Heat transfer mechanisms of laminar flames of hydrogen + oxygen


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Abstract

Increasing the heat transfer from premixed laminar oxy-fuel flames to glass or quartz products is of major importance in the lighting industry. In this paper a laminar flame of hydrogen + oxygen is used as an impinging jet in a stagnation-flow-like configuration to investigate the heating of a glass product. The research was intended to analyze the crucial phenomena determining the heat transfer rate. The time scales of the processes taking place in the flame, the stagnation boundary layer, and the plate are quantified and from this it is shown that these zones can be decoupled. It will also be shown that, as a result, the heat flux entering the plate depends only on the stagnation flow and the plate’s surface temperature. Two cases were studied. In one case the stagnation boundary layer consisting of a burnt and chemically frozen mixture of hydrogen + oxygen or hydrogen + air is studied. In the other case the flow is reactive in the stagnation boundary layer. An analytical approximation for the heat transfer coefficient is derived for the case without a viscous sublayer. The effect of strain rate on the heat transfer coefficients is incorporated in this model. This heat transfer coefficient is compared to numerically calculated heat transfer coefficients for stagnation flows with both reactive and nonreactive boundary layers. Furthermore, it is shown that the stagnation boundary layer is not in chemical equilibrium. First numerical results indicate that surface chemistry can be expected to contribute significantly to the heating process. Surface chemistry is studied numerically by assuming a quartz plate coated with a platinum layer.

Keywords: Hydrogen; Oxy-fuel; Heat transfer; Surface chemistry; Stagnation flame

1. Introduction

Mixtures of a fuel and oxygen are used for many industrial processes. Oxy-fuel flames are used for welding and cutting purposes and for coating processes [1]. High-velocity oxy-fuel (HVOF) spray-
chemical energy into thermal energy rapidly. The fast conversion results in high flame speeds and heat transfer rates. Moreover, the lack of nitrogen in the premixed gas may reduce the yield of nitrogen oxide (NO$_x$) considerably. For melting glass in industrial ovens [4], hydrogen–oxygen mixtures are commonly used [5]. The reduction in energy consumption is considerable when recuperative ovens are replaced by oxy-fuel ovens [5]. Since the late 1960s and early 1970s the lighting industry has been using premixed oxy-fuel combustion. High heat transfer rates leading to short processing times were the main reasons for introducing oxy-fuel combustion. Short processing times are needed both for a high product throughput and for quality. In the lamp-making process the heat has to be applied very locally. Therefore, oxy-fuel flames in an impinging-jet-like configuration are often used. Impinging laminar and turbulent jets have been well studied. Especially the heat transfer from inert gas jets to stagnation walls is well known [6–8]. Oxy-fuel flames are small and mostly laminar. Little is known about impinging laminar jets consisting of premixed oxy-fuel flames.

For the lighting industry, to be able to reduce the costs and to enhance efficiency, more knowledge on the mechanisms of heat transfer from such oxy-fuel flame jets to the glass products has to be gained. The purpose of this study is to gain knowledge on the heat transfer mechanisms from small, high-speed, laminar, oxy-fuel flames to glass products for lamp manufacturing. In this study a hydrogen–oxygen flame was used as an impinging jet in a stagnation-flow-like configuration. The goal of this study was to analyze the crucial heat transfer phenomena to formulate new design rules for the optimization of the heat transfer.

It is anticipated not only that the transport of heat by convection and conduction affects the heat transfer in oxy-fuel flames but also that the production of heat by chemistry effects has a significant influence. These very hot flames contain considerable quantities of free radicals such as O, OH, and H, which might recombine in the cold stagnant boundary layer near the
product, releasing chemical energy and boosting the heat transfer ($\approx 10^2$–$10^3$ J/g mixture). Also recombination reactions of species adsorbed on the product surface might influence the heat transfer rate. The importance of these different mechanisms is investigated here.

It will be assumed in the current study that the burner is close to the product surface so that the distance from the flame to the product is small compared to the typical width of the jet of combusted gases. The configuration in this case can be considered a one-dimensional reacting stagnation flow issuing on a flat plate; see Fig. 1. The heat transfer phenomena within this configuration are assessed analytically and numerically in this paper. Experiments are in progress.

The focus of this study is on high-temperature hydrogen–oxygen flames. Mixtures of hydrogen and air are discussed also so that a comparison can be made between the observed phenomena in oxy-fuel and air-fuel flames. In this study only the thermodynamical properties of quartz will be taken into account since quartz is the main component in lamp glasses and needs the highest heat transfer rate to melt.

In the next section the computation geometry will be presented, together with the conservation equations and boundary conditions for the reacting stagnant flow, for the quartz plate and the inclusion of surface chemistry effects. In Section 3, a time scale analysis will be performed for the typical time scales of (1) the heat production in the reaction layer of the flame and (2) the heat transport by convection. Section 4 gives the typical time scale for the heating of a quartz glass product. Section 5 discusses the heat transfer from a burnt gas to the product when no chemical reactions take place in the stagnant boundary layer. Afterward in Section 6, the effect of recombination reactions in the boundary layer on the heat transfer rate to the product is determined. In Section 7 it will be shown that the reacting stagnation flow is not in chemical equilibrium and that surface chemistry may play a role. The influence of surface chemistry on the heat transfer rate is discussed qualitatively in Section 8.

2. Governing equations

The heat transfer of a hot laminar stagnation flow to an initially relatively cold plate is studied analytically and numerically. For the case of very small distances to the solid surface, the situation can be regarded effectively as one-dimensional. Fig. 1 presents the one-dimensional (1D) computational setup. By fixing the gas composition, temperature, applied strain, and pressure at the nozzle outlet or similarly the left inflow boundary at $x = L = -10^{-2}$ m, a thin laminar premixed flame stabilizes at a position $x_{\text{stab}}$ close to the surface of the plate ($x = 0$). The Reynolds number based on the nozzle diameter and the mean velocity, viscosity, and density of the gas in the nozzle is of the order of $10^2$–$10^3$. The Reynolds number in the flame front based on the typical stream tube width, laminar burning velocity, and viscosity and density in the flame front is of the order of $10^2$. The plate thickness is $D = 5 \times 10^{-3}$ m and at time $t = 0$ the whole plate is set to a uniform temperature of 700 K. Due to the heat transfer from the burnt mixture to the plate, the plate temperature raises in time. Fig. 2 shows the temperature profile in the gas and plate for a hydrogen–oxygen mixture at some time instants. For the present case the flame is stabilized at a distance of around $x_{\text{stab}} = -7 \times 10^{-3}$ m. The temperature raises around $x = x_{\text{stab}}$ to a value close to the adiabatic flame temperature. Then the temperature stays constant until it drops in the thermal boundary layer near the plate, where the flow stagnates to the plate.

In this section the equations describing this 1D chemically reacting flow are formulated. The energy conservation equation for the plate will then be discussed. The boundary conditions and the equations coupling the gas to the solid phase will also be given. Finally, how the boundary conditions have to be modified to take heterogeneous surface reactions into account will be discussed. The conservation equations for the reacting gas mixture include conservation of mass, momentum, and enthalpy in a one-dimensional configuration. Fig. 1 shows this configuration. The density $\rho$, species mass fractions $Y_i$, temperature $T$, and enthalpy $j$ depend only on the leading coordinate $x$. The $y$-velocity component $v$ and the pressure, however, depend on $x$ and $y$. On the centerline ($y = 0$) $v = 0$ but $\partial v/\partial y \neq 0$. To take this derivative...
The conservation of mass is expressed by the continuity equation [14],

\[ \rho \frac{\partial u}{\partial t} + \rho u \frac{\partial u}{\partial x} = \rho K, \]

where \( u \) is the local velocity component in \( x \) direction. The term \( \rho K \) describes the loss of mass in perpendicular direction (\( y \)) due to the stagnating flow. In the one-dimensional stagnation flame the \( y \)-momentum equation reduces to an effective equation for the strain rate (see, e.g., [13] and [15]),

\[ \frac{\partial K}{\partial t} + \rho u \frac{\partial K}{\partial x} = \frac{1}{\gamma G + 1} \left( \rho a^2 - \rho K^2 \right), \]

where \( \gamma_G = 0 \) for a planar and \( \gamma_G = 1 \) for an axisymmetric flow. \( K \) equals the strain rate at the inflow boundary (\( x = L \)) and \( \mu \) is the viscosity. In a chemically reacting flow, a conservation equation for every species \( i \) has to be solved,

\[ \frac{\partial \rho Y_i}{\partial t} + \rho u \frac{\partial Y_i}{\partial x} = \frac{\partial}{\partial x} \left( \rho D_{im} \frac{\partial Y_i}{\partial x} \right) - \dot{r}_i = -\rho KY_i, \]

where \( D_{im} \) is the effective diffusion coefficient, which is calculated using a mixture averaged diffusion model [16,17]. Furthermore, the chemical source term is quantified by \( \dot{r}_i = M_i \sum_k v_{ik} \omega_k \), where \( M_i \) is the molar mass of species \( i \), \( v_{ik} \) is the stoichiometric coefficient of species \( i \) in reaction \( k \), and \( \omega_k \) is the reaction rate of reaction \( k \). The energy conservation equation for the gas is written with regard to the specific enthalpy \( j \) and temperature \( T \):

\[ \frac{\partial \rho j}{\partial t} + \frac{\partial \rho u j}{\partial x} - \frac{\partial}{\partial x} \left( \rho \frac{\partial T}{\partial x} \right) - \frac{\partial}{\partial x} \left( \sum_{i=1}^{N} j_i \rho D_{im} \frac{\partial Y_i}{\partial x} \right) = -\rho K j. \]

The specific enthalpy of the mixture is a mass-weighted sum of the specific enthalpies of all species,

\[ j_i = j_i^0 + \int_{T_0}^{T} c_{p,i}(T^*) dT^*, \]

where \( j_i^0 \) is the chemical formation enthalpy of species \( i \) at reference temperature \( T_0 \), and \( c_{p,i} \) is the specific heat capacity of species \( i \). The set of equations for the gas is closed by the ideal gas law,

\[ \rho = \frac{p \tilde{M}}{RT}, \]

where \( R \) is the universal gas constant and \( \tilde{M} \) is the average molar mass of the mixture. For low Mach number flows, the pressure in this last expression can be taken constant, \( p = p_a \), where \( p_a \) is the atmospheric pressure at the inlet.

For the heating of the plate (\( x > 0 \)) the corresponding energy equation has to be solved,

\[ \rho p c_{pp} \frac{\partial T}{\partial t} = \frac{\partial}{\partial x} \left( \lambda_p \frac{\partial T}{\partial x} \right), \]

where \( \rho_p \), \( c_{pp} \), and \( \lambda_p \) are the density, specific heat capacity, and thermal conductivity coefficients of the plate.

Different transport mechanisms of heat can be considered. One of the mechanisms is the radiative heat flux from the gas to the plate. The heat flux has...
a maximum of the order of $10^2$–$10^3$ W/m$^2$ K (calculated with values from [18–20]) where the absorption bands of H$_2$O are taken into account. The other mechanism is the radiative heat flux from the glass plate to the surroundings. The heat flux has a maximum of the order of $10^4$–$10^5$ W/m$^2$ K (calculated with the black box model above 5 µm). Therefore, the radiative heat flux from the gases to the plate is negligible, while the radiative heat flux from the product to the surroundings comes into play only at high temperatures. On the other hand, radiative transport may cause redistribution of heat within the plate. Later in this paper, we will show that the flow heat flux from the gasses to the product depends only on the gas-plate interface temperature and not on the temperature distribution within the plate. Therefore, radiation is not taken into account.

Boundary conditions for the gas phase have to be defined at the inlet at $x = L$ and at the plate surface at $x = 0$. At the inlet Dirichlet boundary conditions are applied: $u = u_0$, $K = a$, $Y_i = Y_{i,u}$, and $T = T_0$. At $x = 0$ both components of the velocity are zero for all $y$ so that $u = 0$ and $K = 0$. The temperature follows from continuity of heat fluxes from the gas to the plate:

$$\lambda_\text{g} \frac{\partial T}{\partial x} \bigg|_{x=0^-} = \lambda_\text{p} \frac{\partial T}{\partial x} \bigg|_{x=0^+}. \quad (9)$$

This boundary condition holds as long as the plate surface is inert. At $x = D$ the plate is assumed to be adiabatic for simplicity so that $\partial T/\partial x = 0$.

When the surface is catalytically inactive no net mass transport between the gas and the solid phase exists for each species: $\partial Y_i/\partial x = 0$ at $x = 0$. However, when surface reactions take place at $x = 0$, the species mass fluxes are no longer zero ($\partial Y_i/\partial x \big|_{x=0} \neq 0$). The molar flux of species $i$ from the gas toward the plate is defined as $\dot{s}_i$. $\dot{s}_i$ is the adsorption rate minus the desorption rate of species $i$. If the system is not in steady state then a net mass transport between the gas and the solid phase may exist. A Stefan velocity $u_\text{s}$ normal to the surface can be defined according to the mass conservation equation:

$$\rho u_\text{s} = \sum_{i=1}^{N} \dot{s}_i M_i. \quad (10)$$

If the heterogeneous kinetic system at the surface is in steady state the Stefan velocity equals zero. The surface flux depends on the surface impingement rates, the sticking coefficients, the surface reaction constants, the gas composition, and the surface coverages.

For the heterogeneous chemical kinetic model the model of Hellings et al. [21] is used. This model is developed for a platinum surface at low pressures and moderate temperatures. A model for quartz does not exist, but it is expected that the platinum model gives an indication of the importance of surface chemistry. For the homogeneous gas phase chemistry the reduced mechanism of Smooke [11] is used. This mechanism is developed for methane-air combustion with nitrogen acting in third-body reactions. The mechanism is valid here because the adiabatic burning velocity and flame temperature of the hydrogen–air and hydrogen–oxygen mixtures are comparable to those calculated with other mechanisms such as Marinov et al.’s mechanism [9] which was developed for hydrogen–air and hydrogen–oxygen combustion (see Table 1).

### 3. Time scale analysis of the stagnation flame

The behavior of the reacting flow and the most important time scales in the system are studied in this section. The heat transfer rate from the flame to the plate depends on the gas velocity of the impinging flow. The flow velocity $u$ on the other hand changes when the strain rate $K$ changes. By setting the strain $K = a$ at $x = L$ the effective inlet velocity $u_0$ is prescribed. The applied strain rate $a$ is chosen on the basis of the velocity at the inlet, the laminar burning velocity, the distance from the inlet at $x = L$ to the flame front at $x = x_\text{stab}$, and the distance from the flame front to the plate surface at $x = 0$ (see Fig. 1). In Fig. 2 the behavior of $K$ and $u$ is considered. Roughly four regions can be distinguished. Close to the inlet (region I) the mixture is unburnt, so that the strain rate $K = a$ is constant and the velocity decreases linearly from the boundary value to a value close to the burning velocity near the position where the flame front stabilizes in the flow. When the gas enters the combustion zone (region II) the strain rate is not constant anymore due to the density gradient in the flame. The resulting $K$ can be derived from Eq. (3). Once the gas is burnt it approaches chemical equilibrium (region III). If the flame is stabilized relatively far from the stagnation plane compared to the flame thickness, $K$ does not change in time and

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space anymore and the asymptotic value for $K$ can be derived again from Eq. (3). Assuming that $K$ approaches a constant value, it follows that the strain rate of the mixture in the burnt gases is given by

$$K_m = a \left( \frac{\rho_u}{\rho_m} \right)^{1/2},$$

(11)

where $\rho_m$ is the density of the burnt gases, $\rho_m$ is equal to the density of an adiabatic equilibrium mixture in the case of a fast chemistry (hydrogen/oxygen), but this value is not always reached if the chemical processes are slower (hydrogen/air).

Rogg and Peters [15] have shown that $K$ changes in the form of a cosine in the burnt gases approaching the stagnation plane in the case of an opposed twin flame geometry. This is also the case in the present geometry (region III), except for the region almost at the surface of the plate, where the viscous boundary layer affects the velocity profile. In the stagnation boundary layer (region IV) the strain rate decreases again. At the stagnation plane the strain rate equals zero because at $x = 0$ the velocity component parallel to the plate $v$ is zero for all $y$. Eq. (11) indicates that the strain rate which determines the heat transfer to the plate is dependent on the applied strain rate $a$ and on the density of the burnt gases $\rho_m$, which in turn depends on whether the burnt gases reach chemical equilibrium. The strain rates for hydrogen–oxygen mixtures are higher than those for hydrogen–air mixtures because the typical burning velocities for hydrogen–oxygen mixtures are much greater. The hydrogen–oxygen mixtures are studied with applied strain rates of 6000, 8000, and 10,000 s$^{-1}$. The hydrogen–air mixtures have applied strain rates of 2000, 4000, and 6000 s$^{-1}$.

In the remainder of this section, a time scale analysis for the different heating processes in the flame front (region II) and in the stagnation boundary layer (region IV) will be presented to judge the importance of the different processes [22]. To do this the much simpler situation of an inert flow stagnating on a plate is considered.

The chemical time scale of a flame front is typically $\tau_f = \delta_f/\dot{\lambda}_g$, where $\delta_f$ is the flame front thickness and $\dot{\lambda}_g$ is the burning velocity. The typical time scale for the heating of the inert boundary layer $\tau_b$ is determined by convective heating. In this case, the time scale is entirely determined from the strain rate in the flow: $\tau_b = 1/K_m$. The strain rate of a hydrogen–oxygen mixture is of the order $10^4$ s$^{-1}$; so $\tau_b \sim 10^{-4}$ s.

It is assumed that the typical flame front thickness is much thinner than the distance from the plate to the flame front: $\delta_f \ll |x_f|$. For this situation, it can be shown that the chemical time scale of the flame is much smaller than the time scale of the boundary layer. Therefore it is assumed that in an inviscid stagnation flow the velocity decrease is approximately linear ($u \approx -K_mx$). Therefore $K_m \approx -um/|x_f|$, where $um$ is the gas velocity in the burnt gases of the flame, and thus $\tau_b = 1/K_m \approx \lambda_f/um = -(|x_f|/\delta_L)(\rho_m/\rho_u)$, which is larger than $\tau_f = \delta_f/\dot{\lambda}_g$. The properties of the flame are assumed to be constant and thus the thermal diffusivity $\alpha_g = \lambda_g/\rho_g cpg$ is constant. An estimate for the flame thickness is $\delta_f = \alpha_g/\dot{\lambda}_g$ so that the typical chemical reaction time scale is $\tau_f = \alpha_g/\dot{\lambda}_g$. In a hydrogen–oxygen flame at 3079 K, $\alpha_g = 2.72 \times 10^{-3}$ m$^2$/s. With $\delta_L = 9.97$ m/s, $\tau_f \sim 10^{-5}$ s. As expected $\tau_f \ll \tau_b$.

4. Time scale analysis for the quartz glass product

Another limiting time scale is the typical heat-up time scale of the plate. To determine this time scale the heat balance between the quasi-steady stagnation flow and the time-dependent behavior of the plate is studied. The enthalpy equation for the stagnation flow (Eq. (5)) reduces to an equation in which only thermal convection and diffusion are taken into account. Viscosity of the stagnation flow is neglected again. If furthermore it is assumed that the specific heat capacity, thermal conductivity, and density are independent of temperature, the one-dimensional steady energy equation for the stagnation flow becomes

$$-\kappa x \frac{\partial T}{\partial x} = \frac{\partial^2 T}{\partial x^2} \quad (x < 0)$$

(12)

with $\kappa = \rho_g c_p g K_m/\dot{\lambda}_g$. It can be derived from Eq. (12) by integrating twice and inserting boundary condition (Eq. (9)) that the temperature gradient in the plate at the interface is equal to

$$\frac{\partial T}{\partial x} \bigg|_{x=0^+} = (T(0) - T_m) \left( \frac{2\kappa}{\pi} \right)^{1/2} \left( \frac{\dot{\lambda}_g}{\lambda_p} \right),$$

(13)

where $T_m$ is the temperature at $x = -\infty$, which is the temperature of the burnt mixture. Eq. (13) is used as boundary condition for the energy equation of the plate. For the heat-up time scale of the plate, the one-dimensional time-dependent energy Eq. (8) is considered. If the thermal conductivity coefficient $\lambda_p$, the specific heat capacity $c_p$, and the density $\rho_p$ of the plate are chosen to be temperature independent then the thermal diffusivity is defined as $\alpha_p = \lambda_p/\rho c_p$ and the energy equation inside the plate becomes

$$\frac{1}{\alpha_p} \frac{\partial T}{\partial t} = \frac{\partial^2 T}{\partial x^2} \quad (x > 0).$$

(14)
This differential equation is solved using separation of variables, which results in a solution of the form

\[ T(x, t) \sim (C_1 \cos(kx) + C_2 \sin(kx)) \exp(-k^2 \alpha_p t), \]

where \( C_1, C_2, \) and \( k \) are constants which have to be determined from the boundary and initial conditions. From the exponent in Eq. (15) it follows that the typical heat-up time scale of the plate is \( \tau_p = 1/k^2 \alpha_p. \)

The values of \( k \) are found with the boundary condition (Eq. (13)) at \( x = 0^+ \), the adiabatic boundary condition at \( x = D \), and the initial condition which supposes that the plate is initially at a uniform temperature \( T_0 \). Then one can show that \( k \) can be derived from

\[ k D \tan(kD) = \xi D \]

with \( \xi = \lambda_g/\rho_p(2k/\pi)^{1/2} \). There is an infinite number of possible \( k \) values and the solution (Eq. (15)) is a linear combination of all contributions. Small values of \( k \) indicate the slowest time scales, while large values of \( k \) give the fast scales during the initial phase of the heating process. From Eq. (15) it follows that the fast time scales are rapidly exhausted and the smallest \( k \) value is a measure for the time scale of the heating process of the complete plate. The smallest value of \( k \) is such that \( 0 < kD < \pi/2 \).

For the glass plate the thermal conductivity coefficient is \( \lambda_g = 1.4 \text{ W/mK} \), the specific heat capacity \( c_{pp} = 780 \text{ J/kg K} \), and the density \( \rho_g = 2.25 \times 10^3 \text{ kg/m}^3 \). Therefore, \( \alpha_g = 8.00 \times 10^{-7} \text{ m}^2/\text{s} \). Typical plate thicknesses are of the order \( 10^{-3} \text{ m} \) or larger. A typical maximum strain rate for a hydrogen–oxygen flame is \( 1.8 \times 10^4 \text{ s}^{-1} \).

For a gas temperature at the plate of 1500 K, the thermal conductivity coefficient is \( \lambda_g = 0.23 \text{ W/mK} \), the specific heat capacity \( c_{pg} = 2752 \text{ J/kg K} \), the density \( \rho_g = 0.131 \text{ kg/m}^3 \), and thus \( \xi = 696 \text{ m}^{-1} \). When the plate’s thickness is \( D = 5 \text{ mm} \), \( k = 246 \text{ m}^{-1} \) and \( \tau_p = 21 \text{ s} \). When the plate’s thickness is \( D = 1 \text{ mm} \), \( k = 749 \text{ m}^{-1} \) and \( \tau_p = 2.2 \text{ s} \). Thus, for plate thicknesses of the order of millimeters \( \tau_p \) is of the order of seconds. We may conclude from this exercise that the heat-up time scale of the plate is much larger than the typical time scale of the boundary layer and the flame \( (\tau_p \gg \tau_b \gg \tau_f) \). Because these time scales differ significantly, the heating processes of the plate and gas can be decoupled if the plate is more than a millimeter thick. For each value of the surface temperature \( T(0) \) the solution of the gas equations adapts very quickly and is very fast in quasi-steady state. The heat flux from the gas to the plate is therefore independent of the temperature profile in the plate but depends only on the temperature profile of the boundary layer and thus only in a quasi-steady manner on the surface temperature.

\[ q = q(T(0)) \]

So the heat flux to the plate \( q \) can be determined by computing the gas temperature profile, for each value of the interface temperature \( T(0) \) without considering the heating of the solid phase. Stimulated by Eq. (13) we now write \( q = h(T(0) - T_m) \), where \( T_m \) is the maximum temperature of the burnt gases. The focus will be on the influence of the different physical processes on the value of the effective heat transfer coefficient \( h = h(T(0)) \).

One reference solution \( h_{ref}(T(0)) \) for an inert gas burning stagnating on an inert surface (neglecting viscosity so that the Prandtl number \( Pr = 0 \)) follows from Eq. (13).

\[ h_{ref} = \frac{\lambda_g}{T} \left( \frac{2k}{\pi} \right)^{1/2}, \]

which is a function of \( T(0) \) (because the transport coefficients depend on \( T(0) \)) and of the strain rate \( K_m \).

Fig. 3 shows the heat flux \( q \) from a reactive mixture to a plate as a function of the plate surface temperature in the case of three plate thicknesses \( D \). As expected, the lines are almost indistinguishable, although there is a small deviation of the heat flux for the very thin plate. To conclude we may say that this analysis shows that the surface temperature determines the heat flux completely, independent of the plate thickness. However, when the plate becomes very thin, the time scale of the heating of the plate and the time scale of the boundary layer become equally large and the energy distribution in the plate has to be taken into account.

5. Heat transfer of a nonreactive stagnation flow

The influence of viscosity inside the boundary layer close to the plate is studied numerically in this
These effects have been neglected in the analytical evaluation of $h_{\text{ref}}$ so far. The composition of the burnt gases of the flames is frozen manually once they have reached their maximum temperature. In this way, a chemical inert boundary layer of a 1D stagnant flow on a flat plate is created. Because no chemical reactions occur in the boundary layer at the plate, heat transfer from the mixture to the plate has a solely thermodynamic character. The thermodynamic variables such as the specific heat capacity and the thermal conductivity coefficient change within the boundary layer because of the temperature dependency. Also the density is not constant but is derived from the ideal gas law.

In Fig. 4 the heat flux $q$ is plotted as function of the surface temperature $T(0)$ for a hydrogen–oxygen and a hydrogen–air mixture with an applied strain rate of 6000 s$^{-1}$. It shows that the heat input for a frozen hydrogen–oxygen mixture is much higher than that for a frozen hydrogen–air mixture. The heat input into the plate is in both cases almost linear with plate temperature. For practical use, it is common to use an effective heat transfer coefficient $h = q/(T_m - T(0))$. Table 2 shows the maximum flame temperatures $T_m$ for hydrogen–oxygen and hydrogen–air mixtures for different applied strain rates. These maximum temperatures are compared with the adiabatic temperatures. Note that the temperature of the burnt hydrogen–oxygen mixture is almost equal to the adiabatic flame temperature 3079 K for all studied strain rates.

Fig. 5 shows $h/h_{\text{ref}}$ as functions of the surface temperature for the hydrogen–oxygen mixtures. For the strain rate in $h_{\text{ref}}$ the maximum value $K_m$ of Eq. (3) is used, which is equal to 16,986 s$^{-1}$ for a hydrogen–oxygen mixture. From Fig. 5 it can be concluded that the heat transfer coefficient for the frozen hydrogen–oxygen mixture is almost proportional to the analytical coefficient $h_{\text{ref}}$ for all surface temperatures, at approximately $h \approx 0.75h_{\text{ref}}$. The fact that $h \neq h_{\text{ref}}$ is due to viscous effects near the plate. Viscous effects are not taken into account in $h_{\text{ref}}$. For zero viscosity, it appeared numerically that $h \approx h_{\text{ref}}$, which validates the analytical approximation, Eq. (17).

The burnt mixtures of the hydrogen–oxygen are approximately in chemical equilibrium while the burnt hydrogen–air mixture is not. The adiabatic temperature of a stoichiometric hydrogen–air flame is approximately 2390 K. However, the burnt mixtures reach a lower temperature for all strain rates investigated here (see Table 2). It can be concluded that for these strain rates the burnt hydrogen–air mixtures have not reached chemical equilibrium. Fig. 6 shows $h/h_{\text{ref}}$ for the frozen hydrogen–air mixture as function of the surface temperature. The observations for the hydrogen–oxygen mixture also hold for the hydrogen–air mixture. Note that $h \approx 0.6h_{\text{ref}}$ in this case.
Fig. 5 shows the heat transfer coefficients for a frozen mixture with applied strain rate 6000 s\(^{-1}\) is also presented (lower dashed-dotted line).

**6. Heat transfer of a reactive stagnation flow**

In the previous section the heat transfer from a chemically frozen mixture to a flat plate was discussed. Now we will discuss the importance of chemistry in the boundary layer and its effect on the heat transfer. The heat transfer of hydrogen–oxygen and hydrogen–air flames is considered for different strain rates. First, the heat transfers from burnt but still chemically reactive hydrogen–oxygen and hydrogen–air mixtures to a plate are studied. Both mixtures have an applied strain rate of 6000 s\(^{-1}\). In Fig. 4 the heat transfer rates are plotted together with the \(q\) plots for inert boundary layers. It is clear that the heat transfer from the reactive mixture to the body is much higher than that for the frozen mixture. This indicates that extra heat is generated by chemical reactions in the boundary layer, which can be explained as follows. When heat is extracted from the mixture in the boundary layer, the gas temperature drops. The chemical equilibrium shifts and chemistry continues to develop. As a result, more heat is generated by extra combination reactions of radical species such as O, H, and OH, which is not the case when the mixture is frozen. This effect is very large in the burnt hydrogen–oxygen mixture as the amount of free radicals is high.

For different applied strain rates the relative heat transfer coefficients are computed, as in Section 5. Fig. 5 shows the heat transfer coefficients \(h\) of hydrogen–oxygen flames to the body divided by the reference heat transfer coefficient \(h_{\text{ref}}\) for applied strain rates of 6000, 8000, and 10,000 s\(^{-1}\). The corresponding strain rates of the burnt mixture \(K_m\) which are used to determine \(h_{\text{ref}}\) are given in Table 3. In Table 3 also the burnt strain rates for the frozen mixtures are given. For an applied strain rate of \(a = 6000\) s\(^{-1}\) the differences of \(K_m\) between the frozen mixture and the reactive mixtures are relatively small. For the thermodynamic variables and density in \(h_{\text{ref}}\) the same temperature-dependent relations as for the frozen mixture are used. Fig. 5 shows that the relative heat transfer coefficients \(h/h_{\text{ref}}\) for the reactive mixtures are more or less independent of \(K_m\). Therefore, it is concluded that in general the heat transfer coefficient \(h\) has a \(K_m^{1/2}\) dependency like that of \(h_{\text{ref}}\) as predicted by Eq. (17). It can also be observed from Fig. 5 that for an applied strain rate of 6000 s\(^{-1}\), \(h/h_{\text{ref}}\) for the reactive mixture is approximately twice as high as that for the frozen mixture, which corresponds to Fig. 4.

Fig. 6 shows \(h/h_{\text{ref}}\) for burnt but still reactive hydrogen–air mixtures with applied strain rates of 2000, 4000, and 6000 s\(^{-1}\). The influence of chemical reactions in the stagnation boundary layer is much smaller. This is because the radical pool in the hydrogen–air flames is much smaller. It is also seen that the \(K_m^{1/2}\) dependence of \(h\) is not as strict as that for the oxy-fuel flames. At high strain rates the burnt gases are not in equilibrium yet and are affected by the stagnation flow. The chemical and flow time scales are of comparable magnitude and therefore the mixture is not able to convert all its chemical energy into heat. For a higher strain rate the flow time scale \(\tau_b = 1/K_m\) decreases, the maximum flame temperature drops, and \(h/h_{\text{ref}}\) decreases.

**7. Chemical equilibrium**

So far mixtures with reactive and frozen stagnation boundary layers have been studied. When the boundary layer is still reactive, recombination reactions occur and extra heat is produced. If the recombination reactions in the boundary layer are so fast that local chemical equilibrium is reached, the maximum amount of heat transfer is obtained. However, when the mixture in the boundary layer is not in chemical equilibrium some heat can be extracted by recombination reactions at the surface. Surface chemistry may assist to drive the system to local equilibrium. In
this section the chemical composition in the reactive boundary layer will be studied and the results will be compared with those of a boundary layer which has the same temperature but is in local chemical equilibrium.

A plate is heated with a hydrogen–oxygen flame with an applied strain rate of $6000 \text{ s}^{-1}$. The plate is initially at a uniform temperature of 700 K. When the plate surface has reached temperatures of approximately 1530 and 2800 K snapshots are made of the thermal (Fig. 7) and chemical boundary layers (Fig. 8). The temperature profiles are used subsequently to calculate the local chemical equilibrium compositions. Fig. 8 presents the mole fractions of molecular hydrogen, oxygen, and water for a boundary layer with surface temperatures of 1530 and 2800 K. The results are compared with the composition of a mixture with the same local temperature but in local chemical equilibrium. Fig. 8 (left) shows that if the mixture was in chemical equilibrium almost no molecular hydrogen and oxygen would exist close to the stagnation surface at a temperature of 1530 K. All hydrogen and oxygen would be converted to water. However, in the real case there is still a reasonable amount of molecular hydrogen and oxygen present. In the case of local equilibrium and for the higher surface temperature, more hydrogen and oxygen exist close to the stagnation surface. The real system is therefore closer to equilibrium at higher temperatures.

It can be concluded that the chemical boundary layer for a stagnating flame is still far from chemical equilibrium. There is still room for further conversion of chemical energy to heat by other means such as surface chemistry, which is discussed in the next section.

8. Surface chemistry

In this section we will focus on the effect of surface chemistry on the heating of a plate. The gases still contain molecular hydrogen and oxygen that can be converted to water. Due to surface chemistry conversion of hydrogen and oxygen to water, extra heat may be produced to heat the surface, but the importance of this effect is not known.

Catalytic combustion has been well studied, especially the ignition and combustion of pure hydrogen–oxygen mixtures over platinum surfaces at low pressures [21,23]. Also the interaction between homogeneous and heterogeneous chemistry for hydrogen mixtures in stagnation flows on platinum layers at elevated temperatures was studied numerically [24,25] and experimentally [26]. Transient ignition of premixed stagnation-point flows over a catalytic surface was investigated numerically [27]. Also work on the modeling of heterogeneous catalytic combustion in stagnation-point flows has been done recently [28].
Mokhov et al. [29] report from experimental investigations and numerical simulations the behavior of CO and OH in the laminar boundary layer of a combusted propane–air stagnation flow. Hereby the plate temperature, equivalence ratio of the oncoming flow, and catalytic properties of the surface are varied.

Because the main interest is the contribution of surface chemistry to the heat transfer and a kinetic mechanism for quartz does not exist, it is assumed that the surface acts in a way similar to that of platinum. By using platinum as a catalyst the maximum contribution to the heat flux can be determined. For the chemical kinetic model, the model of Hellsing et al. [21] is used. The model of Hellsing et al. [21] has been validated extensively for low pressures and moderate temperatures. Although we do not have a low overall pressure, the partial pressures of hydrogen and oxygen are comparable to the partial pressures for hydrogen and oxygen in Hellsing et al. [21]. The model was extrapolated to high temperatures. Conversion of a reactant to a product by surface reactions has different steps. These steps are absorption, with or without dissociation, diffusion, surface reactions, and desorption. The typical time scale for surface chemistry corresponds to the time scale of the slowest step. If the surface has to remain catalytically active, the slowest step has to be the adsorption step; otherwise the surface will be fully covered and no further significant surface reactions will take place. Therefore let us consider the absorption rate of a reactant in the gas phase. The absorption rate of species $i$ is $r_i = S_i(\theta, T) F_i(p_i, m_i, T)$, where $F_i$ is the impingement rate of species $i$ and $S_i$ is the sticking coefficient [21]. The sticking coefficient may be dependent on the surface coverage $\theta$ and temperature $T$. The impingement rate is the rate of molecules that hit a surface site [30]:

$$F_i = \frac{p_i A_{\text{site}}}{(2\pi m_i k_B T)^{1/2}}. \quad (18)$$

The absorption rate is the number of molecules that is absorbed by a single surface site per unit time. Thus the typical time scale of absorption is $\tau_{si} = 1/r_i$. For hydrogen and oxygen absorption on a platinum surface with the conditions discussed previously the time scales are of the order $10^{-7} – 10^{-4}$ s. In general the absorption of oxygen takes longer than the absorption of hydrogen here, due to a lower partial pressure and sticking coefficient and a higher specific mass of oxygen. Since the absorption time scales are much faster than the typical time scales discussed in Section 3, it can be assumed safely that the surface chemistry follows the gas phase processes quasi-steadily.

A stoichiometric hydrogen–oxygen flame with applied strain rate of 6000 s$^{-1}$ is studied. Fig. 9 (left) shows the species composition of molecular hydrogen and oxygen in the stagnation boundary layer of a hydrogen–oxygen flame, where in one case (solid line) no surface chemistry is taken into account and in the other case (dashed line) surface chemistry is included. The surface temperature is 2800 K. Near the surface the concentrations of hydrogen and oxygen are somewhat lower for the situation where surface chemistry is included than for the situation where no surface chemistry is included. From this we may conclude that more hydrogen and oxygen are converted to water with a reactive surface and extra heat is produced.

Fig. 9 (right) shows the temperature profiles in the stagnation boundary layer for a surface temperature of 2800 K. When no surface chemistry is taken into account, the temperature profile shown is reached after 10 s in the case of a quartz plate of 5 mm thickness. When surface chemistry is taken into account the pro-

![Fig. 9. Species concentration profiles of H$_2$ and O$_2$ (left) and temperature profile (right) of the stagnation boundary layer for a reactive burnt hydrogen–oxygen mixture with (dashed line) and without (solid line) surface chemistry for a surface temperature of 2800 K.](image-url)
file shown is reached after 8.1 s. This means that surface chemistry reduces the heat-up time considerably despite the small differences in species profiles.

Figs. 4 and 5 present the heat flux and heat transfer coefficients as functions of surface temperature for both cases with and without surface chemistry. For high surface temperatures the heat transfer coefficient for a catalytically active surface is higher than that for a noncatalytically active surface.

With the chemical kinetic model studied here, it can be concluded that surface chemistry influences the heat transfer rate significantly. Especially at high temperatures the effect of surface chemistry is significant.

9. Conclusions

In this paper heat transfer from laminar flames of hydrogen + oxygen to a stagnation plane has been studied both analytically and numerically. The influences of strain rate, gas phase chemistry, and surface chemistry have been taken into account. It is shown that the strain rate is the dominating parameter for the heating process, but the other two effects are substantial. All the flames studied (stoichiometric hydrogen + oxygen) have an adiabatic burning temperature of 3079 K. A comparison has also been made with hydrogen–air flames. These have an adiabatic flame temperature of 2390 K at low strain rates (below 2000 s$^{-1}$). For higher strain rates the maximum flame temperature decreases, due to the fact that the flame cools down before full conversion to the final products.

Different time scales have been identified. The time scale of the flame chemistry is of the order of $10^{-5}$ s; the time scale of the boundary layer of the stagnation flow is of the order of $10^{-4}$ s. The heating time scale of the plate depends on the plate thickness. For a quartz plate with a thickness of the order of millimeters, the time scale is of the order of 1 s. The time scale increases with thickness. The heat flux is therefore not dependent on the temperature profile in the plate. The heat flux from the stagnation flow to the plate then depends only on the stagnation flow and the plate surface temperature. A relation for the convective heat transfer coefficient is introduced.

First, the heat transfer of a burnt but chemically frozen mixture stagnating to the plate was studied. The numerically determined heat transfer coefficient for the frozen hydrogen–oxygen mixture is almost proportional to the analytical coefficient. Due to the viscous sublayer, a constant factor of 0.75 difference between the analytically and the numerically determined heat transfer coefficient is observed. For the burnt and frozen hydrogen–air mixture this relation also holds, but this factor now reduces to 0.6.

Second, a numerical study on the heat transfer of a burnt and still chemically active mixture stagnating to the plate was performed. For a burnt hydrogen–oxygen mixture the heat transfer coefficient shows a clear square root dependency on the strain rate, as predicted using the analytical model. However, if the burnt mixture has not reached chemical equilibrium then the square root dependency is not strictly maintained. When the boundary layer is chemically active the heat transfer coefficient is approximately twice as high as that for the frozen mixture. This is in contrast to a hydrogen–air mixture, where the difference between a chemically active and a frozen mixture is not so large due to the smaller amount of free radicals in the reactive flow.

Finally, it is shown that the chemical boundary layer of a stagnation flame is far from chemical equilibrium. Surface chemistry is one of the means to convert some of the remaining chemical energy to heat. By assuming a surface chemistry model for platinum, it is shown that surface chemistry can influence the heat transfer significantly, especially at high temperatures.

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References