Heat Transfer of Laminar Flames of Hydrogen+Oxygen and Methane+Oxygen

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Abstract
Mixtures of a fuel and pure oxygen are used in various industrial processes. Especially in processes where a high heat generation is needed, oxy-fuel combustion is commonly used. One of the processes where a high heat input is needed is the heating and melting of (quartz) glass for the production of lamps. For this purpose, hydrogen+oxygen and methane+oxygen flames in an impinging jet like configuration are often applied. A one-dimensional numerical study was initiated to study the heat transfer from laminar oxy-fuel flames to glass. In this paper, the heat transfer rate of a methane-oxygen flame is compared to the heat transfer rate of a hydrogen-oxygen flame. Thermodynamic and chemistry phenomena are taken into account.

1 Introduction
For the production of lamps burners with mixtures of fuel+oxygen are commonly used. Premixed oxy-fuel burners convert the fuel and oxygen rapidly to products. This results in high flame speeds and high flame temperatures. The heat input from these flames to the object is high. In the lamp making process the high heat transfer rates are applied locally. Therefore, flames in an impinging-jet like configuration are commonly used. This study is performed to gain knowledge on the heat transfer of oxy-fuel flames to (quartz)-glass products.

We consider a situation in which an oxy-fuel mixture is burnt. The combusted gases impinge the object locally, which results in a local heat transfer from the burnt gases to the glass object. Since the distance from the flame to the solid surface is generally small compared to the typical width of the impingement spot, the situation can be regarded as one-dimensional. All numerical computations are performed using the one-dimensional flame code CHEM1D [3].

The heat transfer from the flow to the plate at the stagnation plane is mainly governed by convective heat transfer. The heat transfer rate was described previously for a hot inviscid gas impinging a flat plate [2]. Viscosity influences the heat transfer rate. Also the production of heat by chemical reactions influences the heat transfer significantly. Oxy-fuel flames are relatively hot and contain a lot of free radicals like O, H, OH and CO. Recombination reactions of the radicals in the cold stagnation boundary layer convert chemical energy to extra heat, thus adding an additional heat transfer mechanism.

Figure 1 shows the one-dimensional computational set-up. The flow goes from left to right. It leaves the nozzle at $x = -L$. Then the premixed mixture is ignited and a flame stabilizes at $x = -x_{stab}$. $\delta_f$ is the flame front thickness. The burnt mixture stagnates to the plate at $x = 0$. The plate has a thickness $D$. The Reynolds number based on nozzle diameter $D_n$, mean velocity $u$, density $\rho$ and viscosity $\mu$ of the gas inside 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, viscosity and density is of the order of $10^2$. From these low Reynolds numbers it is concluded that the flow is laminar.

2 Specific objectives
In [1] heat transfer rates are presented for mixtures of hydrogen+oxygen. In this paper results for mixtures of hydrogen+oxygen are compared to computations with mixtures of methane+oxygen. The heat transfer rates from the impinging burnt oxy-fuel mixtures to a product are of main interest. First a theoretical analysis is done with which the typical heat transfer rates for both mixtures can be estimated. The analysis can be done as long as the gas is inviscid and chemically inert. Different gas compositions, although chemically inert, have different thermodynamic properties. Therefore, the effect of the difference of the thermodynamic properties for the different mixtures on the heat transfer rate is studied. Recombination reactions convert chemical energy to heat. The radical and stable species concentrations in the stagnation boundary layer are studied. From this study the influence of incomplete chemistry on the heat transfer rate is determined. Furthermore, it is shown how much chemical energy could be released as heat when the chemistry would be completed to the equilibrium state. It should be noted that the study is not necessarily a quantitative study, but rather a study to compare the effect of different physical and chemical phenomena on the heat transfer for burnt mixtures of methane+oxygen and hydrogen+oxygen.

3 Results and discussion
The theoretical background is discussed briefly in the next sections, but is discussed extensively in [1]. In section 3.1 the governing equations and boundary conditions are presented. Section 3.2 discusses the thermodynamic
To take this derivative into account the local strain rate and space anymore. Then the strain rate of the mixture in is introduced [4].

3.1 Governing Equations

The conservation equations for the reacting gas mixture include conservation of mass, momentum and enthalpy in a one-dimensional configuration. Since, the heating is time dependent the equations are instationary. Figure 1 shows the numerical configuration. The gas density \( \rho_g \), species mass fractions \( Y_i \), temperature \( T \) and enthalpy \( j \) depend only on the leading coordinate \( x \). The pressure and \( y \)-velocity component \( v \) depend on both \( x \) and \( y \). On the centerline \( (y = 0) \) \( v = 0 \) but \( \partial v/\partial y \neq 0 \). To take this derivative into account the local strain rate \( K \) is introduced [4]. \( K \) depends on \( x \) and time. If the distance \( L \) is small compared to the jet width then \( K \) is such that:

\[
  v = Ky. \tag{1}
\]

The conservation of mass is given by the continuity equation [5],

\[
  \frac{\partial \rho_g}{\partial t} + \frac{\partial \rho_g u}{\partial x} = -\rho_g K, \tag{2}
\]

where \( u \) is the local velocity component in \( x \)-direction. \( \rho_g K \) describes the loss of mass in \( y \)-direction, which results from 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. [4] and [6]):

\[
  \frac{\rho_g}{\partial t} + \frac{\partial \rho_g u}{\partial x} = \rho_g \frac{\partial K}{\partial x} = \frac{1}{j+1} \left( \rho_g u a^2 - \rho_g K^2 \right) \tag{3}
\]

where \( j = 0 \) for a planar and \( j = 1 \) for an axisymmetric flow. \( K = a \) equals the strain rate at the inflow boundary \( (x = -L) \) and \( \mu \) is the dynamic viscosity. If the flame is stabilized relatively far from the stagnation plane compared to the flame thickness, \( K \) does not change in time and space anymore. Then the strain rate of the mixture in the burnt gases becomes

\[
  K_m = a \left( \frac{\rho_u}{\rho_m} \right)^{1/2}. \tag{4}
\]

\( \rho_u \) is equal to the density of an adiabatic equilibrium mixture. Because oxy-fuel combustion develops rapidly to equilibrium, the flames considered can be regarded as thin. As a result, the maximum strain rate before the mixture enters the stagnation boundary layer is close to \( K_m \). Therefore, we will take the maximum strain rate as \( K_m \). In a chemically reacting flow, a conservation equation for every species \( i \) has to be solved

\[
  \frac{\partial \rho_g Y_i}{\partial t} + \frac{\partial \rho_g u Y_i}{\partial x} = \frac{\partial}{\partial x} \left( \rho_g D_{im} \frac{\partial Y_i}{\partial x} \right) - \dot{\rho}_i = -\rho_g K Y_i, \tag{5}
\]

with \( D_{im} \) the effective diffusion coefficient, which is determined using a mixture averaged diffusion model [7], [8]. Furthermore, the chemical source term is quantified by \( \dot{\rho}_i = M_i \sum_k v_{ik} \alpha_k \) where \( M_i \) is the molar mass of species \( i \), \( v_{ik} \) the stoichiometric coefficient of species \( i \) in reaction \( k \), and \( \alpha_k \) the reaction rate of reaction \( k \). The model of Smooke [9] is used for all chemical kinetic computations. The energy conservation equation for the gas is written in terms of the specific enthalpy \( j \) and temperature \( T \):

\[
  \frac{\partial \rho_g j}{\partial t} + \frac{\partial \rho_g u j}{\partial x} - \frac{\partial}{\partial x} \left( \rho_g \frac{\partial T}{\partial x} \right) - \frac{\partial}{\partial x} \left( \sum_{i=1}^{N} h_i \rho_g D_{im} \frac{\partial Y_i}{\partial x} \right) = -\rho_g K j. \tag{6}
\]

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

\[
  j_i^0 = j_i^0 + \int_{T_0}^{T} c_{pg,i}(T^*)dT^*, \tag{7}
\]

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

\[
  \rho_g = \frac{pM}{RT} \tag{8}
\]

with \( R \) the universal gas constant and \( M \) the average molar mass of the mixture. For low Mach number flows, the pressure in this last expression is almost constant \( p = p_a \), with \( p_a \) the atmospheric pressure at the inlet.

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

\[
  \rho_p c_{pp} \frac{\partial T}{\partial t} = \frac{\partial}{\partial x} \left( \rho_p c_p \frac{\partial T}{\partial x} \right). \tag{9}
\]

with \( \rho_p \), \( c_{pp} \) and \( \lambda_p \) the density, specific heat capacity and thermal conductivity coefficients of the plate.
The boundary conditions at the gas inlet side \((x = -L)\) are Dirichlet boundary conditions. The inlet temperature \(T = T_u\), pressure and species concentrations \(Y_i = Y_{i,u}\) are defined, together with the applied strain rate \(K = a\). The inlet velocity \(u = u_0\) is then determined from the laminar burning velocity \(s_l\), strain rate, and distance from the flame to the inlet. Mixtures of hydrogen+oxygen and methane+oxygen are used here. The laminar burning velocity of a hydrogen+oxygen mixture is approximately three times the laminar burning velocity of a methane+oxygen mixture. Therefore, it is expected that the inlet strain rate for a hydrogen+oxygen mixture is higher than for a methane+oxygen mixture. However, for reasons of comparison the applied strain rate is set to \(a = 6000\) \(s^{-1}\) for both mixtures. Since the density decrease for a methane+oxygen flame is higher than for a hydrogen+oxygen flame, it is expected that \(K_m\) is higher for a methane+oxygen mixture than for a hydrogen+oxygen mixture. The boundary conditions at the stagnation plane can be Dirichlet or Neumann boundary conditions. Both components of the velocity are zero for all \(y\) so that \(u = 0\) and \(K = 0\) at \(x = 0\). The temperature follows from continuity of conductive heat fluxes at the stagnation plane:

\[
\lambda_g \frac{\partial T}{\partial x} \bigg|_{x=0^-} = \lambda_p \frac{\partial T}{\partial x} \bigg|_{x=0^+}. \tag{10}
\]

At the other plate surface \((x = D)\) the plate is assumed to be adiabatic.

### 3.2 Heat transfer rates

This paper focusses on the heat transfer from flames to products. The products have the geometry of a plate and the thermodynamic properties of glass. Combustion of oxy-fuel mixtures with light fuel compounds is relatively fast. It was shown \([1]\) that the heat flux \(q\) from an impinging jet of burnt gases to a plate can be described in terms of the stagnation wall temperature \(T(0)\) only. This was explained as follows. The typical heating timescale of the stagnation boundary layer is \(\tau_b = 1/K_m\). For an applied strain rate of \(a = 6000\) \(s^{-1}\) the maximum strain rates for the hydrogen+oxygen and methane+oxygen mixtures are \(1.7 \cdot 10^5\) and \(2.0 \cdot 10^5\) \(s^{-1}\), respectively. The typical heating timescale of the product, in this case a plate of 1-5 mm, is of the order of \(10^{-1}-10^1\) s. The typical heating timescale of the stagnation boundary layer is much shorter than the typical heating timescale of the product. It was concluded that the stagnation boundary layer adapts instantly to the plate and is dependent on the wall temperature \(T(0)\) only. Therefore, the heat flux from the stagnation boundary layer to the plate depends only on \(T(0)\). Figure 2 shows the heat fluxes from the combustion gases to the plate for stoichiometric mixtures of hydrogen+oxygen and methane+oxygen. These results are from time-dependent calculations. A plate of 5 mm thickness and uniform temperature of 700 K is heated by stagnating flue gases. With time, the wall temperature \(T(0)\) increases and therefore the temperature difference between the hot flue gases and the wall decreases, which results in a decrease of the heat flux. The overall heat flux from the methane mixture is somewhat lower than that of the hydrogen mixture. The difference is partly a result of the differences in thermodynamic quantities of the burnt mixtures and partly a result of the differences in the chemical composition of the stagnation layer. The influence of the thermodynamic behaviour of the stagnation flow on the heat transfer rate is discussed in this section. The influence of chemistry in the boundary layer on the heat transfer rate is discussed in the next section.
is found [1], [2]:

\[
\h_{\text{ref}} = \lambda_g \left( \frac{2\kappa}{\pi} \right)^{1/2}.
\]

(11)

where \(\kappa\) is a function of the strain rate and is equal to \(\kappa = \rho_g c_{pg}K_m/\lambda_g\). \(\rho_g, c_{pg}\) and \(\lambda_g\) are the density, specific heat capacity and thermal conductivity coefficient of the gas near \(x = 0\) respectively. Figure 3 shows \(h_{\text{ref}}\) as function of \(T(0)\) for both mixtures. Although in our calculations \(K_m\) is higher for a methane-oxygen mixture than for a hydrogen-oxygen mixture \(h_{\text{ref}}\) is comparable. This means that the thermodynamic variables and density in (11) stimulate a higher heat transfer rate for the methane-oxygen mixture than for the hydrogen-oxygen mixture. With the heat flux (see Fig. 2) and adiabatic flame temperature known the heat transfer coefficients \(h\) of the reactive mixtures are determined. Figure 3 shows \(h\) as function of \(T(0)\). For both mixtures the heat transfer coefficient is approximately 50% higher than the expected value for an inert and inviscid stagnation flow. Viscosity reduces the heat transfer rate while chemical reactions in the stagnation layer enhance the heat transfer [1], [10]. In the stagnation boundary layer a lot of radicals exist that can recombine to stable molecules. Recombination leads to extra heat generation and thus leads to a higher heat transfer rate.

### 3.3 Chemical boundary layer

Recombination reactions in the stagnation boundary layer enhance the heat transfer rate from the burnt gases to the plate. This section studies the amount of major free radicals and stable molecules in the stagnation boundary layer. Furthermore, it is studied how far the compositions are from chemical equilibrium.

In the ideal case a stoichiometric hydrogen+oxygen mixture is completely converted to water releasing the maximum amount of heat. However, in reality the burnt mixture contains a lot of radicals. Radicals are in a higher chemical energy state than the stable molecules and possess more chemical and less thermal energy. Figure 4 shows the species concentrations of the major radicals in a burnt hydrogen+oxygen mixture at the stagnation wall \(x = 0\) as function of the wall temperature \(T(0)\). It should be kept in mind that the concentrations presented here are mole fractions and not absolute radical amounts. When the wall is relatively cold (low \(T(0)\)) the overall radical concentration is also low. There is only some H radicals. However at higher surface temperatures the amount of O and especially OH radicals increase. The amount of OH is sensitive to the temperature and is commonly used as temperature indicator. In a burnt methane-oxygen mixture impinging the wall (Fig. 5) equal trends are observed. Although the concentrations of the aforementioned radicals are somewhat lower, we now have a significant amount of CO molecules. In the chemical reaction mechanism for methane+oxygen combustion, a slow step in which much thermal energy is released is the formation of carbon dioxide from carbon monoxide. This is the final step in the reaction mechanism. A lot of CO in the gas indicates that there is a lot of chemical energy that is not released as heat. Because of the significant amount of CO in a burnt methane+oxygen mixture the heat transfer rate from the gas to the plate is lower for a methane+oxygen mixture than for a hydrogen+oxygen mixture.

Cooling of the mixture leads to recombination of radicals to other radicals, to products or even back to the reactants. Reactants and products are stable molecules. Figure 6 shows the reactants and products of the hydrogen+oxygen mixture. At low temperatures, most of the hydrogen and oxygen is converted to water. Due to the large gradients close to the plate and due to the high diffusivity of hydrogen, a significant amount of atomic and molecular hydrogen is observed at \(x = 0\) while the concentrations of atomic and molecular oxygen are
Figure 6: Species mole fractions of stable species H\textsubscript{2}, O\textsubscript{2}, and H\textsubscript{2}O at x = 0 for a mixture of hydrogen+oxygen.

Figure 7: Species mole fractions of stable species H\textsubscript{2}, O\textsubscript{2}, H\textsubscript{2}O and CO\textsubscript{2} at x = 0 for a mixture of methane+oxygen.

low. More upstream relatively more oxygen can be found. At higher wall temperatures the reactant concentrations increase while the water content decreases. The total amount of chemical heat released by recombination reactions becomes smaller at high temperature, but is still significant.

The contribution of the flow to the heat transfer rate is mainly an almost linear function of the temperature difference (T\textsubscript{m} − T(0)). The contribution of the chemical reactions to the heat transfer rate is not necessarily a linear function of (T\textsubscript{m} − T(0)). Therefore, it is possible that in Fig. 3 h increases while the overall heat production by chemical reactions decreases.

Figure 7 shows the concentrations of the major stable species in a burnt methane+oxygen mixture. The amounts of hydrogen and oxygen are comparable. Diffusion of hydrogen with respect to oxygen is not as high as with the hydrogen+oxygen mixture. This is due to the existence of the large amount of the relatively heavy CO\textsubscript{2} molecule and the comparable amount of the in weight comparable CO molecule. Furthermore, the initial unburnt methane+oxygen mixture contains less H elements than in a hydrogen+oxygen mixture.

Conversion from all reactants of the unburnt mixture to stable products would mean a maximum conversion of chemical energy to heat. However, it is not always possible to reach this. The maximum decrease of chemical energy occurs when the mixture is converted from the initial mixture to a mixture in chemical equilibrium. The chemical equilibrium depends on the local conditions, in our case the conditions at the plate surface. However, the stagnating mixture might not have reached chemical equilibrium. The concentrations of the products in the stagnation flow are now compared to the concentrations of the products in a mixture in equilibrium with the same thermal conditions. Figure 8 shows the concentrations of H\textsubscript{2}O in a burnt mixture and in a mixture in equilibrium at x = 0. Obviously, the stagnation flow at the plate surface is not in chemical equilibrium. Only at temperatures close to the adiabatic burning temperature the burnt mixture tends to go to chemical equilibrium. The combustion of oxy-fuel mixtures is relatively fast and therefore the flame front is thin. Chemical equilibrium is reached fast in the flame front. When the burnt gases hit a surface at the same temperature then the mixture will stay in chemical equilibrium. The two lines in Fig. 8 collapse at an adiabatic burning temperature of approximately 3078 K. The same holds for the methane+oxygen mixture (see Fig. 9), where the adiabatic burning temperature is approximately 3045 K. The total difference between the equilibrium values and the burnt methane+oxygen mixture is even larger than for the hydrogen+oxygen mixture. Furthermore, the formation step of CO\textsubscript{2} is a relatively energetic step. Driving the formation of CO\textsubscript{2} enhances the heat transfer significantly. Finally, the composition not only changes with temperature, but also changes when the flow conditions like strain rate change. This not taken into account in this paper.

4 Conclusions

In this paper the heat transfer of laminar hydrogen+oxygen and methane+oxygen flames to a stagnation plane has been studied numerically. It was shown that at a similar applied strain rate the heat transfer rate from a burnt methane+oxygen mixture to a plate does not deviate a lot from a burnt hydrogen+oxygen mixture.

The composition in the stagnation boundary layer is different from a burnt hydrogen+oxygen mixture than for a methane+oxygen mixture. Both stagnation layers have a different density and different values of thermodynamic properties. This affects the heat transfer rate.

Chemical recombination reactions of the radicals in
the stagnation boundary layer influence the heat transfer significantly. Recombination reactions lead to extra heat generation and thus to a higher heat transfer rate. It is shown for a burnt hydrogen+oxygen mixture that at low temperature of the stagnation plane, most of the hydrogen and oxygen is converted to water, releasing the maximum amount of heat. At high stagnation plane temperatures the water content decreases and less heat is released. Furthermore, it is shown that the stagnation boundary layer of a burnt methane+oxygen mixture contains a lot of CO, which has a high chemical energy level. The high CO concentration reduces the heat transfer rate.

Also, it is shown that the chemical boundary layer of a stagnation flame is not in chemical equilibrium, especially at low temperatures. Near the stagnation plane, the difference between the overall product concentration in a burnt mixture and a mixture in chemical equilibrium is larger for a methane+oxygen mixture than for a hydrogen+oxygen mixture. This is mainly due to the large amount of CO for the methane flame. Driving the formation of CO$_2$ would enhance the heat transfer significantly.

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**References**


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Figure 8: Species mole fractions of H$_2$O at $x = 0$ for a reactive burnt hydrogen+oxygen mixture (solid line) and for a mixture in chemical equilibrium (dashed line).

Figure 9: Species mole fractions of H$_2$O and CO$_2$ at $x = 0$ for a reactive burnt methane+oxygen mixture (solid line) and for a mixture in chemical equilibrium (dashed line).