Modeling of Lewis number dependence of scalar dissipation rate transport for Large Eddy Simulations of turbulent premixed combustion

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ABSTRACT

The influences of differential diffusion of heat and mass on the Favre-filtered scalar dissipation rate (SDR) transport have been analyzed and modeled using a priori analysis of Direct Numerical Simulations (DNS) data of freely propagating statistically planar turbulent premixed flames with different values of global Lewis number, Le. The DNS data has been explicitly filtered using a Gaussian filter to obtain the unclosed terms of the Favre-filtered SDR transport equation, arising from turbulent transport (T1), density variation due to heat release (T2), strain rate contribution due to the alignment of scalar and velocity gradients (T3), correlation between the gradients of reaction rate and reaction progress variable (T4), molecular dissipation of SDR (D2), and diffusivity gradients f(D). The statistical behaviors of these terms and their scaling estimates reported in a recent analysis have been utilized here to propose models for these unclosed terms in the context of Large Eddy Simulations (LES) and the performances of these models have been assessed using the values obtained from explicitly filtered DNS data. These newly proposed models are found to satisfactorily predict both the qualitative and quantitative behaviors of these unclosed terms for a range of filter widths \( \Delta \) for all Le cases considered here.

1. Introduction

Lean premixed combustion has been identified as one of the possible ways to reduce pollutant emission from gasoline engines and industrial gas turbines [1]. Lean hydrogen and hydrogen-blended hydrocarbon combustion has the potential to attenuate pollutants and greenhouse gas emissions [2, 3]. However, the flames with abundance of fast diffusing species such as hydrogen either in molecular or in atomic form give rise to a significant level of differential diffusion of heat and mass. The differential diffusion of heat and mass can be characterized by a nondimensional number known as the Lewis number \( Le \), which is defined as the ratio of thermal diffusivity \( \alpha_T \) to mass diffusivity \( D \) (i.e., \( Le = \alpha_T / D \)). In actual premixed combustion it is often not straightforward to assign a single global Lewis number in the presence of several species with different Lewis numbers. Often the Lewis number of the deficient species is considered to be the global Lewis number [4], whereas Law and Kwon [5] proposed a methodology of evaluating the effective Lewis number based on heat release measurements. More recently Dinkelacker et al. [6] proposed an algebraic expression for the effective Lewis number based on mole fractions of major species. A number of previous analyses concentrated on the effects of global Lewis number on different aspects of premixed combustion in isolation [7–28] and the same approach has been adopted here.
Modeling of the differential diffusion arising from non-unity global Lewis number remains pivotal to high-fidelity engineering simulations, which are likely to play important roles in the development of new-generation combustors using either hydrogen or hydrogen-blended fuels. Prediction of the micro-mixing rate of hot products and cold unburned gas plays a key role in the modeling of turbulent reacting flows and a quantity known as the scalar dissipation rate (SDR) characterizes this micro-mixing rate [29, 30–32]. Furthermore, the Favre-mean value of SDR of reaction progress variable $c$ in premixed turbulent flames can be related to the mean reaction rate in the context of Reynolds Averaged Navier Stokes (RANS) simulations [23, 33–35]. The instantaneous SDR of reaction progress variable is defined as [23, 33–38]

$$N_c = D \nabla c \cdot \nabla c$$  \hspace{1cm} (1)

where $D$ is the diffusivity of reaction progress variable $c$. Recent analyses have demonstrated [36–38] that the SDR-based reaction rate closure for RANS can also be used for the modeling of the filtered reaction rate $\bar{w}$ based on the Favre-filtered SDR of a reaction progress variable (i.e., $\bar{N}_c = \rho D \nabla \bar{c} \cdot \nabla \bar{c} / \bar{\rho}$) in the

### Nomenclature

- $c$: reaction progress variable
- $c_m$: thermo-chemical parameter
- $C_p$: specific heat at constant pressure
- $C_V$: specific heat at constant volume
- $C_F$: model parameter
- $C_3, C_4$: model parameters
- $D$: progress variable diffusivity
- $D_1$: eddy diffusivity
- $D_a$: Damköhler number
- $D_t$: molecular diffusivity term
- $D_2$: molecular dissipation term
- $f_b$: burning mode probability density function
- $f_{f_{15}, f_{f_{13}, f_{fT}}}$: model parameters
- $f(D)$: term due to diffusivity gradient
- $K_e$: thermo-chemical parameter
- $K_a$: Karlovitz number
- $L_e$: Lewis number
- $l$: integral length scale
- $M_a$: Mach number
- $M_i$: $i$th component of resolved flame normal
- $N_c$: scalar dissipation rate
- $N_i$: $i$th component of flame normal
- $p$: model parameter
- $Pr$: Prandtl number
- $Q$: general quantity
- $R_e_i$: turbulent Reynolds number
- $R_e_a$: sub-grid Reynolds number
- $S_L$: unstrained laminar burning velocity
- $t$: time
- $t_e$: chemical time scale
- $t_f$: initial turbulent eddy turnover time
- $t_{sim}$: simulation time
- $T$: instantaneous dimensional temperature
- $T^*$: non-dimensional temperature
- $T_{ad}$: adiabatic flame temperature
- $T_0$: reactant temperature
- $T_1, T_2, T_3, T_4$: terms in the transport equation of Favre-filtered scalar dissipation rate

### Subscripts

- $0$: unburned gas value
- $\infty$: burned gas value
- $res$: resolved scale value
- $sg$: sub-grid scale value

### Acronyms

- DNS: direct numerical simulation
- LES: large eddy simulation
- pdf: probability density function
- SDR: scalar dissipation rate
context of Large Eddy Simulation (LES) in the following manner when the filter size \( \Delta \) remains greater than the thermal flame thickness \( \delta_{th} \): 

\[
\delta_{th} = \frac{(T_{ad} - T_0)}{\text{Max}} \frac{\nabla T}{L},
\]

where \( T_{ad} \), \( T_0 \), and \( T \) are the adiabatic flame, unburned gas, and instantaneous temperatures, respectively:

\[
\bar{\bar{\nu}} = \frac{2\rho \bar{N}_c}{(2c_m - 1)} \quad \text{with} \quad c_m = \frac{1}{\int_0^1 [\bar{\nu}]_{L} f_{b}(c) dc}{\int_0^1 [\bar{\bar{\nu}}]_{L} f_{b}(c) dc}
\]

where \( \rho \) is the density and \( \bar{\bar{\nu}} = \bar{\rho} Q / \bar{\bar{\nu}} \) is the Favre-filtered value of a quantity \( Q \) with the over-bar indicating an LES filtering operation. In Eq. (2) \( f_b(c) \) is the reacting mode probability density function (pdf) of \( c \) and the subscript “L” refers to the planar laminar flame conditions. By assuming \( f_b(c) \) as a smooth function, regardless of the exact form, the numerical value of \( c_m \) remains within a range of 0.7–0.9 for typical hydrocarbon–air mixtures [33].

The modeling of SDR not only is useful for the closure of filtered reaction rate but also plays a pivotal role in the closure of micro-mixing rate in the context of pdf methodology [30, 39–41]. For turbulent premixed flames, the Favre-filtered SDR \( \bar{N}_c \) can be modeled either by using an algebraic expression in terms of the resolved quantities or by solving a modeled transport equation. A few recent analyses [36–41] have concentrated on the algebraic closure of SDR for turbulent premixed flames in the context of LES. Algebraic closures are suitable when an equilibrium is maintained between the generation and destruction rates of \( \bar{N}_c \), but this assumption may be rendered invalid under some conditions (e.g., low Damköhler number lean premixed combustion). A number of previous analyses [34, 42–51] concentrated on the modeling of SDR transport in turbulent premixed combustion in the context of RANS simulations. Interested readers are referred to Ref [34], for a detailed review of the existing modeling methodologies for SDR transport in the context of RANS simulations. Recent advancements in high-performance computing have made LES of industrial flows more affordable than in the past, and LES is more successful in capturing unsteady flow features than RANS. However, relatively limited attention has been given to the investigation of SDR transport in the context of LES [52, 53]. Recently, models for the unclosed terms of the SDR \( \bar{N}_c \) transport equation for unity Lewis number flames in the context of LES have been proposed [53], but the differential diffusion effects due to non-unity \( Le \) were not addressed. A recent analysis [28] concentrated on the influences of global \( Le \) on the statistical behaviors of the unclosed terms of the \( \bar{N}_c \) transport equation based on an order-of-magnitude approach, which successfully explained the effects of global \( Le \) and the filter width \( \Delta \) dependences of the Favre-filtered SDR \( \bar{N}_c \) and its transport. It has been found that \( Le \) has significant influences on both the qualitative and quantitative behaviors of the unclosed terms of the SDR \( \bar{N}_c \) transport equation [22, 28]; however, the modeling of \( Le \) effects on these unclosed terms is yet to be addressed, and the present analysis aims to address this gap in the existing literature. In this respect the main objectives of this paper are:

i. To propose models for the unclosed terms of the SDR transport equation in such a manner that the performances of these models remain satisfactory for a range of \( \Delta \) and \( Le \).

ii. To assess the performances of the newly proposed models with respect to explicitly filtered Direct Numerical Simulation (DNS) data.

These objectives are addressed here by conducting a priori analysis using a DNS database of statistically planar turbulent premixed flames with a range of different values of \( Le \) (i.e., \( Le = 0.34–1.2 \)). The details related to mathematical background and numerical implementation are provided in the next section. This is followed by the presentation of the results and subsequent discussion. The main findings are summarized and conclusions are drawn in the final section of this paper.

### 2. Mathematical background & numerical implementation

Three-dimensional DNS simulations with detailed chemistry are now possible, but they remain extremely expensive and need several millions of CPU hours [54] for conducting extensive parametric
variations and carrying out explicit filtering of DNS data using a range of filter widths $\Delta$, as has been carried out in the current study. Thus, the chemical mechanism has been simplified here as a single-step Arrhenius-type irreversible chemical reaction. Under this condition the species field is uniquely represented by a reaction progress variable $c$, which can be defined by using the mass fraction of a suitable reactant $Y_R$ as $c = (Y_{R0} - Y_R)/(Y_{R0} - Y_{R\infty})$, where subscripts $0$ and $\infty$ denote the values in the unburned and burned gases, respectively. The transport equation of $c$ can be used to derive a transport equation of $N_c = \rho D \nabla c \cdot \nabla c / \rho$, which takes the following form [28, 53]:

$$\frac{\partial (\rho \tilde{N}_c)}{\partial t} + \frac{\partial (\rho \tilde{u}_i \tilde{N}_c)}{\partial x_i} = \frac{\partial}{\partial x_i} \left( \rho D \frac{\partial N_c}{\partial x_i} \right) + T_1 + T_2 + T_3 + T_4 - D_2 + f(D) \tag{3}$$

where $u_i$ is the $i$th component of the velocity vector. On the left-hand side of Eq. (3) the terms denote the transient effects and resolved advection of $\tilde{N}_c$, respectively. The term $D_1$ depicts the molecular diffusion of $\tilde{N}_c$, and the terms $T_1$, $T_2$, $T_3$, $T_4$, $(-D_2)$, and $f(D)$ are all unclosed and expressed as follows:

$$T_1 = -\frac{\partial}{\partial x_j} \left[ \rho \tilde{u}_j \tilde{N}_c - \rho \tilde{u}_j \tilde{N}_c \right] \tag{4i}$$

$$T_2 = -\frac{2D}{\rho} \left[ \tilde{w} + \frac{\partial}{\partial x_i} \left( \rho D \frac{\partial c}{\partial x_j} \right) \frac{\partial c}{\partial x_i} \frac{\partial \rho}{\partial x_i} \frac{\partial \rho}{\partial x_j} \right] \tag{4ii}$$

$$T_3 = -2\rho D \frac{\partial c}{\partial x_i} \frac{\partial u_i}{\partial x_j} \frac{\partial c}{\partial x_j} \tag{4iii}$$

$$T_4 = 2D \frac{\partial \tilde{w}}{\partial x_i} \frac{\partial c}{\partial x_i} \tag{4iv}$$

$$(-D_2) = -2\rho D^2 \frac{\partial^2 c}{\partial x_i \partial x_j} \frac{\partial^2 c}{\partial x_i \partial x_j} \tag{4v}$$

$$f(D) = f_1(D) = \frac{2D}{\partial x_k} \frac{\partial (\rho D)}{\partial x_k} \frac{\partial^2 c}{\partial x_i \partial x_j} + \frac{2D}{\partial x_k} \frac{\partial^2 (\rho D)}{\partial x_k \partial x_k} \frac{\partial c}{\partial x_j} - \frac{\partial}{\partial x_j} \left( \rho N_c \frac{\partial D}{\partial x_j} \right)$$

$$- 2\rho D \frac{\partial D}{\partial x_j} \frac{\partial c}{\partial x_i} \frac{\partial c}{\partial x_k} \frac{\partial c}{\partial x_k} + \rho \frac{\partial c}{\partial x_k} \frac{\partial c}{\partial x_k} \frac{\partial D}{\partial x_j} \left[ \frac{\partial D}{\partial t} + \tilde{u}_j \frac{\partial D}{\partial x_j} \right] \tag{4vi}$$

where $\tilde{w}$ is the reaction rate of $c$. The term $T_1$ represents the effects of sub-grid convection, whereas $T_2$ denotes the effects of density-variation due to heat release. The term $T_3$ is determined by the alignment of $\nabla c$ with local strain rate $\epsilon_{ij} = 0.5(\partial u_i / \partial x_j + \partial u_j / \partial x_i)$, and this term is commonly referred to as the scalar-turbulence interaction term. The term $T_4$ arises due to the correlation between $\nabla \tilde{w}$ and $\nabla c$, whereas $(-D_2)$ denotes the molecular dissipation of SDR; these terms will henceforth be referred to as the reaction rate term and the dissipation term, respectively. The term $f(D)$ denotes the effects of $D$ variation. An a priori DNS modeling of the above-mentioned unclosed terms will be discussed in Section 3 of this paper.

For the present analysis, a DNS database of freely propagating turbulent premixed flames has been considered. The simulation domain is taken to be a cube of $24.1\delta_{th} \times 24.1\delta_{th} \times 24.1\delta_{th}$, which is discretized using a uniform Cartesian grid of $230 \times 230 \times 230$ points, ensuring about 10 grid points are kept within $Min(\delta_{th}, \delta_{th})$, where $\delta_t = 1/(Max|\nabla c|)$ is an alternative flame thickness based on $|\nabla c|$ and the values of $\delta_t / \delta_{th}$ for cases A–E (with $Le = 0.34, 0.6, 0.8, 1.0, \text{and } 1.2$) are provided in Table 1. The initial values of the normalized root-mean-square (rms) value of turbulent velocity $u’/S_L$, integral length scale to thermal flame thickness ratio $l/\delta_{th}$, Damköhler number $Da = lS_L/u’\delta_{th}$, Karlovitz number $Ka = (u’/S_L)^{3/2} (l/\delta_{th})^{-1/2}$, turbulent Reynolds number $Re_t = \rho_0u’l/\mu_0$, and $\tau = (T_{ad} - T_0)/T_0$ are presented in Table 1 along with domain and grid sizes, where $\rho_0$ and $\mu_0$ are the unburned gas density and viscosity,
Table 1. Initial values of simulation parameters and nondimensional numbers relevant to the DNS database considered for this analysis.

| Case | Le | $u'/S_L$ | $\delta/\delta_{th}$ | $l/\delta_{th}$ | $\tau$ | Re$_T$ | Da | Ka |
|------|----|---------|----------------|-----------------|--------|-------|----|----|
| A    | 0.34 | 7.5   | 2.17           | 2.45            | 4.5    | 47.0  | 0.33 | 13.0 |
| B    | 0.6  | 7.5   | 1.40           | 2.45            | 4.5    | 47.0  | 0.33 | 13.0 |
| C    | 0.8  | 7.5   | 1.15           | 2.45            | 4.5    | 47.0  | 0.33 | 13.0 |
| D    | 1.0  | 7.5   | 1.0            | 2.45            | 4.5    | 47.0  | 0.33 | 13.0 |
| E    | 1.2  | 7.5   | 0.90           | 2.45            | 4.5    | 47.0  | 0.33 | 13.0 |

For all cases $\tau = 4.5$; $\beta = 6.0$; $Pr = 0.7$; $Ma = S_L / \sqrt{RT_0} = 0.014159$.

respectively, and $S_L$ is the unstrained laminar burning velocity. The flamelet assumption is likely to be valid for the values of $u'/S_L$ and $l/\delta_{th}$ considered here, and all cases considered here represent the thin reaction zones regime combustion according to the regime diagram by Peters [55].

The simulations have been carried out using a well-known DNS code called SENGA [56]. For all cases the boundary conditions in the mean flame propagation direction are considered to be partially nonreflecting, whereas boundaries in the transverse directions are considered to be periodic. A 10th order central difference scheme is used for spatial differentiation for the internal grid points and the order of differentiation gradually drops to a one-sided second-order scheme at the non-periodic boundaries. A low-storage third-order Runge-Kutta method is used for explicit time advancement for all the governing equations. In all cases flame–turbulence interaction takes place under decaying turbulence, which necessitates the simulation time $t_{sim} \geq \max(t_p, t_c)$, where $t_p = l/\nu'$ is the initial eddy turnover time and $t_c = \alpha \tau T_0 / S_L^2$ is the chemical time scale, with $\alpha \tau T_0$ being the unburned gas thermal diffusivity. The simulations have been carried out for about $3.34 t_f = 3.34 l/\nu'$, which amounts to approximately $1.75 \alpha \tau T_0 / S_L^2$ for all cases considered here. Several studies [12–15, 19, 57–61] with either similar or smaller simulation time have contributed significantly to the fundamental understanding and modeling of turbulent premixed combustion in the past. By the time the statistics were extracted, the value of $u'/S_L$ in the unburned reactants ahead of the flame had decayed by about 50%, whereas the value of $l/\delta_{th}$ had increased by about 1.7 times, relative to their initial values. This database has been used in several previous analyses [20–28] and it was shown in Ref [23] that the volume-integrated burning rate for the $Le = 1.0$ and 1.2 flames reached quasi-steady state by the time statistics were extracted. However, the $Le < 1.0$ flames are thermo-diffusively unstable and thus the volume-integrated burning rate increases with time for these cases [23]. The qualitative nature of the statistics was found to remain unchanged and the scaling estimates presented in the next section remain valid since $t = 2.0 l/\nu'$ for all cases considered here.

The unclosed terms of the transport equation of $\bar{N}_c$ have been evaluated by explicitly filtering DNS data using a standard three-dimensional Gaussian filter [28, 53, 57, 58, 60]:

$$G(\vec{r}) = (6/\pi \Delta^2)^{3/2} \exp(-6\vec{r} \cdot \vec{r}/\Delta^2)$$

and the filtered values of a general quantity $Q$ are given by the following integral:

$$\bar{Q}(\vec{x}) = \int Q(\vec{x} - \vec{r})G(\vec{r})d\vec{r}.$$  

In the next section, the results will be presented for $\Delta$ ranging from $\Delta = 0.4 \delta_{th}$ to $\Delta = 2.8 \delta_{th}$. This range of filter widths is comparable to the range of $\Delta$ used in several previous a priori DNS analyses [57, 58, 60], and addresses a range of different length scales from $\Delta$ comparable to $\delta_{th} = 1.75 \delta_c$ ($\delta_c = \alpha \tau T_0 / S_L$ is the Zel’dovich flame thickness) up to $2.8 \delta_{th} = 5.0 \delta_c$, where $\Delta$ is comparable to the integral length scale.

### 3. Results and discussion

The distributions of $\bar{c}$ on $x_1 - x_2$ mid-plane for $\Delta = 0.8 \delta_{th}$, $1.6 \delta_{th}$, and $2.8 \delta_{th}$ at $t = 1.75 t_L$ for cases A–E are shown in Figure 1, which shows an increase in the extent of flame wrinkling with decreasing $Le$. The extent of flame wrinkling can be quantified in terms of the normalized turbulent flame surface area $A_T/A_L$, where the flame surface area is evaluated using the volume integration of the form:

$$A = \int \sqrt{|\nabla \bar{c}|} dV$$

with subscripts “T” and “L” denoting the turbulent and laminar flame values, respectively [28]. The values of $A_T/A_L$ and the normalized turbulent burning velocity $S_T/S_L$ (where $S_T = (\rho_c A_p)^{-1} \int \bar{w} dV$) at $1.75 t_L = \delta_{th}/S_L$ are listed in Table 2, which demonstrates that both
Table 2. The effects of Lewis number on normalized flame surface area $A_T/A_L$ and normalized turbulent flame speed $S_T/S_L$ when the statistics were extracted (i.e., $t = 1.75 \alpha_{ST}/S_L^2$).

| Case | $A_T/A_L$ | $S_T/S_L$ |
|------|-----------|-----------|
| A    | 3.93      | 13.70     |
| B    | 2.66      | 4.58      |
| C    | 2.11      | 2.53      |
| D    | 1.84      | 1.83      |
| E    | 1.76      | 1.50      |

$A_T/A_L$ and $S_T/S_L$ increase significantly with decreasing Lewis number. The burning rate per unit area in turbulent flames increases (decreases) compared with the corresponding laminar value as a result of negative (positive) Markstein length [7–10] for the $Le < 1$ ($Le > 1$) flames. This, in turn, leads to $S_T/S_L > A_T/A_L$ ($S_T/S_L < A_T/A_L$) in the $Le < 1$ ($Le > 1$) flames (see Table 2). It can be further seen from Figure 1 that the flame brush thickens (i.e., the magnitude of $\nabla \tilde{c}$ decreases) and the extent of flame wrinkling decreases with increasing $\Delta$ as a result of the smearing of local information due to the convolution operation associated with LES filtering. As the SDR is related to the reaction rate, and the gradient of the reaction progress variable, the effects of $Le$ on burning rate and $\Delta$ dependence of $\nabla \tilde{c}$ are expected to influence the statistical behavior of SDR $\tilde{N}_c$ and its transport. The effects of $Le$ and $\Delta$ on the statistical behavior of SDR $\tilde{N}_c$ and its transport have been analyzed elsewhere [28] and the current analysis will only concentrate on the influences of global Lewis number on the modeling of SDR transport.

The normalized mean values of $T_1$, $T_2$, $T_3$, $T_4$, $(-D_2)$, and $f(D)$ conditional on $\tilde{c}$ for cases A–E are shown in Figure 2 for $\Delta = 0.4\delta_{th}$ and $\Delta = 2.8\delta_{th}$, Figure 2 shows that $T_2$ and $(-D_2)$ act as source and sink, respectively, in all cases, which is consistent with previous findings [21, 28]. The contribution of $T_4$ is positive for a major portion of the flame brush before becoming negative toward the burned gas side for $\Delta \leq \delta_{th}$ (e.g., $\Delta = 0.4\delta_{th}$); however, for $\Delta > \delta_{th}$ (e.g., $\Delta = 2.8\delta_{th}$) the contribution of $T_4$ remains a leading-order source throughout the flame brush. The term $T_3$ assumes positive values toward the unburned gas side of the flame brush before assuming mostly negative values for the major part of the flame brush in cases D and E, whereas $T_3$ is negative throughout the flame brush in cases A–C for all filter widths. The contribution of $f(D)$ is negative (positive) toward the unburned (burned) gas side of the flame brush for all cases and for all filter widths. The magnitude of $T_1$ is negligible compared with $T_2$, $T_3$, $T_4$, $(-D_2)$, and $f(D)$ for all $\Delta$ in all cases. It can be seen from Figure 2 that the magnitude of all the terms decrease with increasing $Le$ and $\Delta$, which is consistent with previous findings based on DNS data [22, 28]. The observed behaviors of $T_1$, $T_2$, $T_3$, $T_4$, $(-D_2)$, and $f(D)$ in response to $Le$ and $\Delta$ have recently been explained by Gao et al. [28] using a detailed scaling analysis, and the scaling estimates of the filtered SDR and the unclosed terms of the SDR transport equation are provided in Table 3. It is worth noting that $m$ and $n$ in Table 3 are positive numbers with magnitudes greater than unity, and the functions $g(Le)$, $q(Le)$, $\Psi_1(Le)$, and $\Psi(Le)$ increase with decreasing global Lewis number $Le$. It can be seen from the scaling estimates in Table 3 that the magnitudes of the terms $T_1$, $T_2$, $T_3$, $T_4$, $(-D_2)$, and $f(D)$ are expected to increase with decreasing filter width and global Lewis number. Interested readers are referred to [28] for further discussion on the derivation of the scaling estimates of $T_1$, $T_2$, $T_3$, $T_4$, $(-D_2)$, and $f(D)$, and only the modeling of these terms will be discussed in this paper in the following subsections.

### 3.1. Modeling of the turbulent transport term $T_1$

Equation (4i) indicates that the turbulent transport term $T_1$ could be satisfactorily closed if the sub-grid flux of SDR (i.e., $\bar{\rho}u_i\tilde{N}_c - \bar{\rho}u_i\bar{N}_c$) is properly modeled. The sub-grid flux of SDR ($\bar{\rho}u_i\tilde{N}_c - \bar{\rho}u_i\bar{N}_c$) is often modeled using a gradient hypothesis as follows [34]:

$$
(\bar{\rho}u_i\tilde{N}_c - \bar{\rho}u_i\bar{N}_c) = -\bar{\rho}D_t \frac{\partial \tilde{N}_c}{\partial x_i}
$$

(5i)
where $D_{t}$ is the sub-grid scale eddy diffusivity. It has been demonstrated earlier that the turbulent scalar flux of scalar gradients (e.g., flame surface density and SDR) may exhibit counter-gradient (gradient) transport for the flames when counter-gradient (gradient) transport is observed for $(\overline{u'\varepsilon'} - \overline{u'\varepsilon'})$ [20, 22, 23, 62]. Thus, the modeling of $T_{1}$ needs to include both gradient and counter-gradient transports of $(\overline{\rho u_{i}N_{i}} - \overline{\rho u_{i}N_{i}})$.

Figure 1. Distributions of $\tilde{c}$ on $x_{1} - x_{2}$ mid-plane for $\Delta = 0.8\delta_{th}$ (1st column), 1.6$\delta_{th}$ (2nd column), 2.8$\delta_{th}$ (3rd column) for cases A–E (1st–5th row) when the statistics were extracted (i.e., $t = 1.75 \alpha_{th}/S_{L}^{2}$).
Figure 2. Variations of $T_1$ (——), $T_2$ (—), $T_3$ (—), $T_4$ (—), $(-D_2)$ (—), and $f(D)$ (—) conditionally averaged in bins of $\varepsilon$ for $\Delta = 0.4\delta_m$ (1st column), $1.6\delta_m$ (2nd column), and $2.8\delta_m$ (3rd column) in cases A–E (1st–5th row). All the terms are normalized with respect to $q_0^2/\delta_m^2$.

Table 3. Summary of the scaling estimates of the relevant quantities according to Gao et al. [28].

| Quantities | Scaling estimates |
|------------|-------------------|
| $N_c$ | $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-0.5}Re_{\Delta}^{0.5}$ alternatively $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-1}$ |
| $D\nabla \varepsilon, \nabla \varepsilon$ | $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-0.5}Re_{\Delta}^{0.5}$ alternatively $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-1}$ |
| $T_1$ | The above expressions can be combined as $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-0.5}Re_{\Delta}^{0.5}$ alternatively $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-1}$ |
| $T_2$ (see Eq. 7ii) | $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-0.5}Re_{\Delta}^{0.5}$ alternatively $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-1}$ |
| $(T_2)_{res}$ (see Eq. 11ii) | $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-0.5}Re_{\Delta}^{0.5}$ alternatively $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-1}$ |
| $T_3$ (see Eq. 14iii) | $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-0.5}Re_{\Delta}^{0.5}$ alternatively $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-1}$ |
| $T_4$ (see Eq. 14iv) | $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-0.5}Re_{\Delta}^{0.5}$ alternatively $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-1}$ |
| $(-D_2)$ (see Eq. 14i) | $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-0.5}Re_{\Delta}^{0.5}$ alternatively $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-1}$ |
| $(-D_2)_{res}$ (see Eq. 14ii) | $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-0.5}Re_{\Delta}^{0.5}$ alternatively $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-1}$ |
| $f(D)$ (see Eq. 14iii) | $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-0.5}Re_{\Delta}^{0.5}$ alternatively $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-1}$ |
| $(f(D))_{res}$ (see Eq. 14iii) | $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-0.5}Re_{\Delta}^{0.5}$ alternatively $\frac{3}{\delta_m^2} \frac{Le^{-2}Re_{\Delta}^{-1}Da_{\Delta}^{-1}}{\delta_m} \times Le \times Da_{\Delta}^{-1}$ |
Gao et al. [28] demonstrated that the unclosed term $T_1$ can be scaled in the following manner:

$$T_1 \sim \frac{\rho_0 c g(Le) S_I \tilde{N}_c}{\Delta} \sim \frac{\rho_0 c g(Le) S_I^2}{\delta_{th}^2} \times Le \times Da_{\Delta}^{-0.5} Re_{\Delta}^{-0.5} \text{ for } \Delta \gg \delta_{th}$$  \hspace{1cm} (5ii)

where $g(Le)$ is a function increasing with decreasing $Le$, which accounts for flame normal acceleration, $S_I$ is used to scale the sub-grid velocity fluctuations associated with sub-grid scalar gradients, and the sub-grid fluctuations of SDR are taken to scale with the following scaling estimate, which is valid for both gradient and counter-gradient transport:

$$Da_{\Delta} = \Delta S_I / u'_\Delta \delta_{th} \text{ and } Re_{\Delta} = \rho_0 u'_\Delta / \mu_0$$

In Eq. (5ii), $Da_{\Delta}$ is the Damköhler number and $Re_{\Delta}$ is the sub-grid turbulent velocity fluctuation and sub-grid kinetic energy, respectively. One obtains $Da_{\Delta} Re_{\Delta} \sim (\Delta / \delta_{th})^2$ using $\mu_0 \sim \rho_0 S_I \delta_{th}$, which indicates that $Da_{\Delta} Re_{\Delta}$ increases with increasing $\Delta$. Alternatively, one obtains the following expression when the sub-grid velocity fluctuations are taken to scale with $u'_\Delta$ [28]:

$$T_1 \sim \frac{\rho_0 u'_\Delta \tilde{N}_c}{\Delta} \frac{\rho_0 S_I^2}{\delta_{th}^2} \times Le \times Da_{\Delta}^{-1} \text{ for } \Delta \gg \delta_{th}$$  \hspace{1cm} (5iii)

It is worth noting that the scaling estimate given by Eq. (5ii) (Eq. (5iii)) is more appropriate for counter-gradient (gradient) transport. Equations (5ii) and (5iii) can be combined to obtain the following scaling estimate, which is valid for both gradient and counter-gradient transport [28]:

$$T_1 \sim \frac{(\rho u_i N_c - \rho \tilde{u}_i \tilde{N}_c)}{\Delta} \sim \frac{(\rho u_i - \rho \tilde{u}_i \tilde{c}) \tilde{N}_c}{\Delta} \text{ and } (\rho u_i N_c - \rho \tilde{u}_i \tilde{N}_c) \sim \frac{(\rho u_i - \rho \tilde{u}_i \tilde{c}) \tilde{N}_c}{\Delta} \text{ for } \Delta \gg \delta_{th}$$  \hspace{1cm} (5iv)

Gao et al. [53] have recently extended a RANS model proposed by Chakraborty and Swaminathan [51] for the purpose of modeling $(\rho u_i N_c - \rho \tilde{u}_i \tilde{N}_c)$ for the unity Lewis number flames in the context of LES in the following manner:

$$\frac{\rho u_i N_c - \rho \tilde{u}_i \tilde{N}_c}{\Delta} \sim \left\{ \Phi' \tilde{c} \frac{\gamma_1 (\rho u_i - \rho \tilde{u}_i \tilde{c}) - \gamma_2 \rho \tilde{c} (1 - \tilde{c}) u'_\Delta M_i}{\tilde{c} (1 - \tilde{c})} \frac{\tilde{N}_c}{\tilde{c}} \right\}$$  \hspace{1cm} (6i)

where $M_i = -(\partial \tilde{c} / \partial x_i) / | \nabla \tilde{c} |$ is the $ith$ component of the resolved flame normal vector for LES, $\Phi' = 0.7$ is a model parameter, and the following values have been suggested for $\gamma_1$, $\gamma_2$, and $C_F$ [53]:

$$\gamma_1 = 1.8, \quad \gamma_2 = 4.9 - 3.2 \text{erf}(0.15 Re_{\Delta}) \quad \text{and} \quad C_F = 0.11$$  \hspace{1cm} (6ii)

The parameterization given by Eq. (6ii) ensures that $\gamma_2$ assumes an asymptotic value for large values of $Re_{\Delta}$ (i.e., $Re_{\Delta} \rightarrow \infty$). In Eq. (6i), the first term on the right-hand side principally accounts for the effects of flame normal acceleration due to heat release, whereas the last term on the right-hand side of Eq. (6i) represents turbulent transport according to conventional gradient hypothesis. Moreover, the first and second terms on the right-hand side of Eq. (6i) are consistent with the scaling estimates given by Eqs. (5iv) and (5iii), respectively.

The predictions of $J_{\Phi}^+ = \frac{(\rho u_i N_c - \rho \tilde{u}_i \tilde{N}_c) M_i}{\delta_{th} / \rho_0 S_I^2}$ according to Eq. (6i) with $\Phi' = 0.7$ are compared to the corresponding quantity extracted from DNS data for $\Delta = 0.4 \delta_{th}$, $1.6 \delta_{th}$, and $2.8 \delta_{th}$ in Figure 3 for cases $A$–$E$. Figure 3 shows that even though Eq. (6i) predicts $J_{\Phi}^+$ in a reasonable manner in the cases with $Le \approx 1.0$ (e.g., cases C–E), this model does not adequately capture the correct qualitative and quantitative behaviors of $J_{\Phi}^+$ for the flames with $Le \ll 1.0$ (i.e., cases A and B). The model given by Eqs. (6i) and (6ii) does not explicitly account for non-unity Lewis number effects, so it is not surprising that this model does not adequately capture the behavior of $(\rho u_i N_c - \rho \tilde{u}_i \tilde{N}_c)$ for $Le \ll 1.0$ flames where the nondimensional temperature $T^+ = (T - T_0) / (T_{ad} - T_0)$ is significantly different from the reaction progress variable $c$, which alters the distribution of heat release and
thermal expansion within the flame brush compared with the $Le = 1.0$ flames. This behavior is mimicked here by introducing $Le$ dependence of the model parameter $\Phi'$ in the following manner:

$$\Phi' = 0.3(1 - Le) + 0.7$$  \hspace{1cm} (6iii)
The predictions of the model given by Eq. (6i) with \( \Phi' \) according to Eq. (6iii) are also shown in Figure 3, which demonstrates that the model with new parameterization \( \Phi' = 0.3(1 - Le) + 0.7 \) predicts \( J'_{\infty} \) satisfactorily for all filter widths in all cases considered here and the agreement between the predictions of Eq. (6i) and DNS data improves with increasing \( \Delta \) (see Figure 3). The predictions of Eq. (6i) with \( \Phi' \) according to Eq. (6iii) become equal to the corresponding values obtained for \( \Phi' = 0.7 \) for the \( Le = 1.0 \) case, and these two predictions cannot be distinguished from each other for case D in Figure 3. It worth noting that the sub-grid flux of the reaction progress variable (i.e., \( \bar{\rho}u'c - \bar{\rho}u_c \)) requires modeling in LES, and the performances of the models for \( (\bar{\rho}u'N'_{\infty} - \bar{\rho}u_c N'_{\infty}) \) and the turbulent transport term \( T_1 \) depend on the modeling of \( (\bar{\rho}u'c - \bar{\rho}u_c) \). The modeling of \( (\bar{\rho}u'c - \bar{\rho}u_c) \) is beyond the scope of the current analysis and interested readers are referred to recent investigations by Chakraborty and Cant [63] and Gao et al. [64] for further discussion on the modeling of turbulent scalar fluxes in premixed turbulent flames.

### 3.2. Modeling of the density variation term \( T_2 \)

For unity Lewis number flames the gas density \( \rho \) can be expressed as \( \rho_0/(1 + \tau c) \) [33], which leads to an alternative expression for the density variation term \( T_2 \) as [22, 47, 48, 51, 53]:

\[
T_2 = 2(\rho \nabla \cdot \bar{u}N_{\infty}).
\]

However, \( \rho = \rho_0/(1 + \tau T') \neq \rho_0/(1 + \tau c) \) in the non-unity Lewis number flames because the equality between \( T' \) and \( c \) no longer holds. Although \( T_2 = 2(\rho \nabla \cdot \bar{u}N_{\infty}) \) does not strictly hold in non-unity Lewis number flames, the gas density can still be scaled as \( \rho \sim \rho_0/(1 + \tau c) \); thus, the density variation term \( T_2 \) can be scaled for adiabatic flames with low Mach number as follows [28]:

\[
T_2 \sim 2 \left( \rho \frac{\partial u_i}{\partial x_i} N_{\infty} \right) \sim \frac{\rho_0 \tau S_L^2}{Le^{m-1} \delta_{th}^2} \tag{7i}
\]

where \( m \) is a positive number greater than unity (i.e., \( m > 1 \)). The resolved part of \( T_2 \) can be taken to scale as [28]

\[
(T_2)_{res} = -\frac{2\bar{D}}{\bar{\rho}} \left[ \bar{\omega} + \frac{\partial}{\partial x_j} \left( \rho D \frac{\partial c}{\partial x_j} \right) - \frac{\partial \left( \bar{\rho}u'c - \bar{\rho}u_c \right) \rho c}{\partial x_j} \right] \frac{\partial c}{\partial x_i} \frac{\partial \bar{\rho}}{\partial x_i} \tag{7ii}
\]

where \( U_{ref} \) is a velocity scale representing the Favre-filtered velocity components \( \bar{u}_i \). It is worth noting that \( \bar{\rho} = \rho_0/(1 + \tau \bar{c}) \) for unity Lewis number flames yields \( (T_2)_{res} = 2\bar{\rho}D \nabla \bar{c}. \nabla \bar{c}(\bar{u}_i/\bar{c}_i) \); however, the expression \( \bar{\rho} = \rho_0/(1 + \tau \bar{c}) \) does not strictly hold for non-unity Lewis number flames, but \( \bar{\rho} \) and \( (T_2)_{res} \) can still be scaled using \( \rho_0/(1 + \tau c) \) and \( 2\bar{\rho}D \nabla \bar{c}. \nabla \bar{c}(\bar{u}_i/\bar{c}_i) \), respectively.

The scaling estimates given by Eqs. (7i) and (7ii) demonstrate that \( T_2 \) remains of the order of \( \rho_0 \tau S_L^2/\delta_{th}^2 \) irrespective of \( \Delta \). By contrast, the magnitude of \( (T_2)_{res} \) remains comparable to \( \rho_0 S_L^2/\delta_{th}^2 \) for \( U_{ref} \sim S_L \) and \( \Delta = \delta_{th} \), however, the magnitude of \( (T_2)_{res} \) is expected to decrease with increasing \( \Delta \). This suggests that the sub-grid component \( (T_2)_{sg} = T_2 - (T_2)_{res} \) plays an increasingly important role with increasing \( \Delta \), which can be substantiated from Figure 4, where the variations of the mean values of \( T_2 \) and \( (T_2)_{sg} = T_2 - (T_2)_{res} \) conditional on \( \bar{c} \) are shown for cases A–E for \( \Delta = 0.4 \delta_{th}, 1.6 \delta_{th}, \) and \( 2.8 \delta_{th} \).

Gao et al. [53] recently proposed the following model \( T_2 \) for unity \( Le \) flames in the following manner:

\[
T_2 = -\frac{2\bar{D}}{\bar{\rho}} \left[ \bar{\omega} + \frac{\partial}{\partial x_j} \left( \rho D \frac{\partial c}{\partial x_j} \right) - \frac{\partial \left( \bar{\rho}u'c - \bar{\rho}u_c \right) \rho c}{\partial x_j} \right] \frac{\partial c}{\partial x_i} \frac{\partial \bar{\rho}}{\partial x_i} + \beta_{T_2} \tau S_L \left[ \bar{\rho}N_{\infty} - \bar{\rho}D \nabla \bar{c}. \nabla \bar{c} \right] \frac{1}{\delta_{th}(1.0 + K\Delta)^{1/2}} \tag{8}
\]
where $Ka_{\Delta} = (u'_{\Delta}/S_L)^{3/2} (\Delta/\delta_{th})^{-1/2}$ is local sub-grid Karlovitz number and $\beta_{T_2} = 2.7$ is a model parameter. The first term on the right-hand side of Eq. (8) accounts for the resolved component $(T_2)_{res}$, whereas the second term models the sub-grid component. The Karlovitz number dependence in Eq. (8) ensures the diminishing strength of heat release with increasing $Ka_{\Delta}$ [22, 28, 47, 48, 51, 52] as the combustion process is likely to show the attributes of the broken reaction zones regime [55] (where the effects of heat release are weak) for high values of Karlovitz number. The prediction of
Eq. (8) is also shown in Figure 4 for cases A–E for $\Delta = 0.4 \delta_{th}, 1.6 \delta_{th},$ and $2.8 \delta_{th}$. A comparison between the predictions of Eq. (8) and the normalized $T_2$ extracted from explicitly filtered DNS data reveals that Eq. (8) satisfactorily predicts $T_2$ for a range of different filter widths for flames with $Le = 1.0$ (e.g., cases C–E); however, this model significantly underpredicts the magnitude of $T_2$ for the $Le \ll 1.0$ cases (e.g., cases A and B). It can be seen from Eq. (7i) that the magnitude of $T_2$ is expected to increase with decreasing $Le$ due to the strengthening of heat release effects as a result of the enhanced burning rate for the small values of Lewis number (see Table 2). As this effect is missing in Eq. (8), this model underpredicts the magnitude of $T_2$ for the $Le \ll 1.0$ cases (e.g., cases A and B), where the effects of enhanced heat release due to the differential diffusion of heat and mass are particularly strong.

Here the model given by Eq. (8) has been extended in order to account for the effects of enhanced heat release due to the differential diffusion of heat and mass are particularly strong.

The variations of the mean values of $T_3$ conditional on $\tilde{c}$ are shown in Figure 5 for cases A–E at $\Delta = 0.4 \delta_{th}, 1.6 \delta_{th},$ and $2.8 \delta_{th}$. Figure 5 shows that $T_3$ assumes predominantly negative values throughout the flame brush for cases A–C, but this term exhibits weak positive values toward both the unburned gas sides of the flame brush before assuming mostly negative values for the major portion of the flame brush in cases D and E. The term $T_3$ can be expressed as follows [21, 28, 34, 45–48]:

$$T_3 = \frac{2\rho (e_a \cos^2 \alpha + e_b \cos^2 \beta + e_c \cos^2 \gamma) N_c}{\partial t}$$ (10)

where $e_a$, $e_b$, and $e_c$ are the most extensive, intermediate, and most compressive principal strain rates and their angles with $\nabla \tilde{c}$, respectively. Equation (10) suggests that a predominant collinear alignment of $\nabla \tilde{c}$ with $e_a$ ($e_c$) leads to a negative (positive) value of $T_3$. It was discussed elsewhere [21, 28, 34, 45–48].
that $\nabla c$ predominantly aligns with $e_{\alpha}$ when the strain rate induced by the flame normal acceleration overcomes turbulent straining, whereas one obtains preferential alignment of $\nabla c$ with $e_{\gamma}$ when turbulent straining dominates over the strain rate due to flame normal acceleration. The flame normal acceleration strengthens with decreasing $Le$, and thus $\nabla c$ predominantly aligns with $e_{\alpha}$ for the $Le \ll 1$ flames (e.g., cases A and B), leading to negative values of $T_3$ [21, 22, 28]. By contrast, turbulent straining overcomes the flame normal acceleration on both ends of the flame brush for the $Le = 1.0$ cases considered here (e.g., cases C–E), which leads to positive values of $T_3$ both on unburned and on burned

Figure 5. Variations of $T_3$ (solid), $(T_3)_{0s}$ (dashed) conditionally averaged in bins of $\bar{c}$ along with the predictions of Eqs. (12i) and (12iii) (dashed-dotted) and Eqs. (13i) and (13ii) (dotted) for $\Delta \approx 0.4\delta_{th}$ (1st column), $\Delta \approx 1.6\delta_{th}$ (2nd column), and $\Delta \approx 2.8\delta_{th}$ (3rd column) in cases A–E (1st–5th row). All the terms are normalized with respect to $\rho_0 S^2 / \delta_m^2$. 
gas sides of the flame brush for $\Delta = 0.4\delta_{th}$. However, the flame normal acceleration dominates over turbulent straining in the middle of the flame brush where the effects of heat release are strong even in the $Le = 1.0$ cases considered here (e.g., cases C–E), which leads to negative values of $T_3$ for the major portion of the flame brush in these cases.

The effects of $\nabla c$ alignment with $e_a$, on $T_3$ can be scaled in the following manner [28]:

$$T_3 \approx \frac{\rho_0 \tau_{s}\tilde{N}_c}{Le^n\delta_{th}} \approx \frac{\rho_0 \tau_{s}^{2}}{Le^{n-1}\delta_{th}} \quad \text{where} \quad n > 1$$ (11i)

The contribution of $\nabla c$ alignment with $e_{y}$ on $T_3$ can be scaled as follows [28]:

$$T_3 \approx \frac{\rho_0 u'_{A} \tilde{N}_c}{\Delta} \approx \frac{\rho_0 u_{x}^{3}}{\delta_{th}} \times Le \times Pr^{-1/2} \times Ka_{A} \quad \text{for} \quad \Delta \gg \delta_{th}$$ (11ii)

The Lewis number $Le$ dependence in Eq. (11i) (with $n > 1$) accounts for the greater extent of $\nabla c$ alignment with $e_a$ for the flames with $Le < 1.0$. Gao et al. [28] proposed the following scaling estimate of the resolved part of $T_3$:

$$(T_3)_{res} = -2\rho\tilde{D} \frac{\partial c}{\partial x_i} \frac{\partial \tilde{u}_i}{\partial x_j} \frac{\partial \tilde{c}}{\partial x_j} + (1 - f_{T_3})(C_3 - C_4 \tau da_{\Delta}) \frac{u'_{A}}{\Delta} \tilde{p}\tilde{N}_c$$ (12i)

where $C_3$ and $C_4$ are the model parameters and $Da_{\Delta} = S_{L0}\rho_0 \Delta/\tilde{u}'_{A} \delta_{th}$ is the density-weighted local sub-grid Damköhler number. The symbol $f_{T_3}$ is a bridging function in terms of $\Delta S_{L}/\alpha_{T0}$, which ensures that $(T_3)_{res} \approx T_3$ for $\Delta \gg \delta_{th}$ and $T_3$ approaches $(T_3)_{res}$ when the flow is fully resolved:

$$\lim_{\Delta \to 0} T_3 = \lim_{\Delta \to 0} \left(-2\rho\tilde{D} \frac{\partial c}{\partial x_i} \frac{\partial \tilde{u}_i}{\partial x_j} \frac{\partial \tilde{c}}{\partial x_j}\right) = -2\rho\tilde{D} \frac{\partial c}{\partial x_i} \frac{\partial \tilde{u}_i}{\partial x_j} \frac{\partial \tilde{c}}{\partial x_j}$$ (12ii)

Gao et al. [53] proposed the following expressions for the model parameter $C_3$, $C_4$, and $f_{T_3}$:

$$C_3 = 7.5; \quad C_4 = 0.75(1.0 + Ka_{\Delta})^{-0.4} \quad \text{and} \quad f_{T_3} = \exp[-1.05(S_{L0}/\alpha_{T0})^2]$$ (12iii)

It is worth noting that the terms $C_3\tilde{p}(u'_{A}/\Delta)\tilde{N}_c$ and $-C_4\rho_0 c(S_{L0}/\delta_{th})\tilde{N}_c$ are consistent with scaling estimates given by Eqs. (11i) and (11ii), respectively. However, a comparison between Eq. (11i) and $-C_4\rho_0 c(S_{L0}/\delta_{th})\tilde{N}_c$ reveals that the increased alignment of $\nabla c$ with $e_a$ for small values of $Le$ as a result of the strengthening of flame normal acceleration is not accounted for by the model given by Eq. (12i). The effects of flame normal acceleration are expected to weaken with increasing Karlovitz number as the reacting flow field exhibits some attributes of passive scalar mixing for large values of Karlovitz number in the broken reaction zones regime [55]. This behavior is mimicked here by $Ka_{\Delta}$ dependence of $C_4$ in Eq. (12iii).

The predictions of Eq. (12i) with the model parameters given by Eq. (12ii) are compared to $T_3$ extracted from DNS data in Figure 5, which shows that Eq. (12i) adequately captures the qualitative and quantitative behaviors of $T_3$ for the $Le = 1.0$ cases considered here (e.g., cases C–E); however, this model has been found to underpredict the magnitude of the negative contribution of $T_3$ in
the \(Le << 1.0\) cases (e.g., cases A and B) for \(\Delta > \delta_{th}\). It has already been noted that the increased extent of scalar gradient destruction in the \(Le << 1.0\) flames, due to the preferential alignment of \(\nabla c\) with \(e_a\) under strong actions of flame normal acceleration, is not addressed in the model given by Eq. (12i). Thus, this model underpredicts the negative contribution of \(T_3\) for the flames with \(Le << 1.0\). Here Eq. (12i) has been modified in the following manner to account for non-unity Lewis number effects:

\[
T_3 = -2\tilde{p}D \frac{\partial c}{\partial x_j} \frac{\partial u_i}{\partial x_j} \frac{\partial c}{\partial x_j} + (1 - f_{th}) \left| C_3 - C_4 \Gamma(Le) \right| \frac{\mu_A}{\Delta} \tilde{p} \tilde{N}_c
\]  

(13i)

where

\[
\Gamma(Le) = \frac{1.7(1 - \bar{c})^p}{Le^{2.57}} \left( \frac{\delta_L}{\delta_{th}} \right)^{1.3} \quad \text{and} \quad p = 0.2 + 1.5(1.0 - Le)
\]  

(13ii)

The involvement of the function \(\Gamma(Le)\) in Eq. (13i) accounts for the strengthening of \(\nabla c\) alignment with \(e_a\) under strong actions of flame normal acceleration in flames with small values of Lewis number. The presence of \((1 - \bar{c})^p\) helps Eq. (13i) capture the qualitative behavior of \(T_3\) across the flame brush. It can be seen from Figure 5 that the model given by Eq. (13i) provides satisfactory qualitative and quantitative predictions of \(T_3\) for all the flames with different values of \(Le\) for a range of \(\Delta\). It is worth noting that Eq. (13i) approaches Eq. (12i) for \(Le = 1.0\) and thus the predictions of Eqs. (12i) and (13i) cannot be distinguished from each other for case D in Figure 5.

**3.4. Modeling of the combined reaction, dissipation, and diffusivity gradient contribution \([T_4 - D_2 + f(D)]\)**

The variations of the mean values of \([T_4 - D_2 + f(D)]\) conditional on \(\bar{c}\) are shown in Figure 6 for A–E for \(\Delta = 0.4\delta_{th}, 1.6\delta_{th},\) and \(2.8\delta_{th}\). It can be seen from Figure 6 that \([T_4 - D_2 + f(D)]\) acts as a sink (source) term toward the burned (unburned) gas side of the flame brush for \(\Delta = 0.4\delta_{th}\) and \(\Delta = 1.6\delta_{th}\); however, the mean value of \([T_4 - D_2 + f(D)]\) conditional on \(\bar{c}\) assumes predominantly negative values for \(\Delta = 2.8\delta_{th}\). Table 3 shows that the order of magnitudes of \(T_4\), \((-D_2)\), and \(f(D)\) remain comparable according to the scaling analysis by Gao et al. [28] and their magnitudes are expected to increase with decreasing \(Le\). Furthermore, the scaling estimates of \((T_4)_{res}, (-D_2)_{res},\) and \(\{f(D)\}_{res}\) in Table 3 suggest that their contributions are expected to weaken with increasing \(\Delta\), where \((T_4)_{res}, (-D_2)_{res},\) and \(\{f(D)\}_{res}\) are the resolved components of \(T_4\), \((-D_2)\), and \(f(D)\), which are given by

\[
(T_4)_{res} = 2\tilde{D} \frac{\partial \tilde{c}}{\partial x_i} \frac{\partial \bar{c}}{\partial x_j} + 2\tilde{D} \frac{\partial \bar{c}}{\partial x_k} \frac{\partial \tilde{c}}{\partial x_j} \frac{\partial \bar{c}}{\partial x_l}
\]  

(14i)

\[
(-D_2)_{res} = -2\tilde{p}\tilde{D} \frac{\partial \bar{c}}{\partial x_i} \frac{\partial \bar{c}}{\partial x_j} - 2\tilde{p} \frac{\partial \bar{c}}{\partial x_k} \frac{\partial \tilde{c}}{\partial x_j} \frac{\partial \bar{c}}{\partial x_l} + \tilde{p} \frac{\partial \tilde{c}}{\partial x_i} \frac{\partial \bar{c}}{\partial x_j} \frac{\partial \tilde{c}}{\partial x_k}
\]  

(14ii)

\[
\{f(D)\}_{res} = 2\tilde{D} \frac{\partial \bar{c}}{\partial x_k} \frac{\partial \tilde{c}}{\partial x_j} \frac{\partial \bar{c}}{\partial x_l} + 2\tilde{D} \frac{\partial \bar{c}}{\partial x_k} \frac{\partial \tilde{c}}{\partial x_j} \frac{\partial \bar{c}}{\partial x_l}
\]  

(14iii)

Thus, the sub-grid components \((T_4)_{sg}, (-D_2)_{sg},\) and \(\{f(D)\}_{sg}\) are expected to play major roles for \(\Delta >> \delta_{th}\). The aforementioned behaviors of the resolved and sub-grid components of \(T_4\), \((-D_2)\), and \(f(D)\) can be confirmed from Figure 6. It can be seen from Table 3 that the magnitudes of \((T_4)_{sp}, (-D_2)_{sp},\) and \(\{f(D)\}_{sg}\) remain of the order of \(\rho_0 S^2_{\tilde{L}}/\delta_{th}^2 \sim \tilde{p}N_c^2\) for \(\Delta >> \delta_{th}\); however, their magnitudes are expected to increase with decreasing \(Le\), which can indeed be substantiated from Figure 6.
Gao et al. [53] utilized \( T_4 + f(D) - D_2 \) together for unity Lewis number flames by extending an existing RANS model [22, 34, 44, 47, 48, 51] in the following manner:

\[
T_4 + f(D) - D_2
\]

\[
T_4 + f(D) - D_2 = q_0 S^2 \frac{2L}{d th} = \Delta \approx 0.4 \delta_{th} (1\text{st column}), 1.6 \delta_{th} (2\text{nd column}), \text{and } 2.8 \delta_{th} (3\text{rd column}) \text{ in cases A–E (1st–5th row). All the terms are normalized with respect to } \rho_0 S^2 / \delta_{th}^2.
\]

Figure 6. Variations of \([T_4 + f(D) - D_2]\) (—) and \([T_4]_{sg} - [D_2]_{sg} + [f(D)]_{sg}\) (—) conditionally averaged in bins of \( \epsilon \) along with the predictions of Eqs. (15i) and (15ii) (—) and Eq. (16) (—) for \( \Delta = 0.4 \delta_{th} \) (1st column), 1.6 \delta_{th} (2nd column), and 2.8 \delta_{th} (3rd column) in cases A–E (1st–5th row). All the terms are normalized with respect to \( \rho_0 S^2 / \delta_{th}^2 \).
\[
T_4 - D_2 + f(D) = (T_4)_{res} - (D_2)_{res} + \{f(D)\}_{res} - (1 - f_{TD}) \beta_3 (\tilde{c} - c^*) \frac{\left[\tilde{N}_c - \tilde{D} \nabla \tilde{c} \cdot \nabla \tilde{c}\right]^2}{\tilde{c}(1 - \tilde{c})} \quad (15i)
\]

where \( \beta_3 = 5.7; c^* = 1.0 - 0.83 \text{erf} \left[0.5 \frac{\Delta S_L}{\alpha_{T0}} - 2.3\right] \) and \( f_{TD} = \exp \left[ -0.27 \left(\frac{\Delta S_L}{\alpha_{T0}}\right)^{1.7}\right] \); \( (15ii) \)

The involvement of \((\tilde{c} - c^*)/[\tilde{c}(1 - \tilde{c})]\) in Eq. (15i) is required for capturing the qualitative behavior of \([T_4 - D_2 + f(D)]\) across the flame brush, whereas \( f_{TD} \) approaches unity for small values of \( \Delta \) as the terms get fully resolved (i.e., \( \lim_{\Delta \to 0} [T_4 - D_2 + f(D)] = \lim_{\Delta \to 0} [(T_4)_{res} - (D_2)_{res} + \{f(D)\}_{res}] \)). The transition from positive to negative contribution of \([T_4 + f(D) - D_2]\) with increasing \( \Delta \) has been accounted for by \( c^* \). The predictions of Eq. (15i) are shown in Figure 6, which shows that this model captures both the qualitative and quantitative behaviors of \([T_4 + f(D) - D_2]\) for the \( Le = 1.0 \) cases considered here (e.g., cases C–E); however, this model underpredicts the magnitude of \([T_4 + f(D) - D_2]\) significantly for the \( Le \ll 1.0 \) cases (e.g., cases A and B). It is worth noting that the model given by Eq. (15i) does not account for the increased magnitude of \([T_4 - D_2 + f(D)]\) for small values of \( Le \) (see Table 3); hence, perhaps it is not surprising that this model underpredicts the magnitude of \([T_4 + f(D) - D_2]\) for the flames with \( Le \ll 1.0 \) (e.g., cases A and B). The increased magnitude of \([T_4 + f(D) - D_2]\) for the small values of \( Le \) is accounted for by modifying Eq. (15i) in the following manner:

\[
T_4 - D_2 + f(D) = (T_4)_{res} - (D_2)_{res} + \{f(D)\}_{res} - (1 - f_{TD}) \beta_3' (\tilde{c} - c^*) \frac{\left[\tilde{N}_c - \tilde{D} \nabla \tilde{c} \cdot \nabla \tilde{c}\right]^2}{\tilde{c}(1 - \tilde{c})} \quad (16)
\]

with \( \beta_3' = 5.7 Le^{-0.2} \)

where \( c^* \) and \( f_{TD} \) are considered according to Eq. (15ii). The predictions of Eq. (16) are shown in Figure 6, which demonstrates that Eq. (16) captures both the qualitative and quantitative behaviors of \([T_4 + f(D) - D_2]\) for a range of filter widths for different \( Le \) cases considered here. Equations (15i) and (16) become equal to each other for \( Le = 1.0 \) and thus their predictions cannot be separated from each other in case D in Figure 6.

It is worth noting that the combined contribution of the terms \( D_1, T_4, f(D), \) and \( -D_2 \) can be expressed in the following manner if the SDR transport equation is derived based on the kinematic form of the progress variable transport equation (i.e., \( Dc/DT = S_d \nabla c \)) [34, 47]:

\[
D_1 + T_4 - D_2 + f(D) \approx -2 \tilde{D} \nabla \cdot (\rho S_d \tilde{n} \nabla c) / |\nabla c|^2 + 2D \rho S_d \nabla \cdot \tilde{n} \nabla c \quad (17)
\]

where \( S_d = [\tilde{S} \nabla \cdot (\rho D \nabla c)] / (\rho |\nabla c|) \) and \( \tilde{n} = -\nabla c / |\nabla c| \) are the flame displacement speed and local flame normal vector, respectively. Thus, Eq. (17) suggests that the net contribution of \([T_4 - D_2 + f(D)]\) originates due to flame normal propagation and flame curvature. This justifies the modeling of these terms together [21, 34, 44, 47, 48, 53] because the molecular diffusion term \( D_1 \) is a closed term. Although Eq. (16) reasonably captures the qualitative behavior and the magnitude of \([T_4 + f(D) - D_2]\) for all cases considered here, the collective modeling of the terms \( T_4, f(D), \) and \( -D_2 \) may give rise to the loss of their individual physical significances. However, this is one of the first attempts to model the Lewis number effects on the SDR transport equation terms in the context of LES of premixed combustion and thus there is a scope for further improvement of this modeling in the future.

### 3.5. Implications of model implementation

The newly proposed models for the unclosed terms of the SDR \( \tilde{N}_c \) transport equation are summarized in Table 4 for the future potential users of these models. It is worth noting that the flamelet assumption is invoked while deriving these models, so they are expected to remain valid in the corrugated flamelets and thin reaction zones regimes of turbulent premixed combustion [55]. The scaling estimates in Table 3 indicate that the terms \( T_2, T_3, T_4, (-D_2), \) and \( f(D) \) remain leading-order contributors
to the SDR $\tilde{N}_c$ transport and the magnitude of $T_1$ remains negligible compared with the terms $T_2$, $T_3$, $T_4$, $(-D_2)$, and $f(D)$ irrespective of Damköhler and turbulent Reynolds numbers. This is consistent with the observations made from Figure 2. However, the turbulent transport term $T_1$ still needs to be modeled and included in the model implementation for LES for numerical stability.

### 4. Conclusions

The effects of global Lewis number $Le$ on the modeling of the unclosed terms of the transport equation of Favre-filtered SDR $\tilde{N}_c$ have been analyzed based on a priori analysis of a DNS database of freely propagating statistically planar turbulent premixed flames with $Le$ ranging from 0.34 to 1.2. It has been found that $Le$ has profound influence on the statistical behavior of the unclosed terms of $\tilde{N}_c$ transport arising from turbulent transport $T_1$, density variation due to heat release $T_2$, alignment of scalar and velocity gradients $T_3$, correlation between the gradients of reaction rate and reaction progress variable $T_4$, molecular dissipation $(-D_2)$, and diffusivity gradients $f(D)$, and detailed physical explanations have been provided for the observed non-unity Lewis number effects. Recently proposed models for $T_1$, $T_2$, $T_3$, $T_4$, $(-D_2)$, and $f(D)$ for unity Lewis number flames have been extended here to account for the effects of $Le$ based on the scaling estimates of these unclosed terms [28]. The newly proposed models have been found to satisfactorily predict the unclosed terms obtained from explicitly filtered DNS data for a range of $\Delta$ for different values of $Le$. However, it is still essential to implement these models into actual LES simulations for the purpose of a posteriori assessment. Moreover, these models need to be further validated based on detailed chemistry-based DNS simulations. Further validation of these models will form the basis of future investigations.
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