclusions.

2. The mathematical model

In the following analysis we use a cylindrical polar coordinate system, (r, θ, z) , with the origin at the centre of the outlet of the tube, as shown in figure 3. Although we formulate the governing equations for an unsteady flow, in due course we shall look for a steady, permanent-form travelling wave solution (a flame solution).

2.1. The governing equations and boundary conditions

The equations for conservation of mass, momentum and energy are derived using continuum theory for a mixture of reacting species (Buckmaster & Ludford 1982). From these equations we wish to obtain an asymptotic solution that will provide us with some insight into the structure of the flow, so we must make some simplifying assumptions. We first consider simplifying the chemistry of the problem. The reaction of methane with oxygen is complex (Senkan 1992) and we simplify the system so that the reaction is essentially



This model is widely used for premixed flames (Buckmaster & Ludford 1982). Since mass, m , is conserved, $m_f = m_p$, where the subscripts f and p denote properties of the fuel and product respectively. Also, since the sum of the mass fractions, Y_i , is always unity, we have $Y_p = 1 - Y_f$ and hence we only have to solve for the fuel mass fraction which satisfies

$$\rho \left(\frac{\partial Y_f}{\partial t} + u \frac{\partial Y_f}{\partial r} + w \frac{\partial Y_f}{\partial z} \right) = \frac{1}{r} \frac{\partial}{\partial r} \left(r \rho D \frac{\partial Y_f}{\partial r} \right) + \frac{\partial}{\partial z} \left(\rho D \frac{\partial Y_f}{\partial z} \right), \quad (2.1)$$

where ρ is the total density of the fluid mixture, D is the molecular diffusivity of the gases in the mixture (which we assume to be the same for both species) and u and w are the radial and axial velocities. This simplification also affects the equation of state, which becomes

$$p = \frac{R \rho T}{m_f},$$

where R is the molar gas constant and p and T are the total pressure and temperature of the fluid mixture. Since the temperature of the mixture is low enough that there is

no significant chemical reaction in the bulk, there is no creation of mass, momentum or energy there and the mixture of gases behaves as a continuous fluid. The chemical reaction is confined to the inner surface of the tube, where a platinum catalyst is present, and will therefore appear as a boundary condition.

At the boundary between the surface of the tube and the rest of the gas, $r = a$, there is an influx of fuel into the boundary, balanced by an efflux of products produced by the chemical reaction. Surface-catalysed reactions are complex, multi-step processes (see, for example, Wei & Iglesia 2004), each part of which is likely to have Arrhenius temperature-dependence. Since we have already made a huge simplification of the chemistry by just considering fuel and products, we choose the simplest possible temperature dependence for the reaction, namely that there is no reaction for $T < T_c$, and a constant reaction rate for $T \geq T_c$, where T_c is a constant critical temperature. Although the correspondence is not precise, this gives a reaction rate qualitatively similar to an Arrhenius reaction rate $e^{-T_a/T}$. We hope to explore the effect of this more realistic temperature dependence in future publications. Using the law of mass action, we now have

$$\rho D \frac{\partial Y_f}{\partial r} = \begin{cases} -k_0 \rho Y_f, & T \geq T_c \\ 0, & T < T_c \end{cases} \quad \text{at } r = a,$$

where k_0 is the constant rate of the catalysed reaction.

At the inner surface of the tube we also have continuity of velocity normal to the surface, $u = 0$ since gases cannot penetrate the tube wall, and the no-slip condition at the solid boundary, $w = 0$. To obtain the heat-flux boundary condition, we assume that the radial temperature gradient across the tube is small, since the zirconia is thin compared to the radius of the tube. We can therefore combine the boundary conditions at the inner and outer surfaces to obtain

$$k \frac{\partial T}{\partial r} = \begin{cases} k_0 \rho Y_f Q_c, & T \geq T_c \\ 0, & T < T_c \end{cases} \quad \text{at } r = a,$$

where k is the thermal conductivity of the mixture and Q_c is the heat of combustion of methane. The heat losses due to radiation and natural convection are small in comparison to the magnitude of the heat flux through the tube; the non-dimensional radiation heat transfer coefficient is $A = O(10^{-2})$, and the Nusselt number is $Nu = O(10^{-1})$ (see tables 1 and 2). We have therefore neglected them here, although their effect on the solution as a small perturbation over a long distance remains to be investigated. In particular, the appearance of the reaction front as a red band rather than an extended red hot region must be due to cooling by heat losses. We assume that this affects the appearance, but not the leading-order behaviour of the flame.

The length of the tube is much greater than its radius, so we treat it as semi-infinite to leading order. The temperature, mass fraction and mass flow rate at the inlet therefore satisfy

$$T \rightarrow T_{\text{in}}, \quad Y_f \rightarrow 1, \quad 2\pi \int_0^a \rho w r \, dr \rightarrow q_{\text{in}} \quad \text{as } z \rightarrow -\infty, \quad (2.2)$$

where q_{in} is the inlet mass flow rate and T_{in} the inlet temperature.

2.2. Non-dimensionalization

In what follows, dimensional variables are denoted by a star. We non-dimensionalize the governing equations using characteristic scales of the system. As there is now no geometrical length scale in the axial direction, we use the diffusion length scale. The

Physical quantity	Symbol	Value/magnitude	
Inner radius of tube	a	10^{-3}	m
Width of tube	$h = b - a$	2×10^{-4}	m
Mass flow rate at inlet	q_{in}	1×10^{-6}	kg s ⁻¹
Inlet (room) temperature	T_{in}	293	K
Critical temperature of reaction	T_c	$O(10^3)$	K
Density at the inlet ($T = T_{\text{in}}$)	ρ_{in}	$O(1)$	kg m ⁻³
Molecular diffusivity at inlet	D_{in}	$O(10^{-6})$	m ² s ⁻¹
Viscosity of gas mixture at inlet	μ_{in}	$O(10^{-5})$	kg m ⁻¹ s ⁻¹
Thermal conductivity of gas mixture at inlet	k_{in}	$O(10^{-2})$	W m ⁻¹ K ⁻¹
Specific heat at constant pressure of gas mixture	c_p	$O(10^3)$	J kg ⁻¹ K ⁻¹
Specific heat at constant volume of gas mixture	c_v	$O(10^3)$	J kg ⁻¹ K ⁻¹
Stefan–Boltzman constant	σ	5.67×10^{-8}	W m ⁻² K ⁻⁴
Emissivity of zirconia tube	ϵ	$O(10^{-1})$	
Surface heat transfer coefficient	H	$O(1)$	W m ⁻² K ^{-5/4}
Heat of combustion of methane	Q_c	5.570×10^7	J kg ⁻¹
Rate of reaction	k_0	$O(10^{-2})$	m s ⁻¹

TABLE 1. Parameters, their values and their meanings.

dimensionless variables are

$$r = \frac{r^*}{a}, \quad z = \frac{z^*}{qa}, \quad u = \frac{qu^*}{W}, \quad w = \frac{w^*}{W}, \quad t = \frac{Wt^*}{qa}, \quad T = \frac{T^*}{T_{\text{in}}},$$

$$k = \frac{k^*}{k_{\text{in}}}, \quad D = \frac{D^*}{D_{\text{in}}}, \quad \mu = \frac{\mu^*}{\mu_{\text{in}}}, \quad p = \frac{p^* - p_{\text{in}}}{\rho_{\text{in}}W^2}, \quad \rho = \frac{\rho^*}{\rho_{\text{in}}},$$

where qa is the diffusion length scale and

$$q = \frac{Wa}{D_{\text{in}}}.$$

The parameters D_{in} , μ_{in} , k_{in} and ρ_{in} are the diffusivity, viscosity, thermal conductivity and density of the mixture of fluids at the inlet temperature, T_{in} . The axial velocity scale, W , is given by

$$W = \frac{q_{\text{in}}}{\pi a^2 \rho_{\text{in}}}.$$

In terms of these variables, the dimensionless governing equations are

$$\frac{\partial \rho}{\partial t} + \frac{1}{r} \frac{\partial}{\partial r} (r \rho u) + \frac{\partial}{\partial z} (\rho w) = 0, \quad (2.3a)$$

$$\frac{\partial u}{\partial t} + u \frac{\partial u}{\partial r} + w \frac{\partial u}{\partial z} = -q^2 \frac{1}{\rho} \frac{\partial p}{\partial r} + \frac{q}{Re} \frac{1}{3\rho} \left[4 \frac{\partial}{\partial r} \left(\mu \frac{\partial u}{\partial r} \right) + 4 \frac{\mu}{r} \frac{\partial u}{\partial r} - \frac{4u\mu}{r^2} \right. \\ \left. + 3 \frac{\partial}{\partial z} \left(\mu \frac{\partial w}{\partial r} \right) - 2 \frac{\partial}{\partial r} \left(\mu \frac{\partial w}{\partial z} \right) - 2 \frac{u}{r} \frac{\partial \mu}{\partial r} \right] + \frac{1}{Re} \frac{1}{q} \frac{\partial}{\partial z} \left(\mu \frac{\partial u}{\partial z} \right), \quad (2.3b)$$

$$\frac{\partial w}{\partial t} + u \frac{\partial w}{\partial r} + w \frac{\partial w}{\partial z} = -\frac{1}{\rho} \frac{\partial p}{\partial z} + \frac{1}{Re} \frac{1}{3\rho} \left[\frac{1}{r} \mu \frac{\partial u}{\partial z} + 3 \frac{\partial}{\partial r} \left(\mu \frac{\partial u}{\partial z} \right) - 2 \frac{\partial}{\partial z} \left(\mu \frac{\partial u}{\partial r} \right) \right. \\ \left. + 4 \frac{\partial}{\partial z} \left(\mu \frac{\partial w}{\partial z} \right) - 2u \frac{1}{r} \frac{\partial \mu}{\partial z} \right] + \frac{q}{Re} \frac{1}{\rho} \frac{\partial}{\partial r} \left(r \mu \frac{\partial w}{\partial r} \right), \quad (2.3c)$$

$$\rho \left(\frac{\partial Y_f}{\partial t} + u \frac{\partial Y_f}{\partial r} + w \frac{\partial Y_f}{\partial z} \right) = \frac{1}{r} \frac{\partial}{\partial r} \left(r \rho D \frac{\partial Y_f}{\partial r} \right) + \frac{1}{q^2} \frac{\partial}{\partial z} \left(\rho D \frac{\partial Y_f}{\partial z} \right), \quad (2.3d)$$

Symbol	Definition	Typical value	Description
q	$\frac{q_{in}}{\pi a \rho_{in} D_{in}}$	$O(10^2)$	$\frac{\text{rate of fuel flow}}{\text{rate of diffusion}}$
Re	$\frac{q_{in}}{\pi a \mu_{in}}$	$O(10^2)$	$\frac{\text{inertial forces}}{\text{viscous forces}}$
Pr	$\frac{\mu_{in} c_p}{k_{in}}$	$O(1)$	$\frac{\text{momentum diffusivity}}{\text{thermal diffusivity}}$
\mathcal{L}	$\frac{k_{in}}{\rho_{in} D_{in} c_p}$	$O(1)$	$\frac{\text{thermal diffusivity}}{\text{mass diffusivity}}$
γ	$\frac{c_p}{c_v}$	$O(1)$	$\frac{\text{specific heat at constant pressure}}{\text{specific heat at constant volume}}$
M	$\frac{q_{in}}{\pi a^2 \rho_{in} \sqrt{(\gamma - 1) c_p T_{in}}}$	$O(10^{-3})$	$\frac{\text{average speed in fluid}}{\text{average speed of sound in fluid}}$
Q	$\frac{Q_c}{c_p T_{in}}$	$O(10)$	$\frac{\text{heat produced in reaction}}{\text{heat transferred by conduction}}$
Da	$\frac{k_0 a}{D_{in}}$	$O(10)$	$\frac{\text{rate of reaction}}{\text{rate of diffusion}}$
ε	$\frac{h}{a}$	$O(10^{-1})$	$\frac{\text{thickness of tube}}{\text{radius of tube}}$
A	$\frac{\sigma \epsilon T_{in}^3 a}{k_{in}}$	$O(10^{-2})$	$\frac{\text{rate of heat transfer by radiation}}{\text{rate of heat transfer by conduction}}$
Nu	$\frac{Ha T_{in}^{1/4}}{k_{in}}$	$O(10^{-1})$	$\frac{\text{rate of heat transfer by convection}}{\text{rate of heat transfer by conduction}}$

TABLE 2. The dimensionless parameters.

$$\rho \left(\frac{\partial T}{\partial t} + u \frac{\partial T}{\partial r} + w \frac{\partial T}{\partial z} \right) = \mathcal{L} \frac{1}{r} \frac{\partial}{\partial r} \left(rk \frac{\partial T}{\partial r} \right) + \frac{\mathcal{L}}{q^2} \frac{\partial}{\partial z} \left(k \frac{\partial T}{\partial z} \right), \tag{2.3e}$$

$$\rho T = 1, \tag{2.3f}$$

where Re and \mathcal{L} are the Reynolds number and Lewis number respectively, defined by

$$Re = \frac{Wa \rho_{in}}{\mu_{in}}, \quad \mathcal{L} = \frac{k_{in}}{\rho_{in} D_{in} c_p}.$$

Since the Mach number, M , satisfies $M^2 \ll 1$ (see table 2), we have set $M = 0$ in all our calculations. Defining the Prandtl number by $Pr = \mu_{in} c_p / k_{in}$ we can write q in terms of the Reynolds, Prandtl and Lewis numbers as

$$q = Re Pr \mathcal{L}. \tag{2.4}$$

Since both Pr and \mathcal{L} are $O(1)$ parameters, $q = O(Re)$, with $Re \gg 1$. We can therefore think of q as a measure of the mass flow rate.

2.2.1. Boundary conditions

In terms of the dimensionless variables, the boundary conditions at $r = 1$ are

$$u = 0, \quad w = 0,$$

$$\rho D \frac{\partial Y_f}{\partial r} = \begin{cases} -Da\rho Y_f, & T > T_c, \\ 0, & T < T_c, \end{cases} \quad k \frac{\partial T}{\partial r} = \begin{cases} QDa\rho Y_f/\mathcal{L}, & T > T_c, \\ 0, & T < T_c, \end{cases}$$

where T_c is now the dimensionless critical temperature of the reaction, Da is the Damköhler number and Q is the dimensionless heat of reaction

$$Da = \frac{k_0 a}{D_{in}}, \quad Q = \frac{Q_c}{c_p T_{in}}.$$

At $r=0$, u , w , Y_f and T must be finite, and

$$u \rightarrow 0, \quad \int_0^1 r \rho w \, dr \rightarrow \frac{1}{2}, \quad Y_f \rightarrow 1, \quad T \rightarrow 1,$$

as $z \rightarrow -\infty$.

2.3. Further simplifications

A commonly used and reasonable approximation is that the fuel and temperature diffuse at the same rate, and hence that the Lewis number is unity, $\mathcal{L} = 1$ (Buckmaster & Ludford 1982). From this we further assume that the temperature dependences of the thermal conductivity and diffusion coefficient are such that $k = \rho D$. This allows us to add Q times (2.3d) to (2.3e) giving an equation for $QY_f + T$. From the boundary conditions we have

$$\frac{\partial}{\partial r}(QY_f + T) = 0 \quad \text{at } r = 1, \quad QY_f + T \rightarrow Q + 1 \quad \text{as } z \rightarrow -\infty,$$

so $QY_f + T = Q + 1$ is a solution. We therefore only have to explicitly solve for one of either temperature or mass fraction. We choose to solve for temperature.

3. The structure of the asymptotic solution

We now seek an asymptotic structure based on the limit of large q and Da . Physically, this corresponds to a rapid throughput of fuel and fast chemical reaction at the inner surface of the tube. We look for a travelling wave solution, which we expect will develop as the large-time solution. We transform the equations into a frame of reference moving with the flame front at a constant speed, c , which we will have to determine. Experimentally this wave speed is much less than the flow speed and its magnitude will be determined theoretically by the scalings in the asymptotic regions, as we will show in §3.1. Writing $z' = z + ct$ and substituting into (2.3), we obtain

$$\frac{1}{r} \frac{\partial}{\partial r}(r\rho u) + \frac{\partial}{\partial z'}(\rho w') = 0, \quad (3.1a)$$

$$u \frac{\partial u}{\partial r} + w' \frac{\partial u}{\partial z'} = -q^2 \frac{1}{\rho} \frac{\partial p}{\partial r} + \frac{q}{Re} \frac{1}{3\rho} \left[4 \frac{\partial}{\partial r} \left(\mu \frac{\partial u}{\partial r} \right) + 4 \frac{\mu}{r} \frac{\partial u}{\partial r} - \frac{4u\mu}{r^2} \right. \\ \left. + 3 \frac{\partial}{\partial z'} \left(\mu \frac{\partial w'}{\partial r} \right) - 2 \frac{\partial}{\partial r} \left(\mu \frac{\partial w'}{\partial z'} \right) - 2 \frac{u}{r} \frac{\partial \mu}{\partial r} \right] + \frac{1}{Re} \frac{1}{q} \frac{\partial}{\partial z'} \left(\mu \frac{\partial u}{\partial z'} \right), \quad (3.1b)$$

$$u \frac{\partial w'}{\partial r} + w' \frac{\partial w'}{\partial z'} = -\frac{1}{\rho} \frac{\partial p}{\partial z'} + \frac{1}{Re} \frac{1}{q} \frac{1}{3\rho} \left[\frac{1}{r} \mu \frac{\partial u}{\partial z'} + 3 \frac{\partial}{\partial r} \left(\mu \frac{\partial u}{\partial z'} \right) - 2 \frac{\partial}{\partial z'} \left(\mu \frac{\partial u}{\partial r} \right) \right. \\ \left. + 4 \frac{\partial}{\partial z'} \left(\mu \frac{\partial w'}{\partial z'} \right) - 2u \frac{1}{r} \frac{\partial \mu}{\partial z'} \right] + \frac{q}{Re} \frac{1}{\rho} \frac{1}{r} \frac{\partial}{\partial r} \left(r\mu \frac{\partial w'}{\partial r} \right), \quad (3.1c)$$

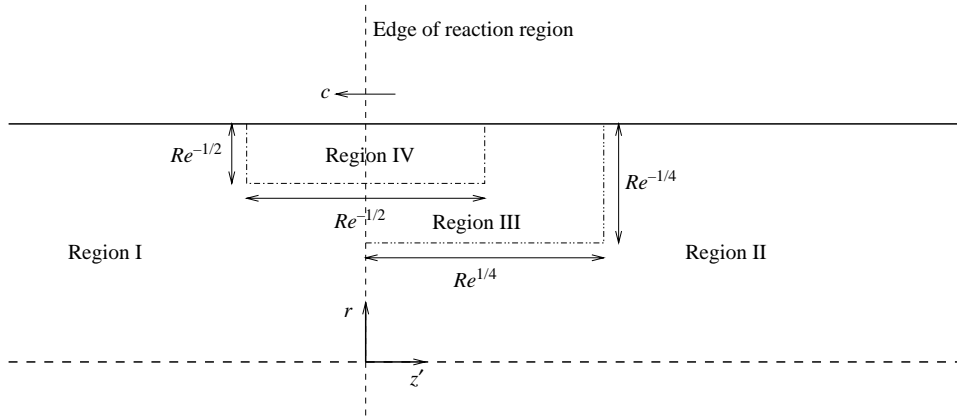


FIGURE 4. Sketch of the tube showing the asymptotic regions.

$$\rho \left(u \frac{\partial T}{\partial r} + w' \frac{\partial T}{\partial z'} \right) = \frac{1}{r} \frac{\partial}{\partial r} \left(r k \frac{\partial T}{\partial r} \right) + \frac{1}{q^2} \frac{\partial}{\partial z'} \left(k \frac{\partial T}{\partial z'} \right), \tag{3.1d}$$

$$\rho T = 1, \tag{3.1e}$$

where $w' = w + c$ is the axial velocity in the new frame of reference. The boundary conditions in this frame of reference become

$$\begin{aligned} u = 0, \quad w' = c, \\ k \frac{\partial T}{\partial r} = \begin{cases} Da\rho(Q + 1 - T), & z' > 0 \\ 0, & z' < 0 \end{cases} \quad \text{at } r = 1. \end{aligned} \tag{3.2}$$

At $r = 0$ we require that u, w' and T are finite, and as $z' \rightarrow -\infty$ we match the flow in the reaction region to the steady inlet flow.

Since we are looking for a travelling wave solution, which is translationally invariant, we fix the origin, the position where the chemical reaction begins, by assuming that $T = T_c$ at $z' = 0, r = 1$. In subsequent calculations, this means that we will prescribe the wave speed and look for a solution. The corresponding critical temperature, T_c , is then the temperature at $z' = 0, r = 1$.

The Damköhler number is large, so changes in the flow due to the reaction occur in a small region close to the reaction front at $z' = 0, r = 1$. This region is the innermost of our asymptotic structure and is shown as region IV in figure 4. The outer regions, I and II, give the $O(1)$ flow in the tube for $z' < 0$ and $z' > 0$ respectively. In $z' < 0$, no chemical reaction occurs and the flow at leading order is just the steady inlet flow. In $z' > 0$, changes in the flow due to chemical reaction at $r = 1$ diffuse out into the centre of the tube. The solution in region III matches together the solutions in regions II and IV.

In regions I and II, the leading-order equations are the large q and Da limits of (3.1) and (3.2). From (2.4) we have $q = Pr Re$, since $\mathcal{L} = 1$. Substituting this into (3.1) and taking the limit of large q we have

$$\frac{1}{r} \frac{\partial}{\partial r} (r\rho u) + \frac{\partial}{\partial z'} (\rho w') = 0, \tag{3.3a}$$

$$\frac{\partial p}{\partial r} = 0, \tag{3.3b}$$

$$u \frac{\partial w'}{\partial r} + w' \frac{\partial w'}{\partial z'} = -\frac{1}{\rho} \frac{\partial p}{\partial z'} + Pr \frac{1}{\rho} \frac{1}{r} \frac{\partial}{\partial r} \left(r \mu \frac{\partial w'}{\partial r} \right), \tag{3.3c}$$

$$\rho \left(u \frac{\partial T}{\partial r} + w' \frac{\partial T}{\partial z'} \right) = \frac{1}{r} \frac{\partial}{\partial r} \left(r k \frac{\partial T}{\partial r} \right), \tag{3.3d}$$

$$\rho T = 1, \tag{3.3e}$$

at leading order. Taking $Da \rightarrow \infty$ in the heat-flux boundary condition at $r = 1$, (3.2), we have simply $\partial T / \partial r = 0$ for $z' < 0$ and $T = 1 + Q$ for $z' > 0$. The equations to be solved in the inner two regions can be found by rescaling (3.1).

We construct a region around the edge of the reaction front in which changes in the heat flux due to the reaction are of $O(1)$. The radial scale is therefore determined by considering the heat-flux boundary condition at the wall, (3.2). Writing $r^2 = 1 - \eta \bar{R}$ and substituting into (3.2), we have a balance of leading-order terms when $\eta = Da^{-1}$. Before we can determine the axial scaling of the inner region we need to know the magnitude of the wave speed. We can determine this by considering the solution in the inlet region and how it matches to the inner region.

3.1. Solution in the inlet region, Region I

No chemical reaction takes place in region I, and the inlet conditions show that $T = 1$, $\rho = 1$, $\mu = 1$ and $k = 1$. The inlet flow is Poiseuille, with $u = 0$ and $w' = 2(1 - r^2) + c$. From experiments we know that the wave speed is much less than the flow speed, so we write $c = \xi C$ where $\xi \ll 1$ and $C = O(1)$ which gives

$$w' = 2(1 - r^2) + \xi C.$$

To obtain the richest balance of terms in the governing equations, we rescale the axial velocity so that it appears at leading order in the innermost asymptotic region. Let \bar{W} be the axial velocity in the inner region so that $w' = \xi(\bar{W} + C)$. In terms of the region IV scaled variable \bar{R} , we obtain

$$\bar{W} = 2\xi^{-1} Da^{-1} \bar{R},$$

so the richest balance occurs when $\xi = Da^{-1}$. This allows us to determine the equations in region IV.

3.2. Equations in region IV

We write $r^2 = 1 - Da^{-1} \bar{R}$, $z' = Da^{-3} \bar{Z}$, $w' = Da^{-1}(\bar{W} + C)$ and substitute into (3.1). At leading order we obtain

$$\frac{\partial}{\partial \bar{R}}(\rho \bar{U}) + \frac{\partial}{\partial \bar{Z}}(\rho(\bar{W} + C)) = 0, \tag{3.4a}$$

$$\begin{aligned} \frac{1}{2} \bar{U} \frac{\partial \bar{U}}{\partial \bar{R}} + \frac{1}{2}(\bar{W} + C) \frac{\partial \bar{U}}{\partial \bar{Z}} = & -\bar{q}^2 \frac{2}{\rho} \frac{\partial \bar{P}}{\partial \bar{R}} + \frac{Pr}{3\rho} \left[8 \frac{\partial}{\partial \bar{R}} \left(\mu \frac{\partial \bar{U}}{\partial \bar{R}} \right) + 6 \frac{\partial}{\partial \bar{Z}} \left(\mu \frac{\partial \bar{W}}{\partial \bar{R}} \right) \right. \\ & \left. - 4 \frac{\partial}{\partial \bar{R}} \left(\mu \frac{\partial \bar{W}}{\partial \bar{Z}} \right) \right] + \frac{Pr}{\bar{q}^2} \frac{1}{2\rho} \frac{\partial}{\partial \bar{Z}} \left(\mu \frac{\partial \bar{U}}{\partial \bar{Z}} \right), \end{aligned} \tag{3.4b}$$

$$\begin{aligned} \bar{U} \frac{\partial \bar{W}}{\partial \bar{R}} + (\bar{W} + C) \frac{\partial \bar{W}}{\partial \bar{Z}} = & -\frac{1}{\rho} \frac{\partial \bar{P}}{\partial \bar{Z}} + \frac{1}{\bar{q}^2} \frac{Pr}{3\rho} \left[3 \frac{\partial}{\partial \bar{R}} \left(\mu \frac{\partial \bar{U}}{\partial \bar{Z}} \right) - 2 \frac{\partial}{\partial \bar{Z}} \left(\mu \frac{\partial \bar{U}}{\partial \bar{R}} \right) \right. \\ & \left. + 4 \frac{\partial}{\partial \bar{Z}} \left(\mu \frac{\partial \bar{W}}{\partial \bar{Z}} \right) \right] + \frac{4Pr}{\rho} \frac{\partial}{\partial \bar{R}} \left(\mu \frac{\partial \bar{W}}{\partial \bar{R}} \right), \end{aligned} \tag{3.4c}$$

$$\rho \bar{U} \frac{\partial T}{\partial \bar{R}} + \rho \left(\bar{W} + C \right) \frac{\partial T}{\partial \bar{Z}} = 4 \frac{\partial}{\partial \bar{R}} \left(k \frac{\partial T}{\partial \bar{R}} \right) + \frac{1}{\bar{q}^2} \frac{\partial}{\partial \bar{Z}} \left(k \frac{\partial T}{\partial \bar{Z}} \right), \quad (3.4d)$$

$$\rho T = 1, \quad (3.4e)$$

where $\bar{U} = -2Da^{-1}u$. The rescaled pressure, \bar{P} , is given by $p = Da^{-2}\bar{P}$, where p is the pressure in the outer regions. The parameter \bar{q} is given by $\bar{q} = qDa^{-2}$, and we assume that $\bar{q} = O(1)$.

The boundary conditions in region IV are

$$\bar{U} = 0, \quad \bar{W} = 0, \quad k \frac{\partial T}{\partial \bar{R}} = \begin{cases} \frac{1}{2}\rho(T - 1 - Q), & \bar{Z} > 0 \\ 0, & \bar{Z} < 0 \end{cases} \quad \text{at } \bar{R} = 0, \quad (3.5a)$$

$$\bar{U} \rightarrow 0, \quad \bar{W} \sim 2\bar{R}, \quad T \rightarrow 1 \quad \text{as } \bar{Z} \rightarrow -\infty, \quad (3.5b)$$

along with matching to the outer solution as $\bar{R}, \bar{Z} \rightarrow \infty$. We can now determine the scalings for the intermediate region, III.

3.3. Equations in region III

Region III is situated between regions II and IV as shown in figure 4. We rescale using $r^2 = 1 - Da^{-1/2}R$, $z' = Da^{-3/2}Z$ and $w' = Da^{-1/2}W + Da^{-1}C$. Substituting into (3.1), we find the leading-order equations to be

$$\frac{\partial}{\partial R} (\rho U) + \frac{\partial}{\partial Z} (\rho W) = 0, \quad (3.6a)$$

$$\frac{\partial P}{\partial R} = 0, \quad (3.6b)$$

$$U \frac{\partial W}{\partial R} + W \frac{\partial W}{\partial Z} = -\frac{1}{\rho} \frac{\partial P}{\partial Z} + \frac{4Pr}{\rho} \frac{\partial}{\partial R} \left(\mu \frac{\partial W}{\partial R} \right), \quad (3.6c)$$

$$\rho U \frac{\partial T}{\partial R} + \rho W \frac{\partial T}{\partial Z} = 4 \frac{\partial}{\partial R} \left(k \frac{\partial T}{\partial R} \right), \quad (3.6d)$$

$$\rho T = 1. \quad (3.6e)$$

The rescaled pressure, P , is given by $p = Da^{-1}P$ and the radial velocity is $U = -2Da^{-1/2}u$. The boundary conditions at the wall are

$$U = 0, \quad W = 0, \quad T = 1 + Q \quad \text{at } R = 0, \quad (3.7)$$

along with matching to the outer regions as $Z \rightarrow 0$ and $R \rightarrow \infty$.

We are now in a position to solve the systems of equations we have obtained in each of the asymptotic regions. We have already described the simple solution in region I, the inlet region, in §3.1. We will consider the solution in the three other regions for two different cases. The more complicated of the two is the $O(1)$ heat release problem, which is given by the variable-density equations. We first briefly consider the case of small heat release, which is significantly more straightforward to tackle than the full problem.

4. Solution for small heat release

The complexity of the systems to be solved in regions II, III and IV can be reduced by assuming that the heat of reaction, Q , is small. This approximation is not very realistic physically, but it simplifies the equations greatly and can be used as a check on the accuracy of our solutions when $Q = O(1)$ in the limit $Q \rightarrow 0$. Moreover, the

solution for $Q \ll 1$ retains most of the features and physics of the full problem. When $Q \ll 1$, the temperature remains close to its inlet value, so it is appropriate to define $T = 1 + Q\hat{T}$. It follows from the equation of state that, to leading order, the density is also constant (the constant-density approximation) and, since the viscosity and thermal conductivity are dependent only on the temperature, we have $\mu = 1$ and $k = 1$ at leading order. Using the constant-density approximation, we therefore find that the flow down the tube is unchanged, with $u = 0$ and $w' = 2(1 - r^2) + Da^{-1}C$ at leading order. The energy equation is now the only non-trivial equation to be solved. In each of the asymptotic regions, the leading-order equation can be found by substituting $u = 0$, $w' = 2(1 - r^2) + Da^{-1}C$, $T = 1 + Q\hat{T}$, $k = 1$ and $\rho = 1$ into (3.3d), (3.4d) and (3.6d) and their respective boundary conditions.

We do not go into details of the solution here, the full analysis is given in Adamson (2004); however, we present the main results to facilitate a comparison with the variable-density model.

4.1. Variation of wave speed with critical temperature

The equations in region IV were solved numerically using a finite difference scheme. The program is set up in such a way that C and \bar{q} are input and the temperature is then found; in particular, the corresponding value of T_c is the temperature at $z' = 0$, $r = 1$. Temperature profiles for different values of the wave speed, C , and hence of the critical temperature, T_c , for $\bar{q} = 1$, are shown in figure 5. Comparing these solutions we find that for small C the temperature increases more rapidly than for large C so that, at a given point, the temperature is greater when the wave speed is smaller. We obtained results at several wave speeds and for each value of C noted the temperature at $\bar{R} = 0$, $\bar{Z} = 0$, which is the critical temperature, \hat{T}_c , to which the solution corresponds. A plot, showing that the wave speed decreases with critical temperature, is shown in figure 6.

4.2. Variation of wave speed with mass flow rate

Taking a fixed value of critical temperature we varied \bar{q} to determine the effect on the wave speed. The results for $\hat{T}_c = 0.2$ are shown in figure 7. Increasing the mass flow rate causes the wave speed to decrease, and reach zero at $\bar{q} \approx 2.48$. For $\bar{q} > 2.48$ the wave speed is negative so that, instead of propagating towards the inlet of the tube, the wave front moves towards the outlet and will eventually be blown out.

The solution for small Q shows that the flame speed depends on both the mass flow rate of fuel into the tube and the critical temperature at which the reaction starts, \hat{T}_c . The latter is related to the amount of catalyst present on the inner surface of the tube; the more catalyst present the lower \hat{T}_c will be. These effects could both be tested experimentally. We make a comparison with experiment in §6, but first we tackle the problem when $Q = O(1)$.

5. Solution for $O(1)$ heat release

When $Q = O(1)$, the solution in the inlet region, which we discussed in §3.1, remains unchanged. In the other three regions, however, we must solve the more complicated system of variable-density equations.

5.1. Region II

In this region we solve (3.3) subject to the boundary conditions

$$u = 0, \quad w' = 0, \quad T = 1 + Q \quad \text{at } r = 1, \quad (5.1a)$$

$$u = 0, \quad w' = 2(1 - r^2), \quad T = 1 \quad \text{at } z' = 0. \quad (5.1b)$$

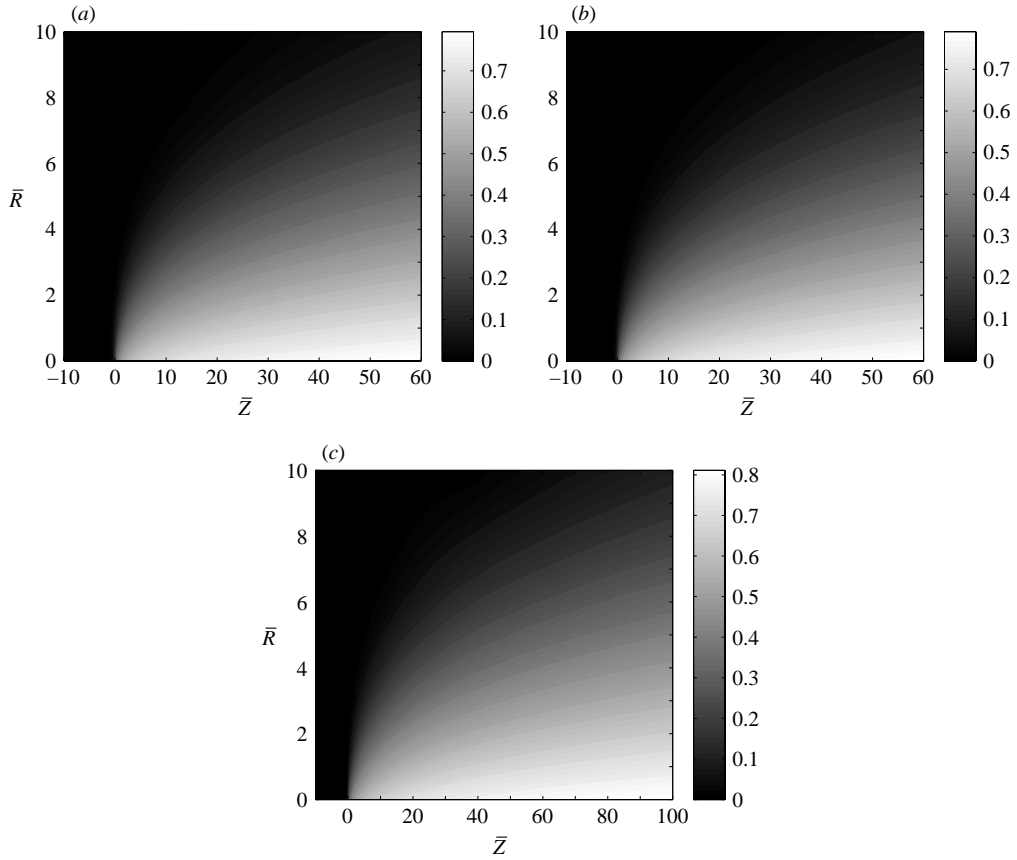


FIGURE 5. Solutions in region IV. (a) $C = 0.5$, (b) $C = 1.0$, (c) $C = 2.0$.

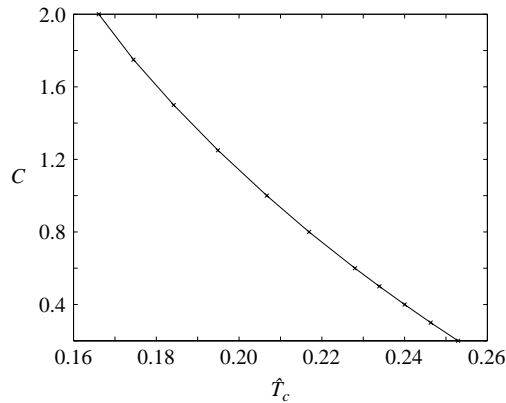


FIGURE 6. Plot of wave speed, C , against critical temperature, \hat{T}_c .

We also require that u, w' and T are finite at $r = 0$. We assume that the fluid behaves like the model fluid described by Stewartson (1964), so that the Prandtl number, $Pr = 1$, and the viscosity and thermal conductivity are directly proportional

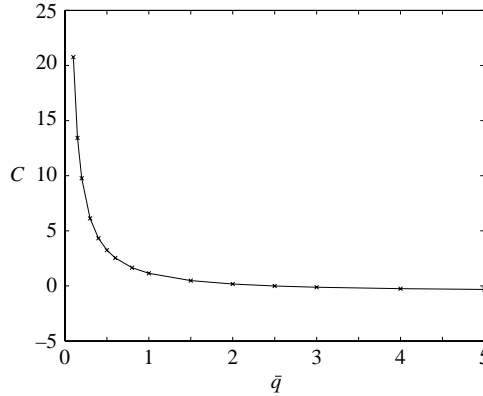


FIGURE 7. Plot of wave speed, C , against mass flow rate of fuel, \bar{q} .

to temperature, $\mu = T, k = T$. In terms of the streamfunction, ψ , defined by

$$\frac{\partial \psi}{\partial z'} = r\rho u, \quad \frac{\partial \psi}{\partial r} = -r\rho w',$$

so that conservation of mass, (3.3a), is satisfied, (3.3c, d) become

$$\begin{aligned} & -3\frac{T}{r^4}\frac{\partial \psi}{\partial z'}\frac{\partial \psi}{\partial r} - \frac{T}{r^3}\frac{\partial \psi}{\partial r}\frac{\partial^2 \psi}{\partial r \partial z'} + \frac{3}{r^3}\frac{\partial T}{\partial r}\frac{\partial \psi}{\partial z'}\frac{\partial \psi}{\partial r} + 3\frac{T}{r^3}\frac{\partial \psi}{\partial z'}\frac{\partial^2 \psi}{\partial r^2} - \frac{1}{r^2}\frac{\partial^2 T}{\partial r^2}\frac{\partial \psi}{\partial z'}\frac{\partial \psi}{\partial r} \\ & - \frac{2}{r^2}\frac{\partial T}{\partial r}\frac{\partial \psi}{\partial z'}\frac{\partial^2 \psi}{\partial r^2} - \frac{T}{r^2}\frac{\partial \psi}{\partial z'}\frac{\partial^3 \psi}{\partial r^3} - \frac{2}{r^3}\frac{\partial T}{\partial z'}\left(\frac{\partial \psi}{\partial r}\right)^2 + \frac{2}{r^2}\frac{\partial T}{\partial z'}\frac{\partial^2 \psi}{\partial r^2}\frac{\partial \psi}{\partial r} \\ & + \frac{1}{r^2}\frac{\partial^2 T}{\partial r \partial z'}\left(\frac{\partial \psi}{\partial r}\right)^2 + \frac{T}{r^2}\frac{\partial \psi}{\partial r}\frac{\partial^3 \psi}{\partial z' \partial r^2} = 3\frac{T^2}{r^4}\frac{\partial \psi}{\partial r} - 6\frac{T}{r^3}\frac{\partial T}{\partial r}\frac{\partial \psi}{\partial r} - 3\frac{T^2}{r^3}\frac{\partial^2 \psi}{\partial r^2} \\ & + \frac{3}{r^2}\left(\frac{\partial T}{\partial r}\right)^2\frac{\partial \psi}{\partial r} + 7\frac{T}{r^2}\frac{\partial T}{\partial r}\frac{\partial^2 \psi}{\partial r^2} + 2\frac{T^2}{r^2}\frac{\partial^3 \psi}{\partial r^3} + 3\frac{T}{r^2}\frac{\partial^2 T}{\partial r^2}\frac{\partial \psi}{\partial r} - \frac{3}{r}\frac{\partial T}{\partial r}\frac{\partial^2 T}{\partial r^2}\frac{\partial \psi}{\partial r} \\ & - \frac{4}{r}\left(\frac{\partial T}{\partial r}\right)^2\frac{\partial^2 \psi}{\partial r^2} - \frac{T}{r}\frac{\partial^3 T}{\partial r^3}\frac{\partial \psi}{\partial r} - 4\frac{T}{r}\frac{\partial^2 T}{\partial r^2}\frac{\partial^2 \psi}{\partial r^2} - 5\frac{T}{r}\frac{\partial T}{\partial r}\frac{\partial^3 \psi}{\partial r^3} - \frac{T^2}{r}\frac{\partial^4 \psi}{\partial r^4}, \end{aligned} \tag{5.2a}$$

$$\frac{\partial \psi}{\partial z'}\frac{\partial T}{\partial r} - \frac{\partial \psi}{\partial r}\frac{\partial T}{\partial z'} = T\frac{\partial T}{\partial r} + r\left(\frac{\partial T}{\partial r}\right)^2 + rT\frac{\partial^2 T}{\partial r^2}, \tag{5.2b}$$

where we have used the fact that $\partial p/\partial r = 0$ to remove the pressure terms. The boundary conditions for ψ and T are

$$\psi = -\frac{1}{2}, \quad \frac{\partial \psi}{\partial r} = 0, \quad T = 1 + Q \quad \text{at } r = 1, \tag{5.3a}$$

$$\psi = -\left(r^2 - \frac{r^4}{2}\right), \quad \frac{\partial \psi}{\partial z'} = 0, \quad T = 1 \quad \text{at } z' = 0. \tag{5.3b}$$

As u and w' must be finite at $r = 0$ we require that

$$\psi = 0, \quad \frac{\partial \psi}{\partial r} = 0,$$

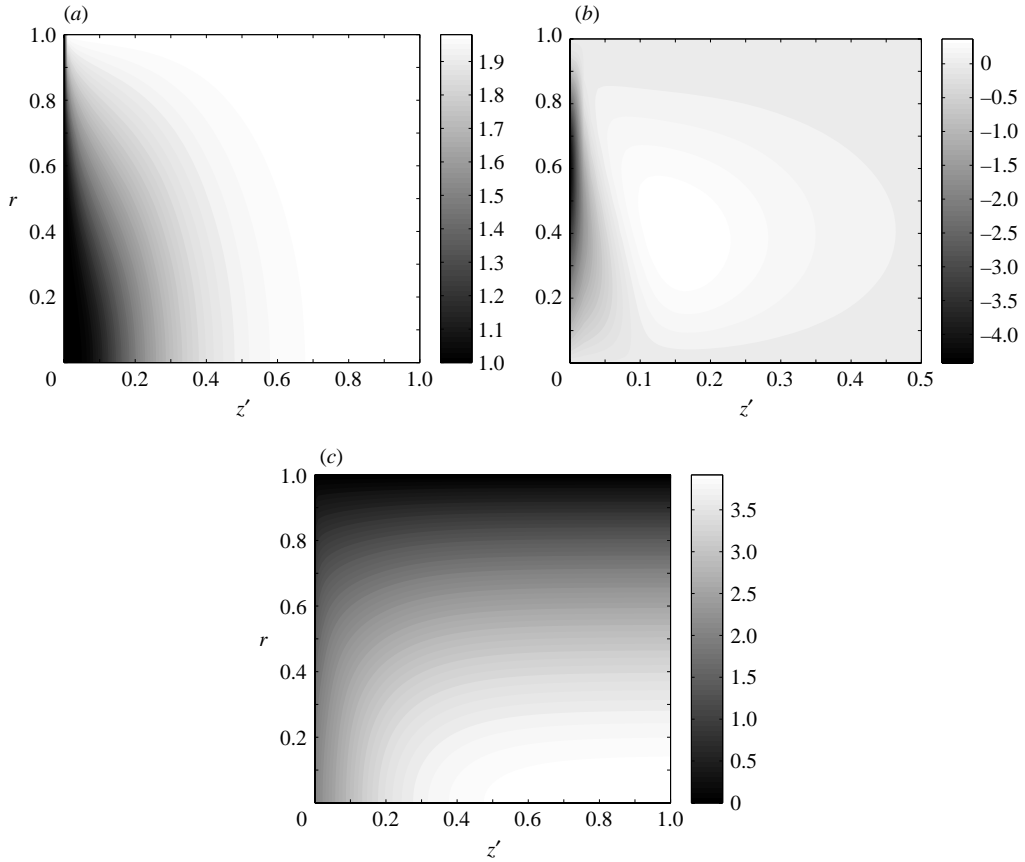


FIGURE 8. Solutions in region II. (a) Temperature, (b) radial velocity, (c) axial velocity.

and also impose the symmetry condition

$$\frac{\partial T}{\partial r} = 0 \quad \text{at } r = 0.$$

Using a forward-stepping implicit finite difference scheme, we discretize the equations on an $m \times n$ grid and write the problem in the form

$$\mathbf{A}_j \mathbf{T}_j = \mathbf{B}_j \mathbf{T}_{j-1}, \tag{5.4a}$$

$$\mathbf{C}_j \boldsymbol{\psi}_j = \mathbf{D}_j \boldsymbol{\psi}_{j-1}, \tag{5.4b}$$

where \mathbf{A}_j , \mathbf{B}_j , \mathbf{C}_j and \mathbf{D}_j are known $m \times m$ matrices, \mathbf{T}_j and $\boldsymbol{\psi}_j$ are vectors of length m that contain the discretized temperature and streamfunction, and $0 \leq j \leq n$. Writing $\mathbf{T}_j = \mathbf{T}_g + \Delta \mathbf{T}$ and $\boldsymbol{\psi}_j = \boldsymbol{\psi}_g$, where \mathbf{T}_g and $\boldsymbol{\psi}_g$ are approximations to the solutions \mathbf{T}_j and $\boldsymbol{\psi}_j$, and $\Delta \mathbf{T}$ is a small correction to the temperature, we obtain an equation for $\Delta \mathbf{T}$ of the form

$$\mathbf{M}_j \Delta \mathbf{T} = \mathbf{r}_j, \tag{5.5}$$

where \mathbf{M}_j is another known $m \times m$ matrix, and \mathbf{r}_j is a known vector of length m . Both \mathbf{M}_j and \mathbf{r}_j contain the approximate solutions \mathbf{T}_g and $\boldsymbol{\psi}_g$. In a similar manner we obtain an equation for $\Delta \boldsymbol{\psi}$. We solve this system for $\Delta \mathbf{T}$ and $\Delta \boldsymbol{\psi}$ in turn and use an iterative procedure to improve the approximations for \mathbf{T}_g and $\boldsymbol{\psi}_g$ until both $\Delta \mathbf{T}$ and $\Delta \boldsymbol{\psi}$ have converged to zero. Some typical solutions are shown in figure 8.

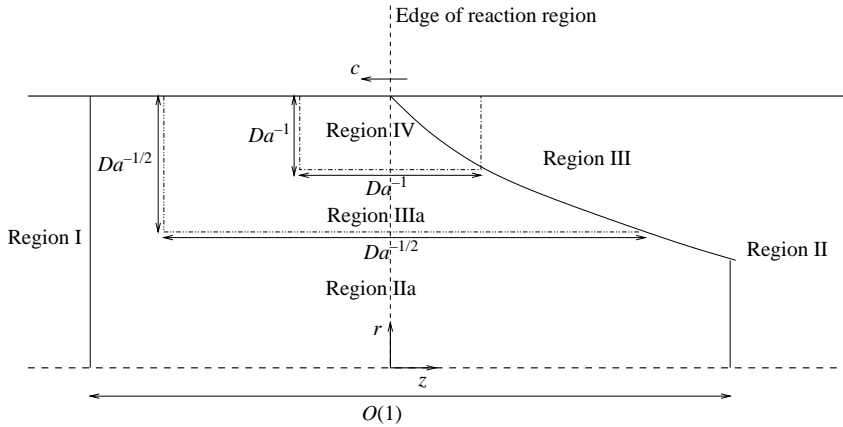


FIGURE 9. Sketch of the tube showing the additional asymptotic regions.

The qualitative form of the solutions is as we might expect, with the temperature decreasing with distance from the wall and increasing as the distance from the edge of the reaction front, z' , increases. The axial velocity increases most quickly close to the wall of the tube where the temperature is highest. As z' becomes large the velocity field tends to a steady Poiseuille profile again, but the maximum velocity here is larger than it was in the inlet region, because of the expansion of the heated gas. The radial velocity increases quickly at small z' as the heated fluid moves away from the wall. In fact, the solution for u jumps from zero at $z'=0$ to a large negative value at the first calculated value of z' . The magnitude of this jump increases as the step size in z' is decreased so that as $z' \rightarrow 0$ there appears to be a singularity in the radial velocity solution. There are no physical singularities in our problem, which suggests that as $z' \rightarrow 0$ the solution in region II becomes non-uniform. Our asymptotic structure should include another region which smooths out the solution for small z' . Figure 9 shows the new asymptotic structure where extra regions, IIa and IIIa, have been added around the reaction front. Region III is drawn as a boundary layer to show more easily how the regions match together. Regions IIa and IIIa match the solutions of regions I and II when $r = O(1)$ and $r = O(Da^{-1/2})$ respectively. We describe the solution in these two new regions in the Appendix.

5.2. Region III

In this region, we solve (3.6) subject to the boundary conditions

$$U = 0, \quad W = 0, \quad T = 1 + Q \quad \text{at } R = 0, \tag{5.6a}$$

$$U, \quad W, \quad T \sim \text{Region IIIa solution} \quad \text{as } R \rightarrow \infty, Z \rightarrow 0. \tag{5.6b}$$

When written in terms of the region III variables, the region IIIa solutions are, to leading order,

$$T = 1 + Da^{-\gamma} \exp\left(-\frac{R^3}{18Z}\right),$$

$$\Psi = R^2 + 2Da^{-\gamma-1} \exp\left(-\frac{R^3}{18Z}\right),$$

where $\Psi = Da(2\psi + 1 + Da^{-1}C)$ and γ is a constant, which can be determined by higher-order matching. The streamfunction, Ψ , is related to the velocities, U and W ,

by

$$\frac{\partial \Psi}{\partial R} = \rho W, \quad \frac{\partial \Psi}{\partial Z} = -\rho U,$$

so conservation of mass, (3.6a), is satisfied. We look for a similarity solution of the form

$$T = T(\eta), \quad \Psi = (3Z)^{2/3} f(\eta), \quad \text{where} \quad \eta = \frac{R}{(3Z)^{1/3}}.$$

After removing the pressure terms from the momentum equations, by differentiating (3.6c) with respect to R , we obtain the governing equations in terms of T and f as

$$\begin{aligned} -2f \frac{df}{d\eta} \frac{d^2 T}{d\eta^2} - 4f \frac{d^2 f}{d\eta^2} \frac{dT}{d\eta} - 2f \frac{d^3 f}{d\eta^3} T - \left(\frac{df}{d\eta}\right)^2 \frac{dT}{d\eta} &= 12 \frac{dT}{d\eta} \frac{d^2 T}{d\eta^2} \frac{df}{d\eta} \\ + 16 \left(\frac{dT}{d\eta}\right)^2 \frac{d^2 f}{d\eta^2} + 4T \frac{d^3 T}{d\eta^3} \frac{df}{d\eta} + 16T \frac{d^2 T}{d\eta^2} \frac{d^2 f}{d\eta^2} + 20T \frac{dT}{d\eta} \frac{d^3 f}{d\eta^3} + 4T^2 \frac{d^4 f}{d\eta^4}, \end{aligned} \quad (5.7a)$$

$$4 \left(\frac{dT}{d\eta}\right)^2 + 4T \frac{d^2 T}{d\eta^2} + 2f \frac{dT}{d\eta} = 0. \quad (5.7b)$$

The boundary conditions at the wall, $\eta = 0$, are

$$f = 0, \quad \frac{df}{d\eta} = 0, \quad T = 1 + Q, \quad (5.8)$$

and as $\eta \rightarrow \infty$ we have

$$T \sim 1 + Da^{-\nu} \exp\left(-\frac{\eta^3}{6}\right), \quad (5.9a)$$

$$f \sim \eta^2 + \frac{1}{(3Z)^{2/3}} 2Da^{-\nu-1} \exp\left(-\frac{\eta^3}{6}\right). \quad (5.9b)$$

Although it appears that the boundary condition in (5.9b) is not in similarity form, we can assume that the term in Z in front of the exponential is part of the next-order solution in region IIIa, and therefore that

$$f \sim \eta^2 + 2Da^{-\nu-1} \exp\left(-\frac{\eta^3}{6}\right), \quad (5.10)$$

to leading order. Differentiating (5.9a) and (5.10) and eliminating the exponential terms gives the boundary conditions

$$\left. \begin{aligned} \frac{df}{d\eta} + \frac{\eta^2}{2} f &\sim 2\eta + \frac{\eta^4}{2} \\ \frac{d^2 f}{d\eta^2} + \frac{\eta^2}{2} \frac{df}{d\eta} + \eta f &\sim 2 + 2\eta^3 \\ \frac{dT}{d\eta} + \frac{\eta^2}{2} T &\sim \frac{\eta^2}{2} \end{aligned} \right\} \text{ as } \eta \rightarrow \infty.$$

We solved (5.7) numerically using a finite difference scheme and the solutions, when $Q = 1$, are shown in figure 10.

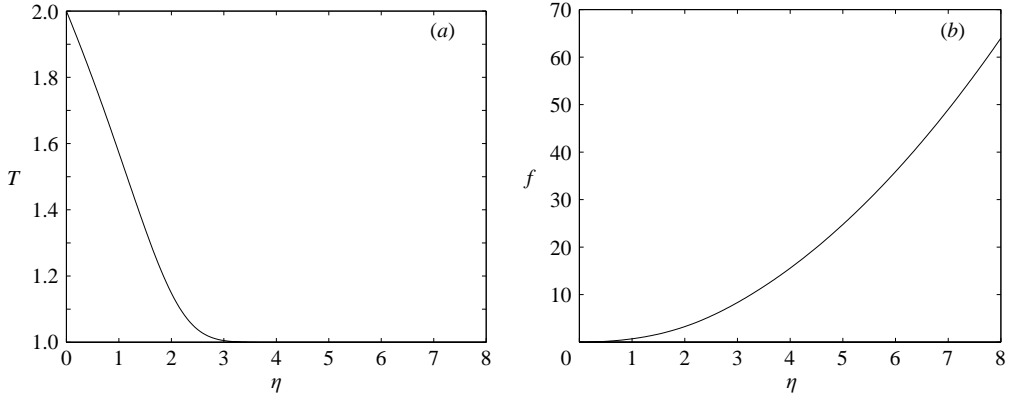


FIGURE 10. Solutions in region III. (a) Temperature, (b) streamfunction.

5.3. Region IV

The equations to be solved in this region are (3.4) with the boundary conditions, (3.5). Differentiating (3.4b) and (3.4c) with respect to \bar{Z} and \bar{R} respectively, we then combine them so as to remove the pressure terms. We define the streamfunction, $\bar{\psi}$, using

$$\frac{\partial \bar{\psi}}{\partial \bar{Z}} = -\rho \bar{U}, \quad \frac{\partial \bar{\psi}}{\partial \bar{R}} = \rho(\bar{W} + C),$$

and, setting $\mu = T, k = T$ as before, we substitute $\bar{\psi}$ and T for u, w', μ, k and ρ into the momentum and energy equations giving

$$\begin{aligned} & -\frac{\partial T}{\partial \bar{R}} \frac{\partial^2 \bar{\psi}}{\partial \bar{Z}^2} \frac{\partial \bar{\psi}}{\partial \bar{Z}} - \frac{1}{2} \frac{\partial^2 T}{\partial \bar{R} \partial \bar{Z}} \left(\frac{\partial \bar{\psi}}{\partial \bar{Z}} \right)^2 - \frac{1}{2} T \frac{\partial \bar{\psi}}{\partial \bar{Z}} \frac{\partial^3 \bar{\psi}}{\partial \bar{R} \partial \bar{Z}^2} + \frac{1}{2} \frac{\partial^2 T}{\partial \bar{Z}^2} \frac{\partial \bar{\psi}}{\partial \bar{R}} \frac{\partial \bar{\psi}}{\partial \bar{Z}} + \frac{\partial T}{\partial \bar{Z}} \frac{\partial \bar{\psi}}{\partial \bar{R}} \frac{\partial^2 \bar{\psi}}{\partial \bar{Z}^2} \\ & + \frac{1}{2} T \frac{\partial \bar{\psi}}{\partial \bar{R}} \frac{\partial^3 \bar{\psi}}{\partial \bar{Z}^3} - 2\bar{q}^2 \frac{\partial^2 T}{\partial \bar{R}^2} \frac{\partial \bar{\psi}}{\partial \bar{Z}} \frac{\partial \bar{\psi}}{\partial \bar{R}} - 4\bar{q}^2 \frac{\partial T}{\partial \bar{R}} \frac{\partial \bar{\psi}}{\partial \bar{Z}} \frac{\partial^2 \bar{\psi}}{\partial \bar{R}^2} - 2\bar{q}^2 T \frac{\partial \bar{\psi}}{\partial \bar{Z}} \frac{\partial^3 \bar{\psi}}{\partial \bar{R}^3} + 4\bar{q}^2 \frac{\partial T}{\partial \bar{Z}} \frac{\partial^2 \bar{\psi}}{\partial \bar{R}^2} \frac{\partial \bar{\psi}}{\partial \bar{R}} \\ & + 2\bar{q}^2 \frac{\partial^2 T}{\partial \bar{R} \partial \bar{Z}} \left(\frac{\partial \bar{\psi}}{\partial \bar{R}} \right)^2 + 2\bar{q}^2 T \frac{\partial \bar{\psi}}{\partial \bar{R}} \frac{\partial^3 \bar{\psi}}{\partial \bar{R}^2 \partial \bar{Z}} = 4 \frac{\partial T}{\partial \bar{R}} \frac{\partial^2 T}{\partial \bar{R} \partial \bar{Z}} \frac{\partial \bar{\psi}}{\partial \bar{Z}} + 2 \frac{\partial T}{\partial \bar{Z}} \frac{\partial^2 T}{\partial \bar{R}^2} \frac{\partial \bar{\psi}}{\partial \bar{Z}} \\ & + 2T \frac{\partial^3 T}{\partial \bar{R}^2 \partial \bar{Z}} \frac{\partial \bar{\psi}}{\partial \bar{Z}} + 16 \frac{\partial T}{\partial \bar{Z}} \frac{\partial T}{\partial \bar{R}} \frac{\partial^2 \bar{\psi}}{\partial \bar{R} \partial \bar{Z}} + 16T \frac{\partial^2 T}{\partial \bar{R} \partial \bar{Z}} \frac{\partial^2 \bar{\psi}}{\partial \bar{R} \partial \bar{Z}} + 10T \frac{\partial T}{\partial \bar{R}} \frac{\partial^3 \bar{\psi}}{\partial \bar{R} \partial \bar{Z}^2} \\ & + 10T \frac{\partial T}{\partial \bar{Z}} \frac{\partial^3 \bar{\psi}}{\partial \bar{R}^2 \partial \bar{Z}} + 4T^2 \frac{\partial^4 \bar{\psi}}{\partial \bar{R}^2 \partial \bar{Z}^2} + 4 \frac{\partial^2 T}{\partial \bar{R} \partial \bar{Z}} \frac{\partial T}{\partial \bar{Z}} \frac{\partial \bar{\psi}}{\partial \bar{R}} + 2 \frac{\partial T}{\partial \bar{R}} \frac{\partial^2 T}{\partial \bar{Z}^2} \frac{\partial \bar{\psi}}{\partial \bar{R}} + 2T \frac{\partial^3 T}{\partial \bar{R} \partial \bar{Z}^2} \frac{\partial \bar{\psi}}{\partial \bar{R}} \\ & + \frac{1}{2\bar{q}^2} \left(3 \frac{\partial T}{\partial \bar{Z}} \frac{\partial^2 T}{\partial \bar{Z}^2} \frac{\partial \bar{\psi}}{\partial \bar{Z}} + 4 \left(\frac{\partial T}{\partial \bar{Z}} \right)^2 \frac{\partial^2 \bar{\psi}}{\partial \bar{Z}^2} + T \frac{\partial^3 T}{\partial \bar{Z}^3} \frac{\partial \bar{\psi}}{\partial \bar{Z}} + 4T \frac{\partial^2 T}{\partial \bar{Z}^2} \frac{\partial^2 \bar{\psi}}{\partial \bar{Z}^2} \right. \\ & + 5T \frac{\partial T}{\partial \bar{Z}} \frac{\partial^3 \bar{\psi}}{\partial \bar{Z}^3} + T^2 \frac{\partial^4 \bar{\psi}}{\partial \bar{Z}^4} \left. \right) + 8\bar{q}^2 \left(3 \frac{\partial T}{\partial \bar{R}} \frac{\partial^2 T}{\partial \bar{R}^2} \frac{\partial \bar{\psi}}{\partial \bar{R}} + 4 \left(\frac{\partial T}{\partial \bar{R}} \right)^2 \frac{\partial^2 \bar{\psi}}{\partial \bar{R}^2} \right. \\ & \left. + T \frac{\partial^3 T}{\partial \bar{R}^3} \frac{\partial \bar{\psi}}{\partial \bar{R}} + 4T \frac{\partial^2 T}{\partial \bar{R}^2} \frac{\partial^2 \bar{\psi}}{\partial \bar{R}^2} + 5T \frac{\partial T}{\partial \bar{R}} \frac{\partial^3 \bar{\psi}}{\partial \bar{R}^3} + T^2 \frac{\partial^4 \bar{\psi}}{\partial \bar{R}^4} \right), \end{aligned} \tag{5.11a}$$

and

$$-\frac{\partial \bar{\psi}}{\partial \bar{Z}} \frac{\partial T}{\partial \bar{R}} + \frac{\partial \bar{\psi}}{\partial \bar{R}} \frac{\partial T}{\partial \bar{Z}} = 4 \left(\frac{\partial T}{\partial \bar{R}} \right)^2 + 4T \frac{\partial^2 T}{\partial \bar{R}^2} + \frac{1}{\bar{q}^2} \left(\frac{\partial T}{\partial \bar{Z}} \right)^2 + \frac{1}{\bar{q}^2} T \frac{\partial^2 T}{\partial \bar{Z}^2}. \tag{5.11b}$$

The boundary conditions become

$$\bar{\psi} = 0, \quad \frac{\partial \bar{\psi}}{\partial \bar{R}} = \frac{C}{T}, \quad T^2 \frac{\partial T}{\partial \bar{R}} = \begin{cases} \frac{1}{2}(T - 1 - Q), & \bar{Z} > 0, \\ 0, & \bar{Z} < 0, \end{cases} \quad \text{at } \bar{R} = 0, \quad (5.12a)$$

$$\bar{\psi}, \quad \frac{\partial \bar{\psi}}{\partial \bar{R}}, \quad T \rightarrow \text{Region IIIa solution} \quad \text{as } \bar{R} \rightarrow \infty, \quad (5.12b)$$

$$\frac{\partial \bar{\psi}}{\partial \bar{Z}} \rightarrow 0, \quad \bar{\psi} \sim C\bar{R} + \bar{R}^2, \quad T \rightarrow 1 \quad \text{as } \bar{Z} \rightarrow -\infty, \quad (5.12c)$$

$$\bar{\psi}, \quad \frac{\partial \bar{\psi}}{\partial \bar{Z}}, \quad T \rightarrow \text{Region III solution} \quad \text{as } \bar{Z} \rightarrow \infty. \quad (5.12d)$$

Since we wish to solve this system numerically, we must smooth the discontinuous boundary condition at $\bar{R} = 0$ to reduce errors close to $\bar{Z} = 0$. We replace the Heaviside function with a hyperbolic tangent giving

$$T^2 \frac{\partial T}{\partial \bar{R}} = \frac{1}{2}(T - 1 - Q) \frac{1}{2}[1 + \tanh(\delta^{-1}\bar{Z})] \quad \text{at } \bar{R} = 0,$$

where δ is a small parameter of the order of the step size in \bar{Z} . We return to the solutions found in regions IIIa and III to obtain boundary conditions as $\bar{R} \rightarrow \infty$ and $\bar{Z} \rightarrow \infty$ respectively.

5.3.1. Boundary conditions as $\bar{R} \rightarrow \infty$

The region IIIa solutions are

$$T = 1 + Da^{-\gamma} \exp(-Da\phi),$$

$$\psi = -\frac{1}{2} - Da^{-1} \frac{C}{2} + Da^{-3/2} \frac{CR^*}{2} + Da^{-1} \frac{R^{*2}}{2} + Da^{-\gamma-2} \exp(-Da\phi),$$

where ϕ is given by (A 8) and t is a root of (A 7). In terms of the region IV variables $R^* = Da^{-1/2}\bar{R}$ and $Z^* = Da^{-1/2}\bar{Z}$, and $t \equiv t(R^*/Z^*)$ therefore remains of $O(1)$ when written in terms of \bar{R} , \bar{Z} . It follows that $\phi = O(Da^{-1})$ and the region IIIa solutions written in terms of the region IV variables are

$$T = 1 + Da^{-\gamma} \exp(-\bar{\phi}),$$

$$\bar{\psi} = C\bar{R} + \bar{R}^2 + 2Da^{-\gamma} \exp(-\bar{\phi}),$$

to leading order, where $\bar{\phi} = Da\phi$ and $\bar{\psi} = Da^2(2\psi + 1 + Da^{-1}C)$. Differentiating these expressions and eliminating exponential terms we obtain

$$\frac{\partial T}{\partial \bar{R}} + \frac{\partial \bar{\phi}}{\partial \bar{R}} T \sim \frac{\partial \bar{\phi}}{\partial \bar{R}},$$

$$\frac{\partial \bar{\psi}}{\partial \bar{R}} + \frac{\partial \bar{\phi}}{\partial \bar{R}} \bar{\psi} \sim C + 2\bar{R} + (C\bar{R} + \bar{R}^2) \frac{\partial \bar{\phi}}{\partial \bar{R}},$$

$$\frac{\partial^2 \bar{\psi}}{\partial \bar{R}^2} + \frac{\partial \bar{\phi}}{\partial \bar{R}} \frac{\partial \bar{\psi}}{\partial \bar{R}} + \frac{\partial^2 \bar{\phi}}{\partial \bar{R}^2} \bar{\psi} \sim 2 + (C + 2\bar{R}) \frac{\partial \bar{\phi}}{\partial \bar{R}} + (C\bar{R} + \bar{R}^2) \frac{\partial^2 \bar{\phi}}{\partial \bar{R}^2},$$

which are the boundary conditions for T and $\bar{\psi}$ as $\bar{R} \rightarrow \infty$.

5.3.2. Boundary conditions as $\bar{Z} \rightarrow \infty$

The region IV solutions must match to the region III solutions as $\bar{Z} \rightarrow \infty$. In region III, we obtained solutions in terms of a similarity variable η , and in terms of

the region IV variables, \bar{R} and \bar{Z} , the region III solutions are given by

$$T = T(\eta), \quad \bar{\psi} = (3\bar{Z})^{2/3} f(\eta), \quad \text{where } \eta = \frac{\bar{R}}{(3\bar{Z})^{1/3}}.$$

We require a boundary condition as $\bar{Z} \rightarrow \infty$, so we only need to know the structure of the solution in region III as $\eta \rightarrow 0$. We let

$$T = 1 + Q + \hat{T}, \quad f = \hat{f},$$

where \hat{T}, \hat{f} are small, and substitute into (5.7), which gives

$$\frac{d^2 \hat{T}}{d\eta^2} = 0, \quad \frac{d^4 \hat{f}}{d\eta^4} = 0,$$

at leading order. The solutions are

$$\hat{T} = A_1 \eta, \quad \hat{f} = A_2 \eta^3 + B_2 \eta^2,$$

where we have satisfied the boundary conditions at $\eta = 0$, given in (5.8). The constants A_1, A_2 and B_2 are unknown but can be eliminated from the solutions to give mixed boundary conditions of the form

$$\left. \begin{aligned} \eta \frac{dT}{d\eta} - T &\rightarrow -(1 + Q) \\ \frac{\eta^2}{6} \frac{d^2 f}{d\eta^2} - \frac{2\eta}{3} \frac{df}{d\eta} + f &\rightarrow 0 \\ -\eta^3 \frac{d^3 f}{d\eta^3} + 4\eta^2 \frac{d^2 f}{d\eta^2} - 10\eta \frac{df}{d\eta} + 12f &\rightarrow 0 \end{aligned} \right\} \text{ as } \eta \rightarrow 0.$$

When these are written in terms of $\bar{\psi}, \bar{R}$ and \bar{Z} we obtain

$$\left. \begin{aligned} 3\bar{Z} \frac{\partial T}{\partial \bar{Z}} + T &\rightarrow 1 + Q \\ 3\bar{Z} \frac{\partial^2 \bar{\psi}}{\partial \bar{Z}^2} + 4 \frac{\partial \bar{\psi}}{\partial \bar{Z}} &\rightarrow 0 \\ 3\bar{Z}^2 \frac{\partial^3 \bar{\psi}}{\partial \bar{Z}^3} + 10\bar{Z} \frac{\partial^2 \bar{\psi}}{\partial \bar{Z}^2} + 4 \frac{\partial \bar{\psi}}{\partial \bar{Z}} &\rightarrow 0 \end{aligned} \right\} \text{ as } \bar{Z} \rightarrow \infty.$$

These boundary conditions complete the system, which we solved using the numerical scheme outlined below.

5.4. Numerical solution

The temperature is given by an array with components, $T_{i,j}$, representing the temperature at \bar{R}_i, \bar{Z}_j on a uniform grid of $m \times n$ points. Similarly, the streamfunction is given by $\psi_{i,j}$. We truncate the semi-infinite range in \bar{R} and infinite range in \bar{Z} so that $0 \leq \bar{R}_j \leq 12$ and $-20 \leq \bar{Z}_j \leq 60$. We calculate the derivatives of the discrete function $T_{i,j}$ using a five-point centred difference formula in the bulk and a four-point biased stencil formulae at the edges of the solution grid. In discrete form, (5.11) are given by

$$A_j \Delta \psi_{j+2} + B_j \Delta \psi_{j+1} + C_j \Delta \psi_j + D_j \Delta \psi_{j-1} + E_j \Delta \psi_{j-2} = R_j, \tag{5.13a}$$

$$a_j \Delta T_{j+2} + b_j \Delta T_{j+1} + c_j \Delta T_j + d_j \Delta T_{j-1} + e_j \Delta T_{j-2} = r_j, \tag{5.13b}$$

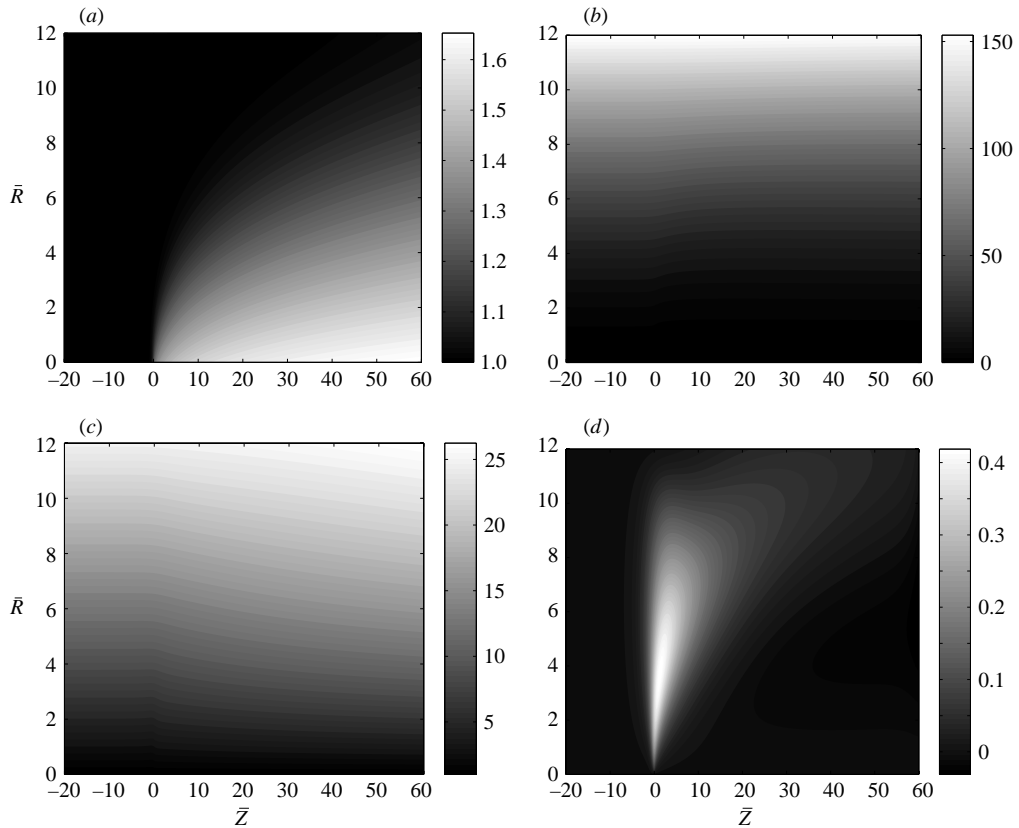


FIGURE 11. Region IV solutions for $C = 1$. (a) Temperature, (b) streamfunction, (c) axial velocity, (d) radial velocity.

where \mathbf{A}_j , \mathbf{a}_j , etc. are known $m \times m$ arrays, $\mathbf{T}_j (=T_{i,j})$, $\boldsymbol{\psi}_j (= \psi_{i,j})$, \mathbf{R}_j and \mathbf{r}_j are vectors of length m and $0 \leq j \leq n$. This system was solved using a Thomas algorithm (Fletcher 1991).

We checked the mesh dependency of the numerical solution, and found the maximum relative errors between solutions on different grids to be of $O(10^{-2})$. To minimize these errors without increasing computational time too much we set $d\bar{R} = 0.1$ and $d\bar{Z} = 0.1$. We obtained results on this grid for a range of values of the wave speed, C , heat of reaction, Q , and mass flow rate, \bar{q} . We began by holding Q and \bar{q} constant and calculated the variation of critical temperature, T_c , with wave speed, C .

5.4.1. Variation of wave speed with critical temperature

We obtained results for a range of values of C . The results for $C = 1$ and $C = 2$ are shown in figures 11 and 12 respectively. The temperature profiles are qualitatively similar to those obtained in the constant-density model, see figure 5. At the point where the reaction starts, $\bar{Z} = 0$, there is a displacement of the streamlines due to the changing density and viscosity of the gas mixture. This causes a large increase in the radial velocity around $\bar{R} = 0$, $\bar{Z} = 0$ as the heated gas moves away from the wall. This behaviour agrees with what we found in region II. The axial velocity also changes rapidly around $\bar{R} = 0$, $\bar{Z} = 0$ and increases as \bar{Z} increases. From the solutions obtained at each value of the wave speed, C , we noted the critical temperature, T_c .

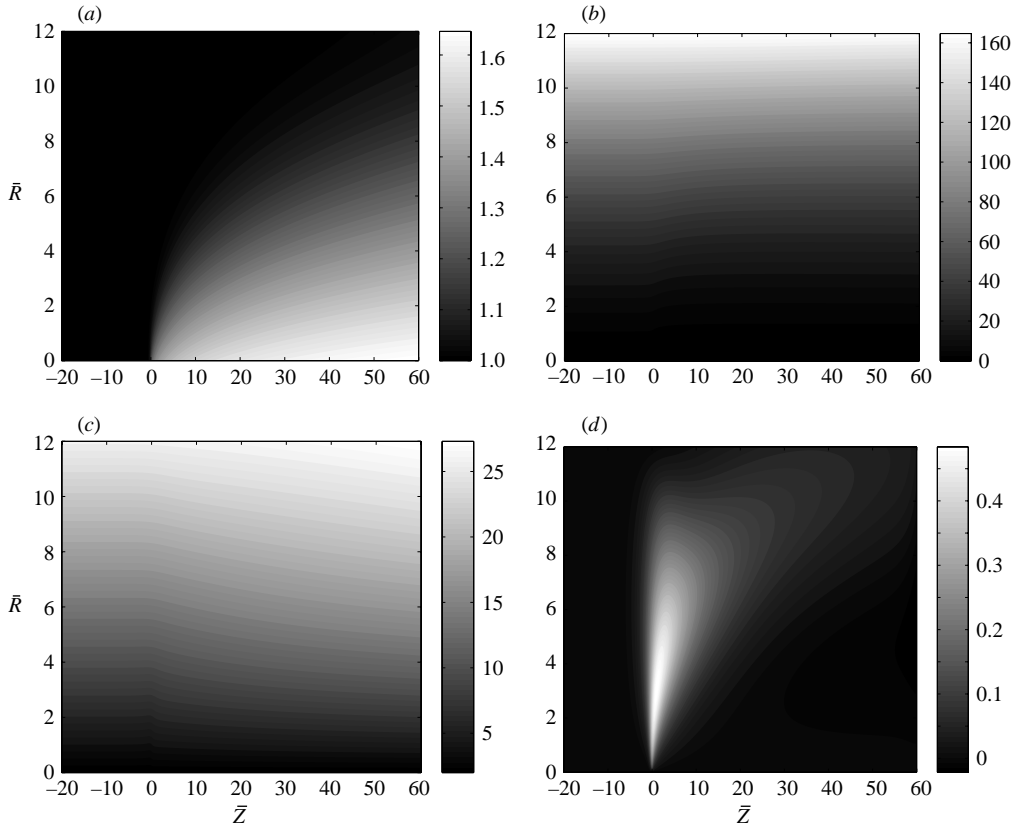


FIGURE 12. Region IV solutions for $C = 2$. (a) Temperature, (b) streamfunction, (c) axial velocity, (d) radial velocity.

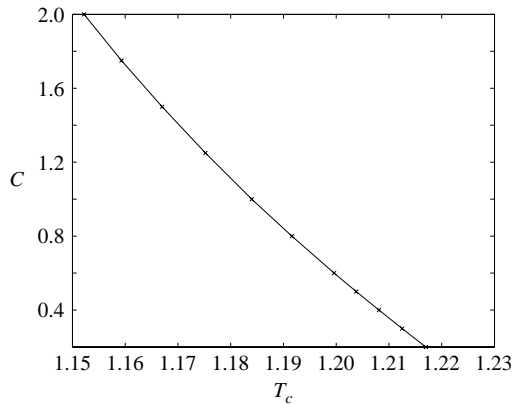


FIGURE 13. Plot of the wave speed, C , against critical temperature of reaction, T_c .

The variation of C with T_c is plotted in figure 13. The result is qualitatively similar to that obtained in the constant-density model (see figure 6). A quantitative comparison of the two models is made by looking at the variation of T_c with heat released from the reaction, Q .

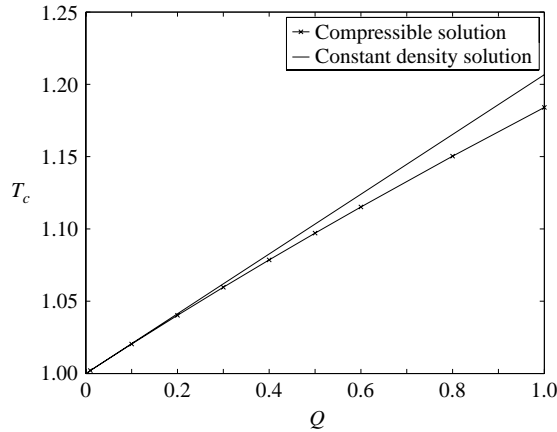


FIGURE 14. Plot of the critical temperature, T_c , against heat released from the reaction, Q .

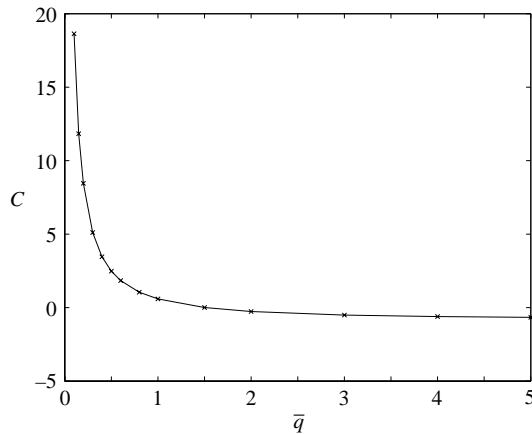


FIGURE 15. Plot of wave speed, C , against mass flow rate of fuel, \bar{q} .

5.4.2. Variation of critical temperature with Q

Our results for Q sufficiently small must be consistent with the results we obtained using the constant-density approximation, $Q \ll 1$, when the temperature was given by $T = 1 + Q\hat{T}$. For $C = 1$, $\bar{q} = 1$ we found that the critical temperature is $\hat{T}_c = 0.21$, and it follows that for small Q the results of the variable-density model for T_c should be given approximately by $T_c = 1 + 0.21Q$. Figure 14 shows that solutions of the variable-density model are in very good agreement with the constant-density result when Q is small, but that for larger values of Q the constant-density approximation predicts too high a value for the critical temperature.

5.4.3. Variation of wave speed with mass flow rate

We now fix Q and T_c and vary the mass flow rate, \bar{q} , to see its effect on the wave speed, C . The results for $Q = 1$, $T_c = 1.2$ are shown in figure 15. They are qualitatively similar to the constant-density results shown in figure 7. The wave speed decreases as \bar{q} increases and reaches zero when $\bar{q} \approx 1.51$. For $\bar{q} > 1.51$ the wave speed is negative, so for sufficiently large mass flow rates, the reaction front propagates towards the outlet of the tube and will eventually be blown out.

Percentage of platinum on surface of tube	0.12	0.19	0.29	0.32
Average wave speed ($\text{m s}^{-1} \times 10^{-4}$)	0.80	1.65	1.95	2.65

TABLE 3. Experimental data.

Mass flow rate ($\text{kg s}^{-1} \times 10^{-6}$)	1.09	1.63	2.18	2.72	3.27
Average wave speed ($\text{m s}^{-1} \times 10^{-4}$)	1.90	2.45	2.65	2.45	2.20

TABLE 4. Experimental data.

6. Qualitative comparison with preliminary experiment

At present, only very limited experimental data are available. However, we can qualitatively compare the preliminary results obtained by Lefevre (2003) with our model. The experiment described in §1 was carried out several times using tubes of zirconia coated on their inner surface with varying amounts of platinum catalyst. The variation of the wave speed, c^* , with the amount of catalyst on the surface of the tube was measured and the results are given in table 3. Two results linking the percentage of platinum on the tube to the critical temperature were obtained experimentally, and from these a linear relationship between the percentage platinum concentration, P , and critical temperature, T^* , was obtained, namely

$$T^* = -500P + 948.$$

Experimentally, the heat released from the reaction is large and gives $Q = O(10)$. However, our models are not valid for large values of Q , due to the simplification made in §2.3. By comparing the $Q = O(1)$ and constant-density results, we know that as Q increases to $O(1)$ values the constant-density model predicts too high a critical temperature at constant wave speed, see figure 14. From this we infer that as Q increases to $O(10)$ values the $O(1)$ model will tend to overestimate the critical temperature. Therefore, as the wave speed decreases with increasing critical temperature, figure 13, we conclude that the values of wave speed at each T_c predicted by the $Q = O(1)$ model will be too high.

Indeed, the experimentally measured values of the wave speed are smaller than those predicted by our model; however, the qualitative behaviour of the results is the same: the wave speed decreases with increasing critical temperature.

Further experimental data relate the mass flow rate of fuel into the zirconia tube to the wave speed, as shown in table 4. The experiments in this case were carried out using a tube coated with 0.4% platinum catalyst. Experimentally, as the mass flow rate increases the wave speed increases until q_{in} reaches a value of about $2 \times 10^{-6} \text{ kg s}^{-1}$. After this point, the wave speed decreases as the mass flow rate increases. Our asymptotic solution is based on a large mass flow rate, so we only expect to obtain agreement with experiment at sufficiently large values of q_{in} . Although, the wave speeds obtained from our model are again too large, the qualitative behaviour of the solutions for $q_{\text{in}} > 2 \times 10^{-6}$ is correct.

Furthermore, the numerical results show that for q_{in} sufficiently large the wave speed becomes negative. Although the available experimental data do not show this, it has been reported that on increasing the mass flow rate sufficiently, the wave stops propagating towards the inlet and the reaction front is blown out of the tube.

We note that the work by Lefevre (2003) is at a very early stage and that the results need to be verified. Further work is also required to extend the experimental results

to include a wider range of wave speeds, temperatures and mass flow rates, especially at large values of q_{in} .

7. Conclusion

We have shown that the reaction front, or surface-catalysed flame, that can propagate in a zirconia tube can be modelled as a steady travelling wave. We found that the speed of this reaction front decreases as the temperature at which the reaction starts, the critical temperature, T_c , increases, and as the mass flow rate of fuel entering the tube, q_{in} , increases. Having found a solution using the constant-density approximation, we found that for larger values of the heat released by the reaction, $Q = O(1)$, the constant-density model predicts too high a value for the wave speed, C . Also, as expected, the wave speeds predicted by our model were higher than those given by the experimental data. There is, however, qualitative agreement with experiment. At a critical value of the mass flow rate, $\bar{q} = \bar{q}_c$, the wave speed is zero and, as \bar{q} increases past this critical value, C becomes negative. Hence, for $\bar{q} > \bar{q}_c$ the reaction front propagates towards the outlet of the tube and the reaction will eventually be blown out. The experimental data available at present show an increase in wave speed with mass flow rate, q_{in} , for small q_{in} and then a decrease as q_{in} increases past a value of about $2 \times 10^{-6} \text{ kg s}^{-1}$. Data at large values of the mass flow rate have not been obtained, although it has been observed experimentally that for large enough values of the mass flow rate the wave propagates towards the outlet of the tube and the reaction is blown out.

There are several possible extensions and improvements to the work in this paper. We hope to obtain better agreement between theory and experimental results by solving for larger values of Q , which could be done by considering the asymptotic solution for $Q \gg 1$. We could also consider some of the effects that we have currently neglected, such as heat loss from the tube, more realistic chemistry and electrochemistry. In addition to any further mathematical analysis, a validation of the current experimental results and more detailed experimental work is required. In particular, further results showing the variation of wave speed with large mass flow rate are needed to verify that the wave propagates backwards for sufficiently large q_{in} .

Appendix. The additional asymptotic regions when $Q = O(1)$

A.1. Equations in region IIa

We return to the original equations, (3.1), to determine the scalings for this new region. We remove the pressure terms from the momentum equations by differentiating (3.1b) with respect to z' and (3.1c) with respect to r and combining the resulting equations. We rescale the axial coordinate using $z' = \zeta \tilde{z}$ with $\zeta \ll 1$, and look for a WKB solution of the form

$$T = 1 + \epsilon^{\tilde{\gamma}} \exp\left(-\frac{\varphi(r, \tilde{z})}{\epsilon}\right),$$

$$\psi = -\left(r^2 - \frac{r^4}{2}\right) - Da^{-1} \frac{Cr^2}{2} + \tilde{\epsilon}^{\tilde{\delta}} \exp\left(-\frac{\tilde{\varphi}(r, \tilde{z})}{\tilde{\epsilon}}\right),$$

where ϵ and $\tilde{\epsilon}$ are small parameters and $\tilde{\gamma}$ and $\tilde{\delta}$ are constants. The streamfunction, ψ , is defined in exactly the same way as in region II. We substitute the expressions for T and ψ into the momentum and energy equations. This gives the leading-order

equations for φ and $\tilde{\varphi}$ as

$$-2(1-r^2)\frac{\partial\varphi}{\partial\tilde{z}} = \left(\frac{\partial\varphi}{\partial r}\right)^2 + \frac{1}{\bar{q}^2}\left(\frac{\partial\varphi}{\partial\tilde{z}}\right)^2, \tag{A 1a}$$

$$\begin{aligned} & -2(1-r^2)\frac{\partial\tilde{\varphi}}{\partial\tilde{z}} \left[\left(\frac{\partial\tilde{\varphi}}{\partial\tilde{z}}\right)^2 + \bar{q}^2\left(\frac{\partial\tilde{\varphi}}{\partial r}\right)^2 \right] \exp\left(-\frac{\tilde{\varphi}}{\tilde{\epsilon}}\right) - 4r\bar{q}^2(1-r^2)^2\frac{\partial\varphi}{\partial\tilde{z}}\frac{\partial\varphi}{\partial r} \exp\left(-\frac{\varphi}{\epsilon}\right) \\ & = \left[2\left(\frac{\partial\tilde{\varphi}}{\partial r}\right)^2\left(\frac{\partial\tilde{\varphi}}{\partial\tilde{z}}\right)^2 + \frac{1}{\bar{q}^2}\left(\frac{\partial\tilde{\varphi}}{\partial\tilde{z}}\right)^4 + \bar{q}^2\left(\frac{\partial\tilde{\varphi}}{\partial r}\right)^4 \right] \exp\left(-\frac{\tilde{\varphi}}{\tilde{\epsilon}}\right) \\ & + 2r(1-r^2)\bar{q}^2\frac{\partial\varphi}{\partial r} \left[\frac{1}{\bar{q}^2}\left(\frac{\partial\varphi}{\partial\tilde{z}}\right)^2 + \left(\frac{\partial\varphi}{\partial r}\right)^2 \right] \exp\left(-\frac{\varphi}{\epsilon}\right), \end{aligned} \tag{A 1b}$$

where we have taken $\epsilon = \tilde{\epsilon} = \zeta = Da^{-2}$ and $\tilde{\delta} = \tilde{\gamma} + 1$. The viscosity and thermal conductivity are $\mu = T$ and $k = T$, as they were in region II. Using (A 1a), we cancel all the terms in φ in (A 1b), which then reduces to

$$-2(1-r^2)\frac{\partial\tilde{\varphi}}{\partial\tilde{z}} = \left(\frac{\partial\tilde{\varphi}}{\partial r}\right)^2 + \frac{1}{\bar{q}^2}\left(\frac{\partial\tilde{\varphi}}{\partial\tilde{z}}\right)^2,$$

and is of the same form as (A 1a). The boundary conditions on φ and $\tilde{\varphi}$ are identical,

$$\exp\left(-\frac{\varphi}{\epsilon}\right) \rightarrow 0 \quad \text{as} \quad \tilde{z} \rightarrow -\infty \quad \text{and} \quad r \rightarrow 0,$$

so we have $\tilde{\varphi} = \varphi$.

The solution in region IIa does not explicitly affect the solution in region IV, since region IIa has no boundaries with region IV (see figure 9). The wave speed is determined in region IV and the variation of C is the main result we wish to obtain from our model. Because of this, and the complexity of the problem, we will not look for a solution of (A 1a). However, in the correct asymptotic limits, the solution of (A 1a) satisfies the appropriate boundary conditions. We will show that, as $r \rightarrow 1$ and $z' \rightarrow 0$, (A 1a) reduces to the equations in region IIIa, which we now derive.

A.2. Region IIIa

We again return to the original equations, (3.1), to determine the scalings for this new region. We eliminate the pressure from the momentum equations as we did in region IIa. We rescale using $r^2 = 1 - \xi R^*$, $z' = \tilde{\xi} Z^*$, with $\xi, \tilde{\xi} \ll 1$, and look for a WKB solution of the form

$$\begin{aligned} T &= 1 + \varepsilon^\gamma \exp\left(-\frac{\phi(R^*, Z^*)}{\varepsilon}\right), \\ \psi &= -\left(r^2 - \frac{r^4}{2}\right) - Da^{-1}\frac{Cr^2}{2} + \eta^\delta \exp\left(-\frac{\tilde{\phi}(R^*, Z^*)}{\eta}\right), \end{aligned}$$

where ε and η are small parameters and γ and δ are constants. The streamfunction, ψ , is defined in exactly the same way as in regions II and IIa. Substituting the expressions for T and ψ into the momentum and energy equations, the leading-order equations for ϕ and $\tilde{\phi}$ are

$$-2R^*\frac{\partial\phi}{\partial Z^*} = 4\left(\frac{\partial\phi}{\partial R^*}\right)^2 + \frac{1}{\bar{q}^2}\left(\frac{\partial\phi}{\partial Z^*}\right)^2, \tag{A 2a}$$

$$\begin{aligned}
 & -2R^* \frac{\partial \tilde{\phi}}{\partial Z^*} \left[\left(\frac{\partial \tilde{\phi}}{\partial Z^*} \right)^2 + 4\bar{q}^2 \left(\frac{\partial \tilde{\phi}}{\partial R^*} \right)^2 \right] \exp\left(-\frac{\tilde{\phi}}{\eta}\right) + 8R^{*2} \bar{q}^2 \frac{\partial \phi}{\partial Z^*} \frac{\partial \phi}{\partial R^*} \exp\left(-\frac{\phi}{\varepsilon}\right) \\
 & = \left[8 \left(\frac{\partial \tilde{\phi}}{\partial R^*} \right)^2 \left(\frac{\partial \tilde{\phi}}{\partial Z^*} \right)^2 + \frac{1}{\bar{q}^2} \left(\frac{\partial \tilde{\phi}}{\partial Z^*} \right)^4 + 16\bar{q}^2 \left(\frac{\partial \tilde{\phi}}{\partial R^*} \right)^4 \right] \exp\left(-\frac{\tilde{\phi}}{\eta}\right) \\
 & \quad - 4R^* \bar{q}^2 \frac{\partial \phi}{\partial R^*} \left[\frac{1}{\bar{q}^2} \left(\frac{\partial \phi}{\partial Z^*} \right)^2 + 4 \left(\frac{\partial \phi}{\partial R^*} \right)^2 \right] \exp\left(-\frac{\phi}{\varepsilon}\right), \tag{A 2b}
 \end{aligned}$$

where we have taken $\varepsilon = \eta = \tilde{\xi} \xi^{-3}$, $\bar{q} = q \tilde{\xi} \xi^{-1}$ and $\delta = \gamma + 2$. We fixed the magnitudes of these parameters by matching next-order terms in the energy equation. The richest balance is obtained when $\varepsilon = Da^{-1}$, $\xi = Da^{-1/2}$ and $\tilde{\xi} = Da^{-5/2}$. All the terms in (A 2b) containing ϕ cancel due to (A 2a), and (A 2b) reduces to

$$-2R^* \frac{\partial \tilde{\phi}}{\partial Z^*} = 4 \left(\frac{\partial \tilde{\phi}}{\partial R^*} \right)^2 + \frac{1}{\bar{q}^2} \left(\frac{\partial \tilde{\phi}}{\partial Z^*} \right)^2,$$

which is of the same form as (A 2a). The boundary conditions on ϕ and $\tilde{\phi}$ are identical,

$$\exp\left(-\frac{\phi}{\varepsilon}\right) \rightarrow 0 \quad \text{as } Z^* \rightarrow -\infty \quad \text{and } R^* \rightarrow \infty, \tag{A 3}$$

so we conclude that $\tilde{\phi} = \phi$.

A.2.1. Matching regions IIa and IIIa

The equation for ϕ in region IIa, (A 1a), should reduce to (A 2a) as $r \rightarrow 1$, $\tilde{z} \rightarrow 0$. Substituting for R^* and Z^* into (A 1a), we obtain

$$-2R^* \frac{\partial \phi}{\partial Z^*} = 4 \left(\frac{\partial \phi}{\partial R^*} \right)^2 + \frac{1}{\bar{q}^2} \left(\frac{\partial \phi}{\partial Z^*} \right)^2,$$

to leading order, where $\phi = Da^{1/2} \varphi$, and this is identical to (A 2a). The region IIa solution for the temperature is given by

$$T = 1 + Da^{-\tilde{\gamma}} \exp(-Da \varphi),$$

and in region IIIa we have

$$T = 1 + Da^{-\gamma/2} \exp(-Da^{1/2} \phi),$$

and, since $\phi = Da^{1/2} \varphi$, the exponential term is the same in both regions. The term in front of the exponential will be affected by the next-order solutions for ϕ and φ so at leading order it is not necessary for these two terms to match. We conclude that the leading-order temperatures in regions IIa and IIIa match as R^* , $|Z^*| \rightarrow \infty$. In a similar manner it follows that the streamfunctions also match to leading order.

A.3. Solution for ϕ

We solve (A 2a) by writing it as a system of characteristic ordinary differential equations in terms of a parameter t (John 1978). Rewriting (A 2a) as

$$F(R^*, Z^*, \phi, \mathcal{P}, \mathcal{Q}) = 2R^* \mathcal{Q} + 4\mathcal{P}^2 + \frac{1}{\bar{q}^2} \mathcal{Q}^2 = 0, \tag{A 4}$$

where

$$\mathcal{P} = \frac{\partial \phi}{\partial R^*}, \quad \mathcal{Q} = \frac{\partial \phi}{\partial Z^*},$$

the characteristic equations are then given by

$$\frac{dR^*}{dt} = 8\mathcal{P}, \quad \frac{dZ^*}{dt} = 2R^* + \frac{2\mathcal{Q}}{\bar{q}^2}, \quad \frac{d\phi}{dt} = -2R^*\mathcal{Q}, \quad \frac{d\mathcal{P}}{dt} = -2\mathcal{Q}, \quad \frac{d\mathcal{Q}}{dt} = 0.$$

On solving these, we obtain

$$R^* = -8at^2 + 8bt + f, \tag{A 5a}$$

$$Z^* = -\frac{16}{3}at^3 + 8bt^2 + 2ft + \frac{2at}{\bar{q}^2} + g, \tag{A 5b}$$

$$\phi = \frac{16}{3}a^2t^3 - 8abt^2 - 2aft + h, \tag{A 5c}$$

$$\mathcal{P} = -2at + b, \tag{A 5d}$$

$$\mathcal{Q} = a, \tag{A 5e}$$

where a, b, f, g and h are functions of s that satisfy

$$\frac{dh}{ds} = b(s)\frac{df}{ds} + a(s)\frac{dg}{ds},$$

and (A 4) when $t=0$. A simple solution is

$$a = s, \quad b = s, \quad f = -2s - \frac{1}{2\bar{q}^2}s, \quad g = 2s + \frac{1}{2\bar{q}^2}s, \quad h = 0,$$

and (A 5a-c) become

$$R^* = -8st^2 + 8st - 2s - \frac{1}{2\bar{q}^2}s, \tag{A 6a}$$

$$Z^* = -\frac{16}{3}st^3 + 8st^2 - 4st + \frac{1}{\bar{q}^2}st + 2s + \frac{1}{2\bar{q}^2}s, \tag{A 6b}$$

$$\phi = \frac{16}{3}s^2t^3 - 8s^2t^2 + 4s^2t + \frac{1}{\bar{q}^2}s^2t. \tag{A 6c}$$

Dividing (A 6b) by (A 6a) eliminates s and, after rearranging the result, we obtain a cubic equation for t ,

$$t^3 - \frac{3}{2} \left(\frac{Z^*}{R^*} + 1 \right) t^2 + \frac{3}{2} \left(\frac{Z^*}{R^*} + \frac{1}{2} - \frac{1}{4\bar{q}^2} \right) t - \frac{3}{8} \left(\frac{Z^*}{R^*} + \frac{1}{4\bar{q}^2} \frac{Z^*}{R^*} + 1 + \frac{1}{4\bar{q}^2} \right) = 0. \tag{A 7}$$

On solving this, the solution for t can be substituted into (A 6c) and, using (A 6a) to eliminate s , we obtain

$$\phi = \frac{4\bar{q}^2 R^{*2} (16\bar{q}^2 t^3 - 24\bar{q}^2 t^2 + 12\bar{q}^2 t + 3t)}{3(16\bar{q}^2 t^2 - 16\bar{q}^2 t + 4\bar{q}^2 + 1)^2}. \tag{A 8}$$

The appropriate root of (A 7) is determined by the boundary conditions on ϕ as $Z^* \rightarrow -\infty$ and $R^* \rightarrow \infty$, given in (A 3). Figure 16 shows the real and imaginary parts of the roots of (A 7) for a range of values of $x = Z^*/R^*$. We take the real, positive root, which for $x > -1$ is t_1 , shown as the solid line in the figure, and for $x \leq -1$ is t_2 , the dot-dashed curve in the figure. The solution for ϕ , after making this choice for t , is shown in figure 17.

The solution in region IIIa is therefore given by

$$T = 1 + Da^{-\gamma} \exp(-Da\phi),$$

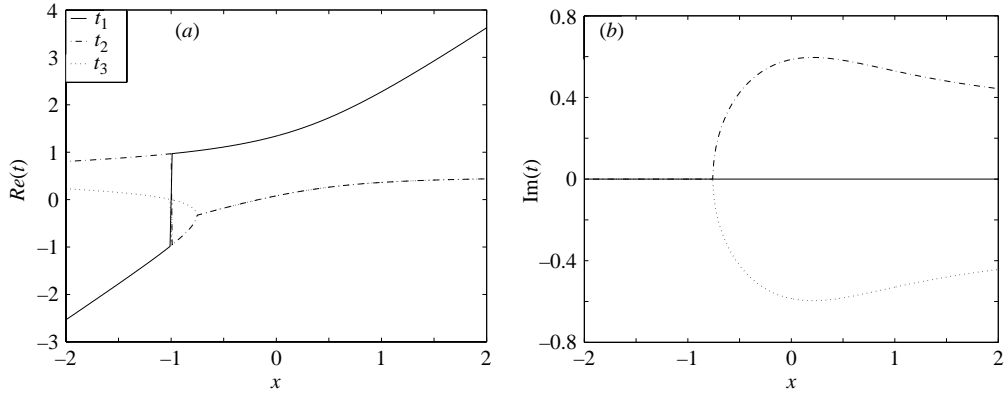


FIGURE 16. Roots of the cubic equation, (A 7). (a) $Re(t)$, (b) $Im(t)$.

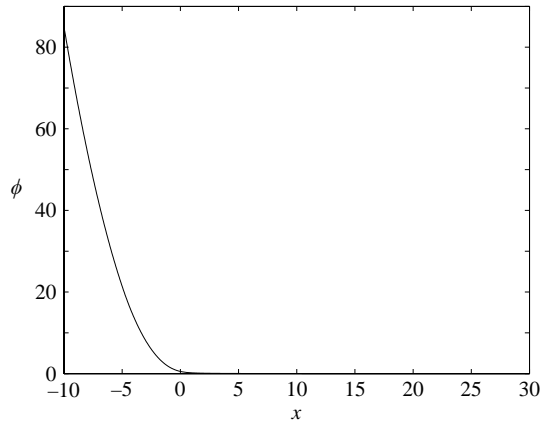


FIGURE 17. Solution for ϕ .

$$\psi = -\frac{1}{2} - Da^{-1}\frac{C}{2} + Da^{-3/2}\frac{CR^*}{2} + Da^{-1}\frac{R^{*2}}{2} + Da^{-(\gamma+2)}\exp(-Da\phi),$$

where ϕ is given by (A 8) and t is the root of (A 7) which gives the correct behaviour of ϕ at each point (R^*, Z^*) as described. We must compare this solution to the numerical solution in region II to determine whether they match in the appropriate limit.

A.3.1. Matching between regions II and IIIa

As we match to region II, the region IIIa variables become large and positive. The correct solution of (A 7) in this case is given by

$$t = \frac{1}{4R^*\bar{q}}\mathcal{F} + \frac{1}{4R^*\bar{q}}(R^{*2} + 4\bar{q}^2Z^{*2})\frac{1}{\mathcal{F}} + \frac{1}{2}\frac{Z^* + R^*}{R^*},$$

where \mathcal{F} is given by

$$\mathcal{F} = (2\bar{q}(4\bar{q}^2 + 3)R^{*3} + 6\bar{q}Z^*R^{*2} + 8\bar{q}^3Z^{*3} + R^*[(64\bar{q}^6 + 96\bar{q}^4 + 36\bar{q}^2 - 1)R^{*4} + (96\bar{q}^4 + 72\bar{q}^2)R^{*3}Z^* + 24\bar{q}^2R^{*2}Z^{*2} + (128\bar{q}^6 + 96\bar{q}^4)R^*Z^{*3} + 48\bar{q}^4Z^{*4}]^{1/2})^{1/3}.$$

In terms of the region II variables we find that to leading order

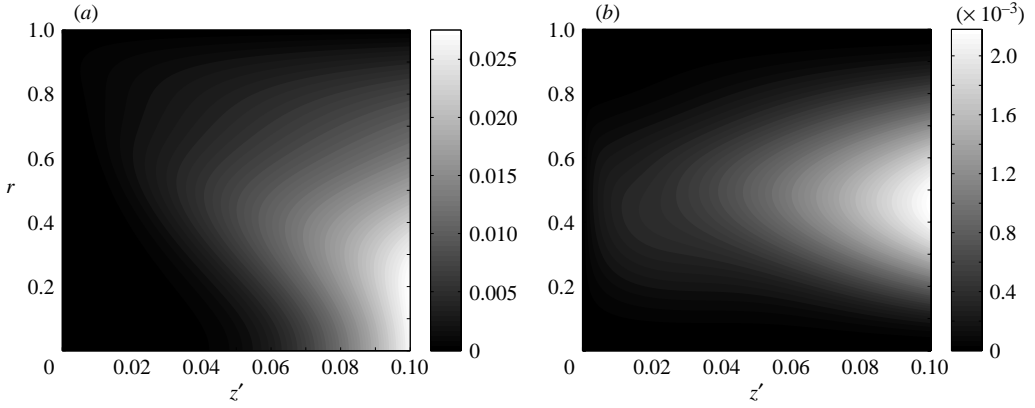


FIGURE 18. Comparison between solutions in regions II and IIIa. (a) f_T , (b) f_ψ .

$$t \sim \frac{3}{2} Da^2 \frac{z'}{(1-r^2)}.$$

Substituting this into the expression for ϕ and taking the leading-order terms we get

$$\phi \sim Da^{-1} \frac{(1-r^2)^3}{18z'}.$$

The region IIIa solutions in terms of r and z' are therefore

$$T = 1 + Da^{-\gamma} \exp\left(-\frac{(1-r^2)^3}{18z'}\right), \tag{A 9a}$$

$$\psi = -\left(r^2 - \frac{r^4}{2}\right) + Da^{-\gamma-2} \exp\left(-\frac{(1-r^2)^3}{18z'}\right), \tag{A 9b}$$

to leading order. We expect that the small correction to the solution in region IIIa will give an $O(1)$ contribution in region II to match with the numerical solution obtained here. The multiplier in front of the exponential term could be found from the next-order solution for ϕ , and the value of γ chosen to make this term $O(1)$ in region II. However, our solution for T satisfies

$$\frac{\partial T}{\partial z'} = \frac{(1-r^2)^3}{18z'^2} (T-1),$$

which gives the leading-order behaviour for T , at small z' , without calculating the next-order correction for ϕ . Similarly, the small- z' behaviour of the streamfunction, ψ , is given by

$$\frac{\partial \psi}{\partial z'} = \frac{(1-r^2)^3}{18z'^2} \left(\psi + \left(r^2 - \frac{r^4}{2}\right) \right).$$

Substituting the region II solution into the expressions

$$f_T = z'^2 \frac{\partial T}{\partial z'} - \frac{(1-r^2)^3}{18} (T-1),$$

$$f_\psi = z'^2 \frac{\partial \psi}{\partial z'} - \frac{(1-r^2)^3}{18} \left(\psi + \left(r^2 - \frac{r^4}{2}\right) \right),$$

we expect to find that f_T and $f_\psi \rightarrow 0$ as $z' \rightarrow 0$. Figure 18 shows these functions for small z' , confirming that the solutions in regions II and IIIa match in the limit $z' \rightarrow 0$.

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