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Citation for published version (APA):

DOI:
10.1080/13647830.2017.1311028

Document status and date:
Published: 01/08/2017

Document Version:
Publisher’s PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

• A submitted manuscript is the author's version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
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To cite this article: Zhen Zhou, Francisco E. Hernández-Pérez, Yuriy Shoshin, Jeroen A. van Oijen & Laurentius P.H. de Goey (2017) Effect of Soret diffusion on lean hydrogen/air flames at normal and elevated pressure and temperature, Combustion Theory and Modelling, 21:5, 879-896, DOI: 10.1080/13647830.2017.1311028

To link to this article: http://dx.doi.org/10.1080/13647830.2017.1311028

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Published online: 12 Apr 2017.
Effect of Soret diffusion on lean hydrogen/air flames at normal and elevated pressure and temperature

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(Received 18 October 2016; accepted 12 March 2017)

The influence of Soret diffusion on lean premixed flames propagating in hydrogen/air mixtures is numerically investigated with a detailed chemical and transport models at normal and elevated pressure and temperature. The Soret diffusion influence on the one-dimensional (1D) flame mass burning rate and two-dimensional (2D) flame propagating characteristics is analysed, revealing a strong dependency on flame stretch rate, pressure and temperature. For 1D flames, at normal pressure and temperature, with an increase of Karlovitz number from 0 to 0.4, the mass burning rate is first reduced and then enhanced by Soret diffusion of H2 while it is reduced by Soret diffusion of H. The influence of Soret diffusion of H2 is enhanced by pressure and reduced by temperature. On the contrary, the influence of Soret diffusion of H is reduced by pressure and enhanced by temperature. For 2D flames, at normal pressure and temperature, during the early phase of flame evolution, flames with Soret diffusion display more curved flame cells. Pressure enhances this effect, while temperature reduces it. The influence of Soret diffusion of H2 on the global consumption speed is enhanced at elevated pressure. The influence of Soret diffusion of H on the global consumption speed is enhanced at elevated temperature. The flame evolution is more affected by Soret diffusion in the early phase of propagation than in the long run due to the local enrichment of H2 caused by flame curvature effects. The present study provides new insights into the Soret diffusion effect on the characteristics of lean hydrogen/air flames at conditions that are relevant to practical applications, e.g. gas engines and turbines.

Keywords: Soret diffusion; hydrogen/air; mass burning rate; cellular flame

1. Introduction

Hydrogen is a promising fuel for the near future, especially for stationary power generation using heavy duty gas turbines [1]. The combustion characteristics of hydrogen differ considerably from traditional hydrocarbon fuels due to its high diffusivity [2]. Therefore, diffusive transport of hydrogen in flames has drawn much attention [3–5].

Diffusive transport in flames is driven by various modes [2,6]. The dominant mode is Fickian diffusion, which is caused by concentration gradients of species. Another mode is Soret diffusion caused by temperature gradients. Soret diffusion is considered as a second-order mass diffusion mode which is usually neglected in reacting flow modelling. Since Soret diffusion drives light species towards high temperature zones and heavy species away from high temperature zones, it has to be taken into account for hydrogen/air flames.
Table 1. Summary of conditions for the studied cases with $\phi = 0.4$. The zero-stretch laminar flame speed ($S_{0u}$), zero-stretch laminar flame thickness ($\delta_{0f}$) and zero-stretch mass burning rate ($m^0$) at the three different conditions are included.

<table>
<thead>
<tr>
<th>Condition</th>
<th>$P$ (bar)</th>
<th>$T_u$ (K)</th>
<th>$S_{0u}$ (cm/s)</th>
<th>$\delta_{0f}$ (cm)</th>
<th>$m^0$ (g/(cm²s))</th>
</tr>
</thead>
<tbody>
<tr>
<td>With Soret diffusion</td>
<td>I(REF)</td>
<td>1</td>
<td>300</td>
<td>22.9</td>
<td>0.061</td>
</tr>
<tr>
<td></td>
<td>II(HP)</td>
<td>5</td>
<td>300</td>
<td>6.36</td>
<td>0.032</td>
</tr>
<tr>
<td></td>
<td>III(HT)</td>
<td>1</td>
<td>500</td>
<td>109.5</td>
<td>0.045</td>
</tr>
<tr>
<td>Without Soret diffusion</td>
<td>I(REF)</td>
<td>1</td>
<td>300</td>
<td>23.6</td>
<td>0.060</td>
</tr>
<tr>
<td></td>
<td>II(HP)</td>
<td>5</td>
<td>300</td>
<td>6.37</td>
<td>0.032</td>
</tr>
<tr>
<td></td>
<td>III(HT)</td>
<td>1</td>
<td>500</td>
<td>115.9</td>
<td>0.044</td>
</tr>
</tbody>
</table>

when accurate prediction of flame behaviour is needed owing to the presence of the light and highly diffusive H₂ and steep temperature gradients. In addition, for lean stretched hydrogen/air flames, the Soret diffusion effect on flame behaviour can also be significantly changed by preferential diffusion due to the local redistribution of molecular hydrogen [4].

A number of numerical studies have been reported in the literature concerning Soret diffusion and its coupling effects with preferential diffusion on laminar hydrogen flames. Ern and Giovangigli [3,7] investigated the effects of Soret diffusion on planar and counterflow hydrogen/air flames, showing that Soret diffusion reduces the zero-stretch laminar flame speed at the lean side and extends the stretch-induced lean extinction limit. Bongers and de Goey [8] studied the effects of Soret diffusion on the laminar flame speed of hydrogen flames, showing that a 10% error in the laminar flame speed is obtained without inclusion of Soret diffusion. Yang et al. [4,9] further studied the Soret diffusion effects in hydrogen/air flames with emphasis on a mechanistic interpretation, clarifying the roles of molecular hydrogen and the hydrogen radical. Grcar et al. [10] studied the Soret diffusion impact on lean hydrogen/air flames and indicated that flame cells computed with Soret diffusion are smaller and sharper. All of the above studies focused on Soret diffusion effects at normal pressure and temperature. Liang et al. [5] investigated the effects of Soret diffusion on the laminar flame speed and Markstein length of stoichiometric hydrogen-enriched syngas flames at normal and elevated pressures and temperatures. It turned out that Soret diffusion reduces the zero-stretch mass burning rate and Markstein length at normal pressure and temperature. It was further shown that the influence of Soret diffusion on the zero-stretch mass burning rate increases, while that on Markstein length decreases, if pressure or temperature is increased. However, the Soret diffusion effect on the stretched mass burning rate at elevated pressure and temperature remains unknown. For 2D freely-propagating flames, although the propagating characteristics of hydrogen/air flames have been widely studied at normal and elevated pressures and temperatures (e.g. see [11–13]), the influence of Soret diffusion on the propagation dynamics of lean hydrogen/air flames is poorly understood at elevated pressure and temperature. Understanding the influence of Soret diffusion on the propagating characteristics of lean hydrogen/air flames at normal and elevated pressure and temperature is of interest and importance from both fundamental and practical viewpoints and is further studied in this work.

In this regard, a systematic numerical study is conducted to evaluate the effect of Soret diffusion on the propagating characteristics of lean premixed hydrogen/air flames at the conditions summarised in Table 1 with an equivalence ratio $\phi = 0.4$. Firstly, the influence of Soret diffusion on 1D flame mass burning rate at elevated pressure and temperature is investigated as a function of Karlovitz number (Ka). The roles of Soret diffusion of H₂
and H are identified at different conditions. Subsequently, a mechanistic interpretation is provided about the effects of pressure and temperature on the Soret diffusion of H$_2$ and H for different values of Ka. Secondly, the extent of the Soret diffusion effect on 2D freely-propagating flames is studied. Such flames are strongly curved and stretched and show cellular instabilities. Soret diffusion changes the flame front evolution significantly and the effects largely depend on temperature and pressure. The global consumption speed with and without Soret diffusion at elevated pressure and temperature is quantified as a function of flame front length. Finally, the correlations between local consumption speed and curvature which represent the early phase and the developed phase of flame front evolution at elevated pressure and temperature are discussed.

2. Governing equations

The flames under study are mathematically described by conservation equations for mass, momentum, energy and species, treating the flow as a continuous, multicomponent, compressible and thermally-perfect mixture of gases. A Newtonian flow is assumed, obeying the ideal gas equation of state, with negligible thermal radiation or gravity effects. The 2D conservation equations for a mixture consisting of $N$ chemical species evolving in time, $t$, and space, $\vec{x}$, can be written in tensor notation as

$$\frac{\partial \rho}{\partial t} + \frac{\partial (\rho u_j)}{\partial x_j} = 0,$$

$$\frac{\partial (\rho u_j)}{\partial t} + \frac{\partial (\rho u_j u_j + \delta_{ij} p)}{\partial x_j} - \frac{\partial \tau_{ij}}{\partial x_j} = 0,$$

$$\frac{\partial (\rho E)}{\partial t} + \frac{\partial [(\rho E + p) u_j]}{\partial x_j} - \frac{\partial (\tau_{ij} u_i)}{\partial x_j} + \frac{\partial q_j}{\partial x_j} = 0,$$

$$\frac{\partial (\rho Y_\alpha)}{\partial t} + \frac{\partial [\rho Y_\alpha (u_j + V_{j,\alpha})]}{\partial x_j} = \dot{\omega}_\alpha,$$

with the indices $i, j = 1, 2$ and $\alpha = 1, \ldots, N - 1$. The Einstein summation convention applies to the indices $i$ and $j$. In the expressions above, $\rho$ is the mixture density, $u_i$ is the mixture velocity, $p$ is the mixture pressure, $T$ is the mixture temperature, $E$ is the total mixture energy (including chemical energy or heat of formation), $Y_\alpha$ is the mass fraction of species $\alpha$, $\dot{\omega}_\alpha$ is the net reaction rate of species $\alpha$, $\tau_{ij}$ is the viscous stress tensor, $q_j$ is the heat flux vector (energy flux due to conduction plus energy flux due to diffusion) and $V_{j,\alpha}$ is the diffusion velocity of species $\alpha$. Additionally, $\delta_{ij}$ denotes the Kronecker delta. By neglecting pressure-gradient terms in the full multicomponent transport formulation while accounting for Soret and Dufour effects, the heat flux vector can be expressed as

$$\vec{q} = \rho \sum_{\alpha=1}^{N} h_\alpha Y_\alpha \vec{V}_\alpha - \lambda \nabla T - \mathcal{RT} \sum_{\alpha=1}^{N} \frac{D_{T\alpha} \nabla X_\alpha}{M_\alpha X_\alpha},$$

and the diffusion velocity is given by

$$\vec{V}_\alpha = \frac{1}{X_\alpha \mathcal{M}} \sum_{\beta \neq \alpha}^{N} \mathcal{M}_\beta D_{\alpha,\beta} \nabla X_\beta - \frac{D_{T\alpha}}{\rho Y_\alpha} \frac{\nabla T}{T}.$$
In Equations (5) and (6), \( R \) is the universal gas constant, \( \lambda \) is the mixture thermal conductivity, \( M \) is the mixture mean molar mass, and \( M_\alpha, X_\alpha, h_\alpha \) and \( D^T_\alpha \) are the molar mass, mole fraction, enthalpy and thermal diffusion coefficient of species \( \alpha \), respectively. The multicomponent diffusion coefficients are denoted by \( D_{\alpha,\beta} \). The last term of Equation (6) is the so-called Soret diffusion.

3. Computational setup

1D and 2D freely-propagating flame configurations are considered in this study. For 1D cases, the zero-stretch and stretched mass burning rates of hydrogen/air flames are calculated at the conditions summarised in Table 1 with and without the inclusion of Soret diffusion utilising the in-house code CHEM1D [14,15]. The 1D domain is from \( x = -0.5 \) to 1 cm. Species and enthalpy are used for the boundary condition. At the unburned side, a Dirichlet type of boundary condition is utilised, while, at the burnt side, Neumann type boundary conditions are implemented. For the calculation of 1D stretched flames, stretch rate is introduced in the 1D governing equations to model mass loss in the other spatial coordinates. Derivation of the 1D governing equations with stretch rate can be found in [16]. In the simulation, constant stretch rates are applied for the stretched flames. More details of the calculation can be found in [17] and thus are not repeated here. Transport properties are computed using the EGLIB library [18,19]. The grid is adaptively refined consisting of at least 500 points so that the flame structure is well resolved.

For 2D cases, flames are simulated at the same conditions as in the 1D cases. The set of governing equations is solved by a body-fitted, multi-block, adaptive mesh refinement (AMR), finite-volume framework that has been originally developed by Groth and co-researchers [20–22]. In the simulations, the computational domain is a rectangular channel with 2 cm in the \( x \)-direction and 1 cm in the \( y \)-direction. The upper and lower boundaries (\( y \)-direction) are periodic, while the left and right boundaries (\( x \)-direction) are a subsonic inflow and a subsonic outflow, respectively. The inlet velocity for each case is set to be the corresponding laminar flame speed. The thermodynamic properties and transport properties are computed using the open-source package CANTERA [23]. The solutions are obtained with up to three levels of refinement, achieving a minimum resolution of 30 m for conditions I (REF) – Reference and III (HT) – Elevated Temperature, and with up to four levels of refinement with a minimum resolution of 15 m for condition II (HP) – Elevated Pressure. These levels of resolution are found to be sufficient to get grid-independent solutions. The computations are initialised with piecewise linear profiles of species mass fractions and temperature by knowing the fresh and burned mixture composition and utilising linear interpolation in the laminar flame layer. The flame is initially located at \( x_f = 1.5 \) cm. The initial spatial perturbation of the flame front is given by

\[
x_f = x_{f0} + A \sin(2\pi n y),
\]

where \( A \) is the amplitude of the disturbance and \( n \) is the wave number of the sinusoidal disturbance. In the simulations, \( A \) is set to 0.1 times the laminar flame thickness and \( n \) is 6.

The 1D zero-stretch flame thickness is defined as

\[
\delta^0_T = \frac{(T_b - T_u)}{|\partial T/\partial s|_{\text{max}}},
\]
where $T_b$ and $T_u$ are the burnt and unburnt gas temperatures and $\partial T/\partial s$ is the gradient of temperature. The reaction mechanism of hydrogen developed by Li et al. [24], which includes 11 species and 21 elementary reactions, is employed for both the 1D and 2D simulations.

4. Results and discussion

4.1. 1D flames

The mass burning rates of hydrogen/air mixtures with an equivalence ratio $\phi = 0.4$ as a function of Karlovitz number at the three conditions predicted with Soret diffusion are compared with those without Soret diffusion, as shown in Figure 1. The Karlovitz number is defined as

$$Ka = \frac{\delta_0}{S^U_0} K,$$

where $K$ is the flame stretch rate. The mass burning rate is extracted from the inner layer position defined at the position of maximum heat release rate. It is observed that, for condition I (REF), Soret diffusion reduces the mass burning rate slightly for $Ka < 0.2$ and enhances the mass burning rate slightly for $Ka > 0.2$. At the higher pressure of condition II (HP), however, Soret diffusion enhances the mass burning rate at all tested $Ka$ values. The increment of the mass burning rate caused by Soret diffusion enlarges with the increase of $Ka$. On the contrary, at the higher temperature of condition III (HT), Soret diffusion reduces the mass burning rate at all tested values of $Ka$. The decrease of mass burning rate caused by Soret diffusion diminishes with the increase of $Ka$. It should be noted that the influence of Soret diffusion on the mass burning rate for $Ka = 0$ at $\phi = 0.4$ decreases with increasing pressure, while that at $\phi = 1$ increases with the increase of pressure [5], which indicates that the response of the Soret diffusion effect to pressure depends on the equivalence ratio. However, the response of the Soret diffusion effect to temperature at
\( \phi = 0.4 \) is consistent with that at \( \phi = 1 \) [5]. This demonstrates that the Soret diffusion effect on mass burning rate is not straightforward and strongly depends on the coupling of flame stretch, equivalence ratio, pressure and temperature.

As the Soret diffusion in hydrogen/air flames significantly impacts the distributions of \( \text{H}_2 \) and \( \text{H} \) [4], the effects of Soret diffusion of \( \text{H}_2 \), \( \text{H} \) and \( \text{H}_2+\text{H} \) on the mass burning rate are quantified and compared for the three conditions. The results are scaled by the corresponding mass burning rate for different \( \text{Ka} \) values predicted without Soret diffusion. In Figure 2, it is demonstrated that the Soret diffusion impact on the mass burning rate is dominated by the Soret diffusion of both \( \text{H}_2 \) and \( \text{H} \) for conditions I (REF) and III (HT) and dominated by Soret diffusion of \( \text{H}_2 \) for condition II (HP). Figure 2(a) shows that, at condition I (REF), Soret diffusion of \( \text{H}_2 \) reduces the mass burning rate for \( \text{Ka} \) in the range 0–0.04. For \( \text{Ka} \) > 0.04, it enhances the mass burning rate. However, at all the tested values of \( \text{Ka} \), Soret diffusion of \( \text{H} \) reduces the mass burning rate. It is observed in Figure 2(b) that, at condition II (HP), Soret diffusion of \( \text{H}_2 \) enhances the mass burning rate and Soret diffusion of \( \text{H} \) reduces the mass burning rate only slightly at all the tested values of \( \text{Ka} \). In Figure 2(c), Soret diffusion of \( \text{H}_2 \) and \( \text{H} \) at condition III (HT) shows the same trend as that in condition I (REF). Nevertheless, at condition III (HT) the influence on mass burning rate caused by Soret diffusion of \( \text{H}_2 \) decreases, while that caused by Soret diffusion of \( \text{H} \) increases, as compared with their respective counterparts at condition I (REF). A mechanistic interpretation of these observations is provided in the following.

The effect of Soret diffusion on the mass burning rate of lean hydrogen/air flames with the increase of \( \text{Ka} \) mainly results from the coupling effect between preferential diffusion and Soret diffusion. Owing to the preferential diffusion, Soret diffusion leads to additional changes in local equivalence ratio and affects the chemical reaction rates. This is illustrated in Figure 3 for condition I (REF). The calculation of local equivalence ratio is based on the definition provided in [25], which is expressed as

\[
\phi = \frac{0.5 Z_H}{Z_O},
\]

where \( Z_\alpha \) is the element mass fraction of \( \alpha \), with \( \alpha = \text{H} \) or \( \text{O} \). According to previous studies [4,5], the major relevant reaction affected by the Soret diffusion of \( \text{H} \) is \( \text{H} + \text{O}_2 = \text{OH} + \text{O} \) (R1) (chain branching), which is plotted in Figure 3 as well. Figure 3 shows that
the local equivalence ratio and reaction rate of $\text{R1}$ for $Ka = 0$ and $0.4$ at condition I (REF) are affected by Soret diffusion as a function of normalised temperature. The normalised temperature is defined as

$$c = \frac{(T - T_u)}{(T_h - T_u)},$$

where $T$ is the local temperature. For $Ka = 0$, the local equivalence ratio is reduced by both $H_2$ Soret diffusion and total Soret diffusion throughout the flame and the reaction rate of $\text{R1}$ is reduced by total Soret diffusion slightly. For $Ka = 0.4$, the local equivalence ratio in the preheat zone is reduced, but in the high temperature region it is enhanced by both $H_2$ Soret diffusion and total Soret diffusion. In this case, the reaction rate of $\text{R1}$ is enhanced by $H_2$ Soret diffusion and total Soret diffusion due to the local enrichment at high temperature.
should be noted that the reaction rates of R1 with total Soret diffusion for both $Ka = 0.0$ and 0.4 are slightly lower than those with $H_2$ Soret diffusion only, due to the inclusion of $H$ Soret diffusion.

To illustrate further the Soret diffusion effect on the mass burning rate for different $Ka$ values at elevated pressure and temperature, the $H_2$ and $H$ Soret diffusion fluxes for three different $Ka$ values at the three conditions as a function of normalised temperature are presented in Figure 4. The Soret diffusion fluxes are scaled by the corresponding mass burning rates. The changes in local equivalence ratio and mass fraction of $H$ due to Soret diffusion for the corresponding cases are plotted in Figure 5. The changes are determined with respect to the solution without Soret diffusion as a function of the normalised temperature, $c$. The reaction zone and the active zone of R1 for $Ka = 0.4$, represented by two vertical dash–dotted lines at the three conditions, are superimposed in Figure 5 (left) and Figure 5 (right) for reference, respectively. The left and right boundaries of the reaction zone are defined at the points of maximum heat release rate and 0.02 times maximum heat.
release rate. The active zone of R1 is the region where the reaction rate of R1 is larger than 50% of its peak value [5]. Additionally, the Soret diffusion flux is non-negligible only when both $\nabla T/T$ and the concentration of these species are large. Therefore, the $\nabla T/T$ and mass fractions of $H_2$ and $H$ for $Ka = 0.0$ and 0.4 at the three conditions are plotted in Figure 6. It is seen in Figure 4 that the Soret diffusion flux of $H_2$ is positive, and it peaks at the upstream side of the flame for the three conditions. The peak at the upstream side is because both $\nabla T/T$ and the mass fraction of $H_2$ at the upstream side of the flame are larger than those at the downstream side, as shown in Figure 6. It reflects that $H_2$ is transported by Soret diffusion of $H_2$ from the upstream side of the flame to the downstream side. For the three conditions, the Soret diffusion flux of $H_2$ increases with increasing $Ka$ at the upstream side, while it decreases slightly at the downstream side. This is because both the $H_2$ concentration and $\nabla T/T$ increase with the increase of $Ka$ at the upstream side due to preferential diffusion effects (Figure 6). However, both $\nabla T/T$ and the $H_2$ concentration
Figure 6. Effects of flame stretch rate, pressure and temperature on the temperature gradient normalised by temperature itself and the mass factions of H$_2$ and H. (Colour online)

decrease slightly as a function of Ka at the downstream side for conditions I and I and the H$_2$ concentration decreases for condition II. Consequently, more H$_2$ is transported to the high temperature region of the flames by Soret diffusion of H$_2$ with the increase of Ka and less H$_2$ is moved out. Indeed, as shown in Figure 5, the increase in local equivalence ratio in the high temperature region caused by Soret diffusion of H$_2$ increases with the increase of Ka, which leads to the rise of the mass burning rate. This finally explains the increase of mass burning rate at all the tested conditions as a function of Ka in Figure 2.

In this paragraph, the influences of pressure and temperature on the Soret diffusion of H$_2$ are interpreted. With the increase of pressure, the net Soret diffusion flux of H$_2$ increases greatly due to the increase of $\nabla T/T$ (Figure 6), as compared with that at 1 bar. However, the increase of mass burning rate (Figure 1) is larger than the increase of Soret diffusion flux of H$_2$ due to the increase of pressure from 1 to 5 bar. As a result, the Soret diffusion flux of H$_2$ scaled with the mass burning rate decreases with pressure (Figure 4). This effect is larger with increasing Ka. A similar effect is observed with the increase of temperature (Figure 4). Furthermore, the increase in scaled Soret diffusion flux of H$_2$ is weaker at conditions II (HP) and III (HT) than that at condition I with the increase of Ka. This indicates that the increase in H$_2$ transported to the high temperature zone at conditions II (HP) and III (HT) is less than that at condition I with the increase of Ka. This leads to the fact that, for condition III (HT), the fraction of the reaction zone with enhanced local equivalence ratio is less than that at condition I (REF) for a fixed Ka, as shown in Figure 5 (left). Therefore, the influence on mass burning rate caused by Soret diffusion of H$_2$ is less sensitive to the increase of Ka at elevated temperature as observed in Figure 2(c). However, for condition II (HP), the reaction zone becomes thinner with the increase of pressure. The fraction of the reaction zone with enhanced local equivalence ratio is larger than that at condition I (REF) for a
fixed increased \( \text{Ka} \). Therefore, the influence on mass burning rate caused by Soret diffusion of \( \text{H}_2 \) is more sensitive to the increase of \( \text{Ka} \) at elevated pressure, as shown in Figure 2(b).

Now the effect of Soret diffusion of \( \text{H} \) on mass burning rate at elevated pressure and temperature as a function of \( \text{Ka} \) is explained in the following. Although the influence of Soret diffusion of \( \text{H} \) is not sufficient to change the local equivalence ratio, it affects the reaction rates of the relevant reactions for the \( \text{H} \) radical. Figure 4 shows that the scaled Soret diffusion flux of \( \text{H} \) is positive, indicating that Soret diffusion of \( \text{H} \) moves \( \text{H} \) radicals from the active zone of \( \text{R1} \) to the downstream side of the flame for all the test values of \( \text{Ka} \). This is why the change of the mass fraction of \( \text{H} \) is negative in Figure 5 (right) for \( \text{Ka} = 0 \). Furthermore, Soret diffusion of \( \text{H} \) increases with the increase of \( \text{Ka} \) due to the increase of \( \nabla T/T \) and the mass fraction of \( \text{H} \) (Figure 6). The increments in \( \nabla T/T \) and the mass fraction of \( \text{H} \) result from preferential diffusion effects. This would imply that the mass fraction of \( \text{H} \) in the active zone of \( \text{R1} \) is reduced by the Soret diffusion flux of \( \text{H} \) with the increase of \( \text{Ka} \), leading to a reduction in mass burning rate. However, it is seen in Figure 5 that, for the three conditions, the mass fraction of \( \text{H} \) in the flames increases with the inclusion of Soret diffusion at elevated \( \text{Ka} \). This is because the increment in the mass fraction of \( \text{H} \) caused by the enhancement of local equivalence ratio in the reaction zone is larger than the reduction in the mass fraction of \( \text{H} \) caused by Soret diffusion of \( \text{H} \). At elevated pressure, the scaled Soret diffusion flux of \( \text{H} \) decreases due to the reduction of the mass fraction of \( \text{H} \) in the flame (Figure 4). On the contrary, the Soret diffusion flux of \( \text{H} \) increases at elevated temperature. This is caused by the increase of the mass fraction of \( \text{H} \) in the flame (Figure 4). This explains why the reduction in mass burning rate caused by the Soret diffusion of \( \text{H} \) decreases with pressure and increases with temperature, as seen in Figure 2.

To sum up, the influence of Soret diffusion on the 1D flame mass burning rate at different conditions mainly results from the competing effects of Soret diffusion of \( \text{H}_2 \) and \( \text{H} \). The Soret diffusion of \( \text{H}_2 \) eventually increases the mass burning rate with the increase of \( \text{Ka} \). This is due to the enhancement of the local equivalence ratio in the large part of the reaction zone caused by the Soret diffusion of \( \text{H}_2 \). This effect is enhanced by the increased pressure and suppressed by the increased unburned mixture temperature, which is mainly because of the decrease in reaction zone thickness with pressure and the increase in reaction zone thickness with temperature. However, the Soret diffusion of \( \text{H} \) decreases the mass burning rate with the increase of \( \text{Ka} \). The reason is that the Soret diffusion of \( \text{H} \) transports H radicals out of the active zone of \( \text{R1} \) in all the cases tested. This effect is suppressed by the increased pressure and enhanced by the increased temperature. This is because the mass fraction of \( \text{H} \) in the active zone of \( \text{R1} \) decreases with pressure and increases with temperature.

4.2. 2D flames

In this subsection, the time-dependent evolution of 2D lean hydrogen/air flames with and without Soret diffusion is analysed. Firstly, the flame front evolution and global propagating characteristics are discussed. Secondly, the local flame structures are analysed for the three conditions.

For a thermal-diffusively unstable planar flame, a small perturbation triggers the formation of cellular structures, as shown in Figure 7. The flame front is defined at the position of the normalised temperature (progress variable) \( c = 0.3 \). It is seen that the initially perturbed planar flames at \( x = 1.5 \) cm evolve into cellular flames rapidly, and the leading edges are convex towards the reactants for all the three conditions. It should be noted that, although the flame shapes with adaptive mesh and uniform mesh are different, their dynamic
behaviour is similar. In a statistical sense, the flames are similar. For all the cases, in the early phase of flame front evolution, the initial small perturbation is amplified, which leads to the formation of small scale cellular structures. After a certain period of time, small flame cells merge together forming bigger flame cells. This phenomenon is consistent with previous results [13]. However, for the flames at condition II (HP), at some moments, the flame propagates into the combustion product, which is also reported in a number of papers, such as in [26]. It is caused by the strong coupling effects of hydrodynamic instability and thermal-diffusive instability at elevated pressures. While, for the flames at condition III (HT), the flame fronts with and without Soret diffusion seem to evolve into a stable and nearly planar flame. This phenomenon is observed repeatedly for two extra cases simulated with increased wavenumber (12) and extended domain (2 cm × 2 cm) at condition III (HT). Thus, it is expected that the flame is stabilised by the increased unburned mixture temperature and lower thermal expansion.

In order to quantify the flame front \((c = 0.3)\) evolution further, with and without Soret diffusion, a Fourier analysis is performed. The evolution of amplitudes of different Fourier modes of the instantaneous flame front is presented in Figure 8. The amplitude is scaled by the laminar flame thickness. It is seen in Figure 8 that, at condition I (REF), in the initial period from 0 to 5 ms, the initial wave number \((k_6)\) dominates the evolution of both flames with and without Soret diffusion. The amplitude of \(k_6\) for the flame with Soret diffusion is considerably amplified, while it is just slightly amplified for the flame without Soret diffusion. After 5 ms, the \(k_2\) and \(k_4\) modes, which represent large flame cells, are excited for both flames with and without Soret diffusion. Moreover, the excitation of \(k_2\) and \(k_4\) with Soret diffusion occurs earlier than that without Soret diffusion. Figure 8 shows that at condition II (HP) the evolution of different Fourier modes is more chaotic and changes significantly with time. During the time period from 4 to 6 ms, the amplitudes of \(k_8\) and
$k_9$ with Soret diffusion are larger than those without Soret diffusion. Interestingly, the excitation of $k_1$ and $k_2$ with Soret diffusion takes place earlier than those without Soret diffusion. Figure 8 shows that, at condition III (HT), the amplitudes of the excited modes show less difference between the flames with and without Soret diffusion, as compared with the conditions I (REF) and II (HP).
Figure 9. Instantaneous flame length and global consumption speed for the flames with and without Soret diffusion at the three different conditions. Red Crosses: with Soret diffusion; black circles: without Soret diffusion. The global consumption speed is scaled with the corresponding laminar flame speed. The flame length is scaled with the width of the computational domain. (Colour online)

The change of the global consumption speed depends on the wrinkling of the flame front due to the coupling effects of hydrodynamic instability and thermal-diffusive instability. The global consumption speed is computed using the definition in [10,27], given by

$$S_c^G = \frac{-1}{A_L(\rho Y_{H_2})} \int_{\Omega} \omega_{H_2} \, d\Omega,$$

where $A_L$ is the width of the computational domain, $\rho Y_{H_2}$ is the inlet hydrogen mass density, $\omega_{H_2}$ is the mass consumption rate of molecular hydrogen, and $d\Omega$ is a differential element of area. The control volume $\Omega$ is the entire domain. The instantaneous flame front length is extracted based on the iso-line of $c = 0.3$. Figure 9 shows a scatter plot of scaled global consumption speed versus scaled flame length for the three conditions. The global consumption speed is scaled with the corresponding laminar flame speed. The flame length is scaled with the width of the computational domain. The global consumption speeds for the initial several time steps are omitted due to the initialisation with the approximation for the species profiles. It is observed that the scaled global consumption speed increases with the increase of the scaled flame length for the three conditions. Furthermore, at conditions I (REF) and II (HP), the scaled global consumption speed with Soret diffusion is higher than that without Soret diffusion. However, at condition III (HT), the scaled global consumption speed with Soret diffusion is lower than that without Soret diffusion. This observation is consistent with the results from the 1D flame simulation (Figure 2). Based on the analysis of the 1D flame, it is inferred that the enhancement of global consumption speed with Soret diffusion at conditions I (REF) and II (HP) results from the strong increase in the burning intensity in positively stretched and curved regions caused by the Soret diffusion of H$_2$. The reduction of global consumption speed with Soret diffusion at conditions III (HT) is due to the strong negative influence on burning intensity caused by Soret diffusion of H.

The correlation between local consumption speed and flame curvature is also analysed with and without Soret diffusion. The local consumption speed is computed in the same way as in [27]. It is defined as

$$S_c^\ell = \frac{-1}{A_c(\rho Y_{H_2})} \int_{\Omega_{\text{local}}} \omega_{H_2} \, d\Omega_{\text{local}},$$
where $A_c$ is the length of the flame front inside the control volume. The local control volume is the area between the two adjacent local normals, which are uniformly distributed along the flame front. The local norms follow the gradient of $c$. The flame front for the computation of the local consumption speed is defined at an isothermal line which passed through the peak local $H_2$ consumption rate of the flames. The curvature, $\sigma$, is simply computed throughout the domain by using the formula, $\sigma = -\nabla \cdot \vec{n}$, where $\vec{n}$ is the normal unit vector, defined as $\vec{n} = -\nabla T/\|T\|$. The curvature of the flame front is obtained by interpolation. Based on this definition of flame curvature, the flame curvature is positive in the regions which are convex towards the reactants and negative at cusps. In this analysis, two instantaneous snapshots for each case are taken, namely at 5 and 20 ms, which represent the early phase and developed phase of the simulation, respectively. The local consumption speed versus flame curvature at 5 and 20 ms are plotted in Figure 10, where the local consumption speed is scaled with the corresponding zero-stretch laminar flame speed. It is shown that Soret diffusion affects the positive correlation between the local consumption speed and the flame curvature. The local consumption speed with the inclusion of Soret diffusion is more sensitive to flame curvature than that without Soret diffusion at conditions I (REF) and II (HP), reflecting a more negative Markstein number indicative of a more thermal-diffusively unstable flame. At the three conditions, flames predicted with Soret diffusion at 5 ms exhibit a more pronounced reduction in burning intensity in cusps than those at 20 ms when compared to their counterparts without Soret diffusion. This is because the flames at 5 ms are sharper and more so when Soret diffusion is included. At conditions I (REF) and II (HP), the maximum local consumption speed predicted with and without Soret diffusion at 5 ms shows larger difference than that at 20 ms. The difference is even...
larger at condition II (HP) than that at condition I (REF). However, at condition III (HT), Soret diffusion has only a small influence on the maximum local consumption speed at both 5 and 20 ms. Additionally, a larger amount of local extinction is observed at condition II (HP). The effect of Soret diffusion significantly enhances thermal-diffusive instability at elevated pressure. This is because the Markstein number decreases, indicative of a more thermal-diffusively unstable flame at elevated pressure, as shown in Figure 1. However, the enhancement in thermal-diffusive instability caused by Soret diffusion is suppressed at elevated temperature, owing to the larger Markstein number at elevated temperature.

To sum up, the influence of Soret diffusion on 2D freely propagating flame evolution depends significantly on the pressure and temperature. In the early phase of flame evolution, at normal pressure and temperature, the flames computed with Soret diffusion display more curved flame cells than those without Soret diffusion. This effect is enhanced by pressure while it is suppressed by temperature. The global consumption speed is enhanced by Soret diffusion at normal and elevated pressures. This is because the burning intensity in positively stretched and curved regions is strongly increased by Soret diffusion of H₂. However, the global consumption speed is reduced by Soret diffusion at elevated temperature. This is due to the negative influence on burning intensity caused by the Soret diffusion of H. The correlation between local consumption speed and flame curvature indicates that a more thermal-diffusively unstable flame is predicted with the inclusion of Soret diffusion at normal and elevated pressures. However, this is not the case at elevated temperature.

5. Conclusions

The influence of Soret diffusion on lean hydrogen/air flames at normal and elevated pressure and temperature has been systematically investigated. Firstly, the effect of Soret diffusion on the mass burning rate as a function of Ka is studied by the simulation of 1D flames. It is found that, at normal pressure and temperature, Soret diffusion first reduces the mass burning rate and then enhances the mass burning rate with the increase of Ka. This results from the competing effects of Soret diffusion of H and H₂. Soret diffusion of H₂ first reduces and then enhances the mass burning rate with the increase of Ka. This effect is enhanced by the increased pressure and suppressed by the increased unburned mixture temperature. However, Soret diffusion of H decreases the mass burning rate continuously with the increase of Ka. This effect is suppressed by the increased pressure and enhanced by the increased temperature.

Secondly, the dynamics of 2D lean hydrogen/air flames is analysed. It is found that, at normal pressure and temperature, in the early phase of flame evolution, the flames computed with Soret diffusion display more curved flame cells and for these flames the excitation of Fourier modes representing larger flame cells is slightly easier than those without Soret diffusion. Pressure enhances this effect while temperature reduces it. The influence of Soret diffusion of H₂ on global consumption speed is enhanced by elevated pressure. The influence of Soret diffusion of H on global consumption speed is enhanced by elevated temperature. Soret diffusion affects the flame structure and dynamics more in its early phase of evolution than in the long term phase due to the local enrichment of H₂ caused by flame curvature effects. The numerical results demonstrate that it is important to take Soret diffusion into consideration when hydrogen/air flames are computed at elevated pressures. However, this is not the case at elevated temperature. Future research will be conducted on the influence of Soret diffusion on the ignition of hydrogen/air flame at elevated pressure and temperature.
Acknowledgements
The authors thank Professor Clinton Groth for providing access to the Computational Framework for Fluids and Combustion (CFFC) code.

Funding
The financial support is gratefully acknowledged of the Dutch Technology Foundation (STW) [Project 13549].

Disclosure statement
No potential conflict of interest was reported by the authors.

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