Effects of Soret diffusion on the laminar flame speed and Markstein length of syngas/air mixtures

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Abstract

The effects of Soret diffusion on premixed syngas/air flames at normal and elevated temperatures and pressures are investigated numerically including detailed chemistry and transport. The emphasis is placed on assessing and interpreting the influence of Soret diffusion on the unstretched and stretched laminar flame speed and Markstein length of syngas/air mixtures. The laminar flame speed and Markstein length are obtained by simulating the unstretched planar flame and positively-stretched spherical flame, respectively. The results indicate that at atmospheric pressure the laminar flame speed of syngas/air is mainly reduced by Soret diffusion of H radical while the influence of H_2 Soret diffusion is negligible. This is due to the facts that the main reaction zone and the Soret diffusion for H radical (H_2) are strongly (weakly) coupled, and that Soret diffusion reduces the H concentration in the reaction zone. Because of the enhancement in the Soret diffusion flux of H radical, the influence of Soret diffusion on the laminar burning flux increases with the initial temperature and pressure. Unlike the results at atmospheric pressure, at elevated pressures the laminar flame speed is shown to be affected by the Soret diffusion of H_2 as well as H radical. For stretched spherical flame, it is shown that the Soret diffusion of both H and H_2 should be included so that the stretched flame speed can be accurately predicted. Similar to the laminar flame speed, the Markstein length is also reduced by Soret diffusion. However, the reduction is found to be mainly caused by Soret diffusion of H_2 rather than that of H radical. Moreover, the influence of Soret diffusion on the Markstein length is demonstrated to decrease with the initial temperature and pressure.
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1. Introduction

Mass diffusion is essential in laminar flames and it strongly affects, or even determines, the fundamental combustion processes such as ignition, flame propagation, and extinction [1,2]. The dominant mode of mass transport is the Fickian diffusion, which is due to the appearance of concentration gradient. In the modeling of multi-component

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reactive flow, usually only the Fickian diffusion is considered. However, in the presence of very light or heavy species and steep temperature gradient, Soret diffusion [1–3], which drives light (heavy) species toward (away from) the hot zone, is also important for the mass transport.

The effects of Soret diffusion on laminar flames have been extensively studied in the literature. Except the work on heavy species such as metal particle [4], soot [5], and *n*-heptane [6], most of the studies are focused on the Soret diffusion of light species. For examples, Ern and Giovangigli [7,8] studied the influence of Soret diffusion on the flame structure and extinction of hydrogen/air and methane/air mixtures; Bongers and De Goey [9] investigated the effects of Soret diffusion on the laminar flame speed of hydrogen/air and methane/air mixtures; Grcar et al. [10] examined the Soret effects in lean hydrogen/air premixed flames; and Yang et al. [11,12] presented a mechanistic interpretation on Soret diffusion in hydrogen/air and *n*-butane/air flames. In these studies [4–12], discernable effects of Soret diffusion were observed.

However, to our knowledge, in the literature there is no study investigating the effects of Soret diffusion on syngas/air flames. Currently, syngas (synthesis gas) is expected to play an important role in future energy production, particularly for stationary power generation using Integrated Gasification Combined Cycle (IGCC) systems [13]. However, there are gaps in the fundamental understanding of syngas combustion characteristics, especially at elevated temperatures and pressures that are relevant to practical combustors. Syngas is primarily composed of H_2 and CO, and usually it has high concentration of light species, hydrogen [13]. Therefore, it is expected that Soret diffusion will affect syngas/air flames. Previous studies [4–12] only examined the Soret diffusion at normal temperature and pressure. Therefore, the influence of Soret diffusion at high temperatures and pressures remains unknown. Since syngas combustion in gas-turbine occurs at high temperatures and pressures, it is of interests to understand the effects of Soret diffusion at elevated temperatures and pressures as well as at normal conditions.

The objective of this study is to computationally assess and interpret the effects of Soret diffusion on syngas/air flames at normal and elevated temperatures and pressures. Two most important parameters of premixed syngas/air flames are investigated: one is the laminar flame speed (or laminar burning flux) and the other is the Markstein length. The laminar flame speed is defined as the propagation speed of an adiabatic planar flame relative to the unburned gas [1,2] and it affects the fuel burning rate in combustion engines. The Markstein length characterizes the variation in the local flame speed due to the influence of external stretching [14]. It determines the

flame instability related to preferential diffusion and is one of the basic input physicochemical parameters in certain turbulent combustion models [14]. In the next section, the problem specification and numerical methods are presented. Then in Section 3, the effects of Soret diffusion on the laminar flame speed and Markstein length of syngas/air mixtures are discussed. Finally, the conclusions are given in Section 4.

2. Problem specification and numerical methods

Two premixed flame configurations are considered in this study: one is the unstretched freely-propagating planar flame and the other is the positively-stretched propagating spherical flame. The laminar flame speeds of syngas/air mixtures at different conditions (equivalence ratio, temperature, and pressure) with and without Soret diffusion are obtained by simulating the one-dimensional propagating planar flame using the CHEMKIN–PREMIX code [15]. In all simulations, the detailed chemical mechanism for syngas oxidation developed by Davis et al. [16] is used and it consists of 13 species and 38 elementary reactions. The number of grid points is always kept to be above 800 so that the flame structure is well resolved and the results are grid-independent.

In order to get the Markstein length of syngas/air mixtures, the one-dimensional expanding premixed spherical flame is simulated using the in-house code A-SURF [17,18]. A-SURF solves the conservation equations of one-dimensional, compressible, multi-component, reactive flow in a spherical coordinate using the finite volume method [17,18]. The CHEMKIN packages [19] are incorporated into A-SURF to calculate the temperature- and component-dependent thermodynamic and transport properties as well as the reaction rates based on the detailed chemistry of Davis et al. [16]. A-SURF has been successfully validated and used in previous studies [17,18,20,21]. Details on the governing equations, numerical schemes, and code validation can be found in Refs. [17,18] and thus are not repeated here. The computational domain is set to be $0 \leqslant r \leqslant 50$ cm and a multi-level, dynamically adaptive mesh is used. The propagating spherical flame is initiated by a small hot pocket (1–2 mm in radius) of burned product surrounded by static fresh mixture at initially specified temperature and pressure.

3. Results and discussion

3.1. Effects of Soret diffusion on unstretched laminar flame speed

In order to assess the effects of Soret diffusion on the laminar flame speed of syngas/air mixtures,

we compare the laminar flame speeds evaluated with and without including Soret diffusion of all species. The results are shown in Fig. 1. It is seen that activating Soret diffusion leads to a reduction in the laminar flame speed. The same observation is found in previous studies on hydrogen/air and methane/air mixtures [7–9,11] and the mechanistic interpretation on the reduction of laminar flame speed by Soret diffusion will be provided later. According to Fig. 1, the influence of Soret diffusion on the laminar flame speed increases with the equivalence ratio and hydrogen content in syngas. This is reasonable since the influence of Soret diffusion is mainly caused by the light species, H_2 and H [11].

Soret diffusion is resulted from temperature gradient, and the steepness of the temperature distribution in a premixed planar flame is known to be strongly affected by the initial temperature and pressure. Therefore, the influence of Soret diffusion is expected to depend on the initial temperature and pressure. In the following we only focus on the stoichiometric syngas/air mixture with equal amounts of H_2 and CO (i.e. H_2:CO = 50%:50% vol.). The laminar flame speeds at different temperatures and pressures are presented in Fig. 2(a). The inclusion of Soret diffusion is shown to always reduce the laminar flame speed. Regarding to the initial temperature effect, it is observed that the influence of Soret diffusion on the laminar flame speed monotonically increases with the initial temperature. However, different from the initial temperature, the increase in pressure is shown to reduce the influence of Soret diffusion: at atmospheric pressure (1 atm), the reduction of the laminar flame speed by Soret diffusion is in the range of 10–20 cm/s, while at P = 25 atm, it is in the range of 2–4 cm/s. Noticing that for the laminar planar flame propagation the eigenvalue is the laminar burning flux ($m = \rho_u S_u$, where ρ_u is the unburned gas density) instead of the laminar
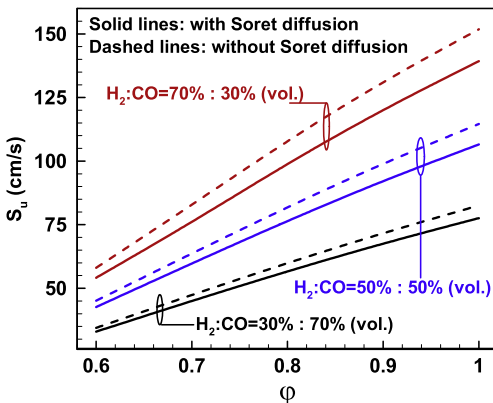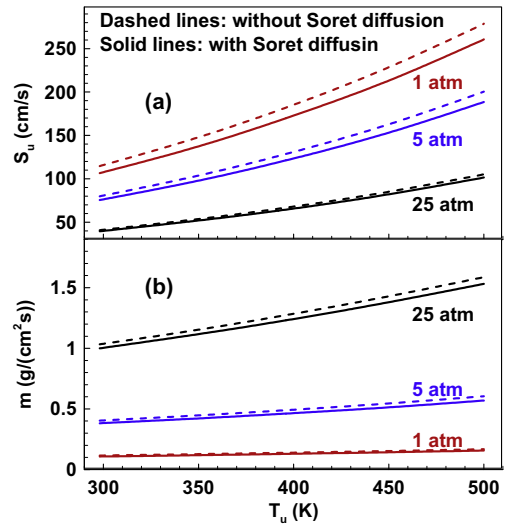
Fig. 2. The influence of Soret diffusion on the (a) laminar flame speed and (b) laminar burning flux of stoichiometric syngas/air mixture at different temperatures and pressures (H_2:CO = 50%:50% vol.).

flame speed (S_u) [2], we plot the laminar burning fluxes at different initial temperatures and pressures in Fig. 2(b). It is seen that the influence of Soret diffusion on the laminar burning flux increases with the pressure. Therefore, the results in Fig. 2 demonstrate that the influence of Soret diffusion depends on the initial temperature and pressure.

To further demonstrate the effects of the initial temperature and pressure, we plot the reduction in laminar burning flux (δm) caused by including Soret diffusion of different species in Fig. 3. It is seen that the influence of Soret diffusion is mainly caused by H_2 and H. At P = 1 atm, Fig. 3 shows that the reduction caused by H Soret diffusion dominates over that by H_2 Soret diffusion. However, with the increase of the pressure, the reduction caused by H_2 Soret diffusion monotonically increases while that caused by H Soret diffusion changes non-monotonically. Therefore, unlike the observation at atmospheric pressure [11], at elevated pressures the laminar burning flux is affected by the Soret diffusion of H_2 as well as H. Figure 3(b) indicates that at normal and elevated temperatures, the reduction in laminar burning flux slightly increases with the initial temperature. (The relative reduction in the laminar flame speed or burning flux is found to change slightly with the initial temperature and decrease with the pressure.) In the following we shall provide a mechanistic explanation on this observation.

Before interpreting the effects of the initial temperature and pressure, we need first understand how Soret diffusion affects the laminar flame speed of syngas/air mixture at normal temperature and
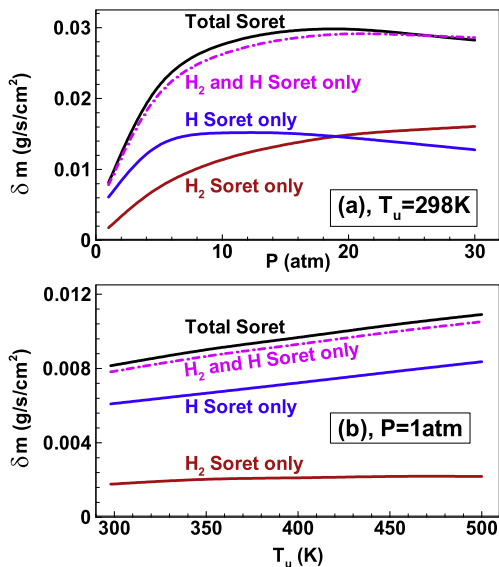
Fig. 1. The influence of Soret diffusion on the laminar flame speed of syngas/air mixtures at T_u = 298 K and P = 1.0 atm (φ is the equivalence ratio).

Fig. 3. The reduction in laminar burning flux due to Soret diffusion at different (a) pressures and (b) initial temperatures.
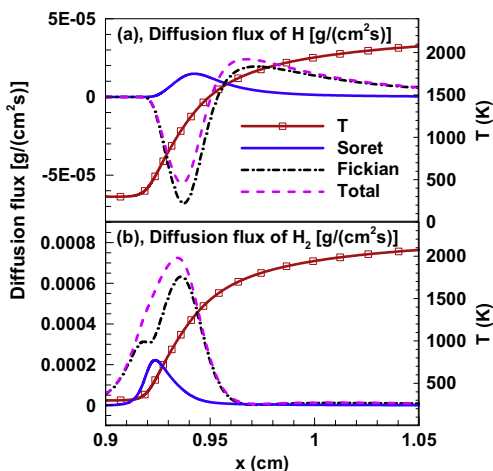

Fig. 4. Fickian and Soret diffusion fluxes of (a) H and (b) H_2 in the laminar premixed flame of stoichiometric syngas/air mixture at $T_u = 298$ K and $P = 1.0$ atm.

pressure (298 K and 1 atm). Using the same strategy of Yang et al. [11] (to separately suppress or activate Soret diffusion of individual species in simulation), we find that at $P = 1$ atm the change in the laminar flame speed of syngas/air mixture is mainly caused by Soret diffusion of H radical, and that the influence of H_2 Soret diffusion is much smaller (also demonstrated by Fig. 3(b)). To interpret this phenomenon, we present the Fickian and Soret diffusion fluxes of H and H_2 in Fig. 4. The temperature profiles are superimposed for reference. For H radical, it is seen that the Fickian diffusion is negative first and then becomes positive, and that the Soret diffusion flux mainly spread in the interval of negative Fickian diffusion flux. Unlike Fickian diffusion, Soret diffusion makes more H radical transport to the high temperature region (downstream), leading to the reduction of the H radical concentration in the reaction zone. However, for H_2, Fig. 4(b) shows that both the Fickian diffusion and Soret diffusion move H_2 downstream.

In the unstretched planar flame, enthalpy conservation is maintained and the adiabatic flame temperature is not affected by Soret diffusion. Therefore, the influence of Soret diffusion on the laminar flame speed is mainly due to the resulting change in the chemical reaction rates. In the oxidation of syngas, the main reactions related to H and H_2 are the chain branching and chain propagation reactions: $H + O_2 = OH + O$ (R1), $H_2 + O = H + OH$ (R2), and $H_2 + OH = H + H_2O$ (R3) [2]. The rates of these reactions together with the reaction $H + O_2 + M = HO_2 + M$ (R12) are

plotted in Fig. 5. It is seen that they are obviously affected by Soret diffusion. Since it is the coupling between Soret diffusion and major reactions that leads to the reduction of the laminar flame speed, we demonstrate the coupling in Fig. 6. In order to illustrate the major reaction zone, we choose (R1) and (R3) as the major relevant reactions for H radical and H_2, respectively. At the temperature region between two vertical lines shown in Fig. 6, the reaction rate is above 50% of its peak value and this region is referred to as the main reaction zone. It is seen that for H radical, the peak Soret diffusion flux occurs in the main reaction zone, while for H_2, the Soret diffusion flux is biased towards the upstream of main reaction zones. Since the main reaction zone for (R2) is on the right side of that for (R3) as shown in Fig. 5, the Soret diffusion flux of H_2 is decoupled with the main reaction zones of H_2-relevant reactions (R2 and R3). Consequently,
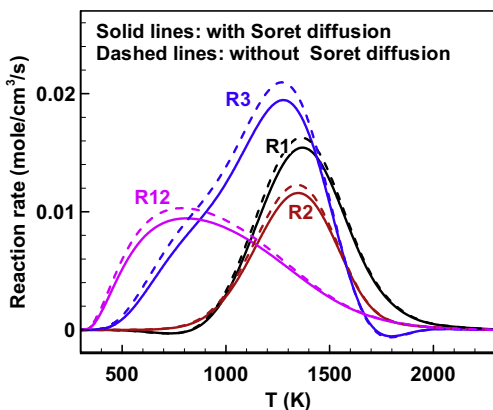

Fig. 5. Effects of Soret diffusion on the reaction rates of major reactions ($T_u = 298$ K and $P = 1.0$ atm).
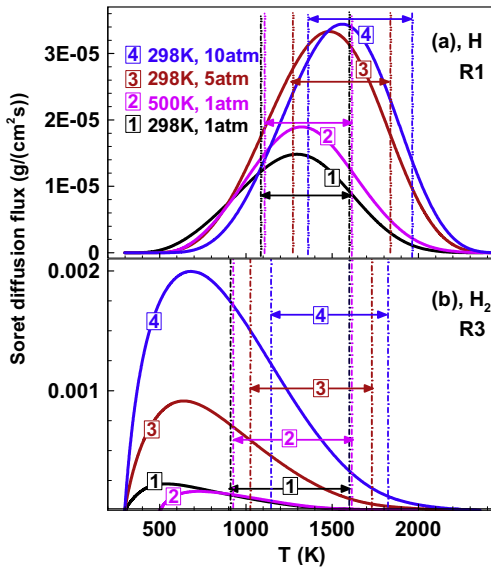
Fig. 6. Coupling between Soret diffusion and chemical reaction. The solid lines represent the Soret diffusion flux of H or H$_2$. At the temperature region denoted by the arrowed-lines between two vertical dash-dotted lines, the reaction rate of (R1) or (R3) is above 50% of its peak value.

Fig. 7. Effects of initial temperature (a) and pressure (b) on the "driving force" and mole fraction of H radical.

the influence of H$_2$ Soret diffusion on the laminar flame speed of syngas/air is negligible. Different from H$_2$, the H Soret diffusion is demonstrated by Fig. 6(a) to be strongly coupled with the main reaction zone of the chain branching reaction (R1). Therefore, the coupling shown in Fig. 6 for $P = 1$ atm basically explains why the laminar flame speed of syngas/air mixtures is mainly affected by Soret diffusion of H radical.

The effects of initial temperature and pressure can also be interpreted by the coupling between Soret diffusion flux and major reaction rates. With the increase of the initial temperature (from 298 K to 500 K), Fig. 6 shows that the Soret diffusion fluxes of both H and H$_2$ become larger (explained later in Fig. 7(a)) while the coupling remains unchanged. Therefore, at higher temperature, the laminar burning flux, $m = \rho_u S_u$, is more strongly influenced by Soret diffusion (see Figs. 2(b) and 3(b)). With the change of the pressure from 1 atm to 5 atm, Fig. 6 shows that the Soret diffusion fluxes of both H and H$_2$ increase greatly (explained later in Fig. 7b). Therefore, the reduction in laminar burning flux (δm) caused by H Soret diffusion and that caused by H$_2$ Soret diffusion both increase with the pressure, which is consistent with results in Fig. 3(a). With the increase of the pressure from 5 atm to 10 atm, Fig. 6 shows that the Soret diffusion flux of H radical changes slightly while that of H$_2$ increases significantly. Consequently, similar changes (as observed in
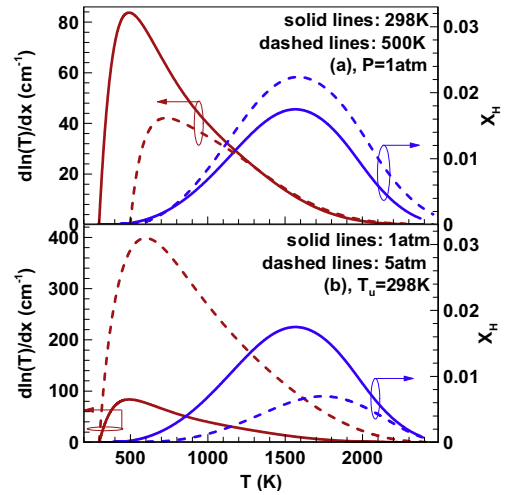
Fig. 3(a)) happen to the reduction in laminar burning flux caused by H Soret diffusion and that caused by H$_2$ Soret diffusion. Moreover, Fig. 6 shows that the increase in the Soret diffusion caused by increasing the initial temperature is much less than that by increasing the pressure. This explains the dependence of the reduction in laminar burning flux on the initial temperature and pressure shown in Fig. 3.

Now we explain why the Soret diffusion flux of H radical increases with the initial temperature and pressure, which is observed in Fig. 6(a). According to transport theory [1–3], Soret diffusion flux is proportional to the temperature gradient normalized by temperature itself, grad T/T (it is $d\ln(T)/dx$ for one-dimensional case), which is referred to as the "driving force" of Soret diffusion [11,12]. Only in the region where the "driving force" and concentrations of light species are both large, is the Soret diffusion flux non-negligible. Figure 7 shows the effects of initial temperature and pressure on the "driving force" as well as the mole fraction of H radical. According to Fig. 6(a), the Soret diffusion flux of H radical peaks around 1200–1400 K. Near this temperature region, Fig. 7(a) demonstrates that the "driving force" is almost unaffected by the initial temperature. Since the mole fraction of H radical increases with the initial temperature (as shown in Fig. 7(a)), the Soret diffusion flux of H radical should also increase with the initial temperature. Therefore, the increase of the H Soret diffusion flux with the initial temperature shown in Fig. 6(a) is caused by the increased amount of H radical in the reaction zone. In fact, Fig. 7(a) shows that the "driving force" decreases with the initial temperature at the low temperature regime ($T < 1000$ K) in which the Soret diffusion flux is consequently reduced (see

Fig. 6(a)). Though steeper temperature gradient occurs at higher initial temperature, the driving force, $d\ln(T)/dx$, actually becomes smaller. This is because the increase in T is greater than the increase in dT/dx when the initial temperature is increased from 298 K to 500 K. At an elevated pressure (5 atm), Fig. 7(b) shows that the "driving force" is greatly improved while the mole fraction of H radical is reduced. In the temperature region of 1300–1500 K, the improvement in the "driving force" is shown to be much larger than the reduction in the mole fraction of H radical. As a result, the H Soret diffusion flux also increases with the pressure, as shown in Fig. 6(a).

3.2. Effects of Soret diffusion on stretched flame speed and Markstein length

Unlike the unstretched premixed planar flame, in practical combustion process of sygnas, the flame propagation speed and stability are strongly affected by stretch rate. In the following we shall examine the influence of Soret diffusion on stretched flame speed and Markstein length of syngas/air mixtures.

Figure 8 shows the change of the flame propagation speed, S_b, with the flame radius, R. When the compression-induced flow [22] is negligible, the stretched flame speed with respect to the burned gas is $S_b = dR/dt$ (the flame front, R, is defined in simulation as the position where the maximum heat release rate occurs). The results in Fig. 8 demonstrate that the Soret diffusion of CO has little influence while the Soret diffusion of H and H_2 strongly affects the stretched flame speed. For an outwardly propagating spherical flame, the stretch rate is $K = 2R^{-1}dR/dt = 2S_b/R$ [2] and thereby it is inversely proportional to the flame radius. At large flame radius ($R > 3$ cm in Fig. 8 and $K < 500$ s^{-1} in Fig. 9), the stretch effect

on the flame propagation speed is relatively small and hence the influence of Soret diffusion is similar to that on the unstretched planar flame. At $R > 3$ cm, Fig. 8 shows that the Soret diffusion of H_2/H has weak/strong influence on the flame speed. These results are consistent with those on the unstretched laminar flame speed discussed in the previous subsection.

At small flame radius (thus large stretch rate) of $0.5 < R < 1.5$ cm, Fig. 8 indicates that the flame propagation speed is strongly affected by the Soret diffusion of H_2 as well as H. Similar to the unstretched planar flame at high pressures, for stretched spherical flame the Soret diffusion of both H and H_2 should be included so that the stretched flame speed can be accurately predicted. It is well known that for stretched spherical flame, the enthalpy conservation is not maintained due to the coupling between the positive stretch rate and preferential diffusion of heat and species [2]. Since Soret diffusion modifies the diffusion flux of H_2 as well as H (see Fig. 4), the influence of Soret diffusion is greatly promoted by the stretch rate, especially at small flame radius (large stretch rate).

For moderately stretched spherical flame, there is a linear relationship between S_b and K [14]:

$$S_b = S_b^0 - L_b K \tag{1}$$

where S_b^0 is the flame propagation speed at zero stretch rate. According to Eq. (1), the influence of Soret diffusion on S_b shown in Fig. 8 can be interpreted by examining how Soret diffusion affects L_b. The Markstein length L_b can be obtained from the linear extrapolation between S_b and K according to Eq. (1). The flame radius range used for linear extrapolation is from 1.0 cm to 2.5 cm so that the effects of ignition [17], compression [22], and nonlinear behavior
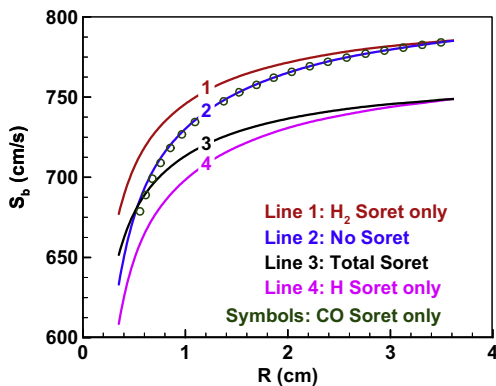

Fig. 8. The influence of Soret diffusion on the change of the spherical flame propagation speed with the flame radius for stoichiometric syngas/air mixture (H_2:CO = 50%:50% vol., T_u = 298 K and P = 1.0 atm).
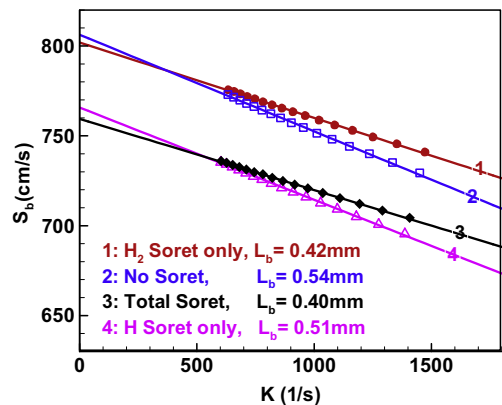

Fig. 9. The influence of Soret diffusion on the change of the spherical flame propagation speed with the stretch rate (symbols are data in the range of $1.0 \leqslant R \leqslant 2.5$ cm, lines denote linear fitting).

[23–25] are minimized. Figure 9 demonstrates the effects of Soret diffusion of different species on the change of S_b with K. It is seen that at large stretch rate (small flame radius), the Soret diffusion of H and that of H_2 both affect the stretched flame speed. As mentioned before, at small stretch rate (say $K < 500\ s^{-1}$), the inclusion of H_2 Soret diffusion is shown to only slightly affect S_b, while Soret diffusion of H radical affects S_b much more pronounced. However, contrary to the laminar flame speed at small or zero stretch rate, the Markstein length (its value is shown in Fig. 9) is only slightly affected by activating H Soret diffusion while strongly affected by allowing for Soret diffusion of H_2 (the relative reduction in Markstein length is $1 - 0.40/0.54 = 26\%$). It is demonstrated in Fig. 9 that only considering H_2 Soret diffusion leads to nearly the same amount of reduction in Markstein length as that by allowing for Soret diffusion of all species, and that the Markstein length without Soret diffusion of all species is close to that of including only H Soret diffusion. Therefore, the reduction of Markstein length is mainly caused by the Soret diffusion of H_2 rather than that of H radical.

The influence of Soret diffusion on Markstein length shown in Fig. 9 is mainly due to the dependence of the Markstein length on the Lewis number. As shown in Fig. 4(b), the inclusion of H_2 Soret diffusion promotes the mass diffusion of H_2 and thereby effectively increases the mass diffusivity of H_2. As a result, the Lewis number of H_2 (inversely proportional to mass diffusivity) is reduced when Soret diffusion of H_2 is activated. Since the Markstein length decreases monotonically with the decrease of the Lewis number [2,14], the Markstein length is reduced by allowing for H_2 Soret diffusion. Since the Soret diffusion flux of H is much less than that of H_2 (as shown in Figs. 4 and 6), the influence of Soret diffusion on the preferential diffusion between heat and H radical in a stretched flame is negligible. Consequently, for Markstein length, the Soret diffusion of H_2 plays a dominant role, while the influence of H Soret diffusion is negligible. This observation is also consistent with the theory in Ref. [26] stating that the Markstein length is mainly affected by the fuel Lewis number rather than the radical Lewis number.

We also investigate the influence of Soret diffusion on the Markstein length of syngas/air mixtures at elevated temperatures and pressures. The results are plotted in Fig. 10. Unlike the laminar flame speed/burning flux shown in Fig. 2, the influence of Soret diffusion on the Markstein length is shown to decrease with the initial temperature and pressure. Compared with results at normal temperature ($T_u = 298$ K), at higher initial temperature ($T_u = 500$ K) the relative increase in the mass diffusivity of H_2 due to Soret diffusion becomes smaller, resulting in less reduction in
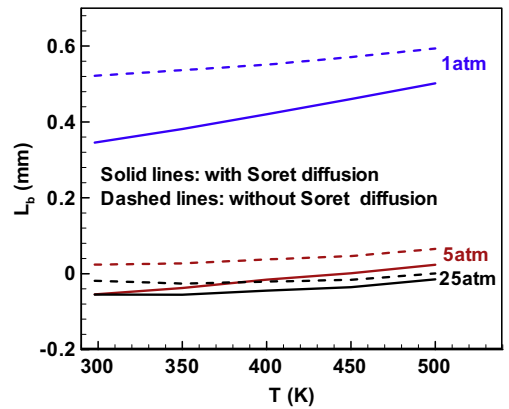

Fig. 10. The influence of Soret diffusion on the Markstein length of stoichiometric syngas/air mixture at different temperatures and pressures.

the Lewis number of H_2. Therefore, as shown in Fig. 10, the reduction in Markstein length caused by activating Soret diffusion decreases with the initial temperature. Since the Markstein length is proportional to the flame thickness which is inversely proportional to the pressure, the Markstein length decreases greatly with the pressure [2]. As a result, the reduction in Markstein length caused by activating Soret diffusion also decreases with the pressure.

4. Conclusions

The effects of Soret diffusion on the laminar flame speed and Markstein length of syngas/air mixtures at normal and elevated temperatures and pressures are studied numerically. It is found that Soret diffusion quantitatively influences the propagation of premixed syngas/air flame. Moreover, the influence is shown to be strongly affected by the initial temperature and pressure.

For the propagation of unstretched planar flame, the laminar flame speed and laminar burning flux of syngas/air mixture are found to be reduced by the inclusion of Soret diffusion. At atmospheric pressure, the reduction in laminar burning flux is mainly caused by the Soret diffusion of H radical rather than H_2. This is because the main reaction zone and the Soret diffusion of H (H_2) are strongly (weakly) coupled. With the increase of the initial temperature and pressure, the Soret diffusion flux of H radical is promoted and the coupling between H Soret diffusion and main chemical reaction remains. Consequently, the influence of Soret diffusion on the laminar burning flux increases with both the initial temperature and pressure. However, with the increase of the pressure, the reduction caused by H_2 Soret diffusion monotonically increases while that

caused by H Soret diffusion changes non-monotonically. Unlike the case at atmospheric pressure, the laminar burning flux at elevated pressures is greatly affected by the Soret diffusion of H_2 as well as H radical.

For the propagation of positively-stretch spherical flame, it is shown that the stretched flame speed is strongly affected by the Soret diffusion of H_2 as well as H, and that the influence of Soret diffusion is promoted by the positive stretch rate especially at small flame radius. Similar to the laminar flame speed, the Markstein length is also reduced by Soret diffusion. However, the reduction is mainly caused by Soret diffusion of H_2 rather than that of H radical. This is due to the facts that the mass diffusivity is increased (thus the Lewis number is reduced) by the inclusion of the Soret diffusion of H_2 and that the Markstein length decreases monotonically with the decrease of the Lewis number. Furthermore, the influence of Soret diffusion is demonstrated to decrease with the initial temperature and pressure.

Acknowledgement

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