Scalar fluctuation and its dissipation in turbulent reacting flows

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The dissipation rate of a scalar variance is related to mean heat release rate in turbulent combustion. Mixture fraction is the scalar of interest for non-premixed combustion and a reaction progress variable is relevant for premixed combustion. A great deal of work is conducted in past studies to understand the spectra of passive scalar transport in turbulent flows. A very brief summary of these studies to bring out salient characteristics of passive scalar spectrum is given first. Then, the classical analysis of reactive scalar spectrum is revisited in the lights of recent understanding gained through analyzing the scalar spectrum deduced from direct numerical simulation data of both non-premixed and premixed combustion. The analysis shows that the reactive scalar spectral density in premixed combustion has a dependence on Karlovitz and Damköhler numbers, which comes through the mean scalar dissipation rate appearing in the spectral expression. In premixed combustion, the relevant scale for the scalar dissipation rate is shown to be of the order of the chemical length scale and the dissipation rate is not influenced by the scales in the inertial-convective range unlike for the passive scalar dissipation rate. The scalar fluctuations produced near the chemical scales cascade exponentially to larger scales. These observations imply that the passive scalar models cannot be extended to premixed combustion.

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I. INTRODUCTION

The mixing of fuel and air in non-premixed turbulent combustion plays vital role in sustaining the combustion processes and the rate of this mixing is influenced by turbulence, molecular diffusion and also combustion itself. The influence of combustion comes through thermal expansion effects on turbulence and turbulence-chemistry interaction. Both turbulence and combustion are multiscale phenomena and hence one can expect a cross-scale influence of one another. However, Bilger[1] pointed out that the mixing rate at small scales directly influences combustion by deducing that the instantaneous reaction rate per unit volume for species i is given by $\dot{\omega}_i = -N_\xi d^2 Y_i / d\xi^2$, where ξ is a chemically conserved scalar, known as mixture fraction. The mass fraction of species i is Y_i and the small-scale mixing rate is denoted by N_ξ and is defined as $N_\xi = \mathscr{D}(\nabla\xi \cdot \nabla\xi)$, where $\mathscr{D}$ is the molecular diffusivity which is taken to be the same for all the species involved in the combustion process. This small scale mixing rate, N_ξ, is commonly known as the scalar dissipation rate of the mixture fraction. Bilger[2] showed that the combustion and associated heat release effects are local in non-premixed combustion and thus the theories and models developed for passive scalar transport in non-reacting turbulence can be employed for non-premixed combustion.

The central role of small-scale mixing in turbulent premixed combustion was shown by Bray.[3,4] The rate of this mixing between hot and cold mixtures is signified by the scalar dissipation rate of reaction progress variable, c, which may be defined using either a scalar mass fraction or temperature. Also, the reaction progress variable is not a conserved or passive scalar like ξ and its dissipation rate is defined as $N_c = \mathscr{D}_c(\nabla c \cdot \nabla c)$. Hereafter, we shall drop the subscript for $\mathscr{D}$ and interpret it in the context of the scalar under consideration. The scalar dissipation rate is not only influenced by turbulence but also by combustion, molecular diffusion and the interactions among these physical processes.[5–7] Hence, the theory and modeling of passive scalar transport in turbulence cannot be extended directly to premixed combustion and this shall be further asserted through the scalar spectrum analysis discussed in the next section. Also, the heat release in premixed combustion affects the kinematic relationship between the turbulence and scalar fields.[8–11] The scalar gradient vector aligns with the most extensive principal strain rate in premixed combustion when the heat release effects overwhelms turbulent straining whereas it aligns with the most compressive principal strain rate in non-premixed combustion[12] and for passive scalar transport.[13–15] This possible change in the alignment brings leading order effects to the scalar mixing (scalar dissipation rate) physics and its modeling.[16,17]

Typically, the scalar dissipation rate of a passive scalar such as mixture fraction is estimated using a linear relaxation model written as $\widetilde{N}_\xi \simeq C_\xi \sigma_\xi^2 / \tau_t$, where the $\sim$ represents density weighted average for Reynolds-Averaged-Navier-Stokes (RANS) calculations or density weighted filtered value for a large eddy simulation (LES). The symbols σ_ξ^2 and τ_t represent variance of ξ and turbulence timescale respectively. This model introduced first by Corrsin[18] is valid when the molecular diffusion is smaller than the turbulent dispersion of scalar concentration.[19] This condition is satisfied in many reacting and non-reacting turbulent flows and thus, the above linear relaxation model is commonly used for RANS calculations. Also, it can be extended to subgrid dissipation rate in LES with dynamic procedure[20] to estimate the model constant, C_ξ, with an understanding that σ_ξ^2 and τ_t implying the subgrid variance and turbulence timescale respectively. The inadequacy of this model for premixed combustion was demonstrated in past studies for RANS[7,21] and LES[22–25] calculations. This is because of the strong influence of combustion effects on turbulence, scalar-turbulence interaction and their feedback on combustion.

A reasonable way to include these close coupling into the modelling of the scalar dissipation rate is to deduce a model through careful analysis of the transport equation for N_c. Furthermore, the scalar dissipation rate is required in almost all of combustion modeling approaches in one form or another. For example, the mean or filtered scalar dissipation rate is required for eddy breakup model and the approaches like conditional moment closure[26] and transported joint scalar PDF[27–31] (probability density function) require conditional dissipation rate. The flame surface density is also known to be related to the conditional dissipation rate.[32] Professor O′Brien has made seminal contributions to the topic of scalar mixing in turbulent flows with and without chemical reactions, specifically in the views of PDF approach. Hence, we think that it is appropriate to revisit the problem of scalar mixing in turbulent reacting flows to bring together recent understandings and developments.

The aim, here, is not to do an exhaustive and broader review but to revisit the classical ideas put forward by O′Brien, Corrsin and others for passive and reactive scalar spectra, and thereby the scalar dissipation rate, in combusting flows of engineering interest. This is conducted specifically in the light of recent findings on the scalar mixing physics in reactive flows. This paper is organised as follows. The scalar spectra are discussed in the next section. The scalar dissipation rate appearing in the scalar spectrum needs to be modeled which is discussed in Section III. Some new interpretation of reactive scalar spectrum is discussed in Section IV and the conclusions are summarised in the final section.

II. SCALAR AND ITS DISSIPATION SPECTRA

The spectral density, $E_q(k,t)$, of scalar fluctuation contains many important characteristics of the scalar field, q, representing either the mixture fraction or reaction progress variable. The integral of this spectral density over all the possible wavenumbers, k, yields the scalar variance, σ_q^2 which is given by the two-point correlation function $R_q(\mathbf{r} = 0)$. For variable density flows, this correlation function is defined as[33]

$$R_q(\mathbf{r}) = \frac{\overline{[\rho(\mathbf{x})q''(\mathbf{x})q''(\mathbf{x}+r)) + \rho(\mathbf{x}+r)q''(\mathbf{x}+r)q''(\mathbf{x}))]}}{2\overline{\rho}},$$

where q'' is the fluctuation of q over its density weighted mean, $\widetilde{q}$, and ρ is the local fluid density. The spectral density is given by

$$E_q(k,t) = \int_{-\infty}^{\infty} \Phi_q(\mathbf{k},t)\delta(|\mathbf{k}| - k)\ d\mathbf{k}, \tag{1}$$

where Φ_q is the spatial Fourier transform of R_q and $\mathbf{k}$ is the wavenumber vector. The spectral density obeys the following equation when the scalar and turbulence fields are homogeneous[18,25,34]

$$\frac{\partial E_q}{\partial t} = T(k,t) - 2\mathscr{D}k^2 E_q + 2\mathscr{F}\left(\widehat{\dot{\omega}}_q\right), \tag{2}$$

where $\mathscr{F}\left(\widehat{\dot{\omega}}_q\right)$ is the spectral density of the reaction related terms appearing in the transport equation for $\sigma_q^2 = R_q(0)$ (see equation 3.40 of Kolla et al.[33]) The term $T = -\partial\left[s(k,t)E_q\right]/\partial k$ is the spectral transfer of scalar fluctuations from one wavenumber to another. The quantity sE_q is the spectral flux across a wavenumber k.

For a passive scalar like mixture fraction, the reaction rate is zero and thus Eq. (2) becomes $\partial\left[sE_\xi\right]/\partial k = -2\mathscr{D}k^2 E_\xi$ for a stationary state and the right-hand-side of this expression coming from the molecular diffusive flux in $\mathbf{r}$ space is expected to contribute for large wavenumbers. Hence, this expression simplifies further for wavenumbers in the inertial-convective range, yielding $E_\xi \sim s^{-1}$ and $s(k)$ is obtained through physical consideration. This has been done in past studies for Schmidt numbers (defined as $Sc = \nu/\mathscr{D}$, where ν is the kinematic viscosity) of the order one, larger and smaller than unity.[35–39] The results of these studies are summarised in Fig. 1 and it is important to note that the above deduction is valid only for the intertial-convective and viscous-convective ranges of wavenumbers marked in this figure.[40] These past studies suggested that $s \sim \varepsilon^{1/3}k^{5/3}$ and $k\sqrt{\varepsilon/\nu}$ for the inertial-convective and the viscous-convective ranges respectively, where ε is the dissipation rate of the turbulent kinetic energy, K. Hence, the well-known behaviours $k^{-5/3}$ and k^{-1} are observed for the inertial-convective and viscous-convective

ranges,[35–38] which were also verified through direct numerical simulations (DNSs).[40,41] However, for the inertial-diffusive range, where the viscous effects are negligible but the diffusive effects are important, the spectral balance is $\partial \left[s E_\xi \right] / \partial k = -2 \mathscr{D} k^2 E_\xi$. By using the expression for s in the inertial-convective range in this spectral balance, one can deduce that

$$E_\xi = A_\xi \varepsilon^{-1/3} k^{-5/3} \exp \left[\frac{-3 A_1}{2} \left(\frac{k}{k_{oc}} \right)^{4/3} \right],$$

(3)

where A_ξ is a constant which is evaluated using the dissipation rate of the scalar variance, $\chi_\xi = 2 \overline{\rho \mathscr{D} \left(\nabla \xi'' \cdot \nabla \xi'' \right)} / \overline{\rho} = 2 \mathscr{D} \int k^2 E_\xi \, dk$ and this gives $A_\xi = 2 \chi_\xi / 3$ for the inertial-convective range.[18,37] The factor $2/3$ is known as the Obukhov-Corrsin constant. For $k \gg k_{oc}$ (viscous-diffusive range), there is an exponential decrease of E_ξ from $k^{-5/3}$ with a portion showing $k^{-17/3}$ behaviour.[34] The wavenumber corresponding to the Obukhov-Corrsin length scale is denoted using $k_{oc} = \left(\varepsilon / \mathscr{D}^3 \right)^{1/4}$ and is related to the Kolmogorov wavenumber, k_k, through $k_{oc} = k_k Sc^{3/4}$. Chemically reacting turbulent flows with $Sc \ll 1$ are rare for terrestrial conditions and applications. Although there are practical applications such as mixing of two liquids with chemical reactions satisfying $Sc \gg 1$ condition, we limit ourselves to the case of $Sc \sim O(1)$ by considering engineering and technological applications involving gaseous flows.
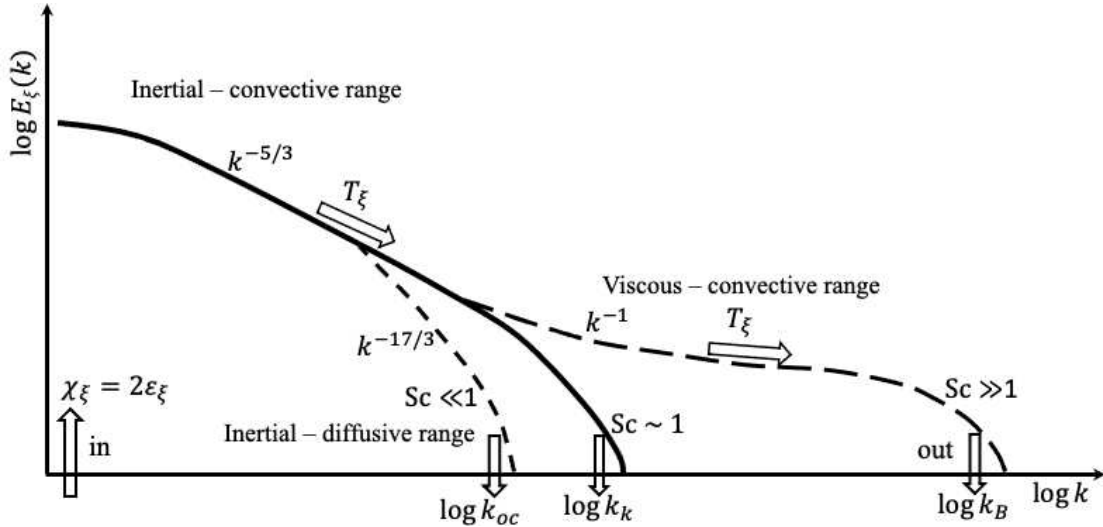

FIG. 1. Schematic of stationary passive scalar spectrum, E_ξ, for three limiting cases.[40] Adopted with permission from the book Ten chapters in turbulence 2012. Copyright 2012 Cambridge University Press.

The assumption of homogeneous flow used while writing Eq. (2) can be questioned for reacting flows, specifically for premixed combustion. However, the shapes of velocity and scalar

spectra computed using DNS data of non-premixed[42] and premixed[33] flames (involving one-step and multi-step chemical kinetics) suggest that it may be reasonable to make this approximation to deduce further insights into the spectral behaviour of scalar fluctuations in reacting flows. Also, velocity and scalar dissipation spectra deduced using DNS data of premixed flames lend support for this view.[43]. Hence, we shall use Eq. (2) for further analysis here. However, one must be cautious in interpreting and applying the results and insights discussed in this paper and the above conditions should be borne in mind.

Since the mass fraction of a reactive scalar, Y_i, in non-premixed combustion can be expressed as a liner function of mixture fraction, if all the molecular diffusivities are the same and the chemical timescale is very much shorter than a typical turbulent timescale, one can see that Y_i spectra are directly related to the mixture fraction spectrum. However, combustion introduces an additional length scale, which can be of the order of Kolmogorov scale or thinner. Hence, the reactive scalar spectra, despite its direct relation to the mixture fraction spectra, may not collapse together if one employs the diffusive scale such as Obukhov-Corrsin length scale which is commonly used for passive scalar spectrum.[42] Perhaps, the appropriate scale to use for the diffusive-reactive range, $k \gg k_{oc}$, may be the reaction zone thickness which can be defined[44] as $\delta_r \simeq \sqrt{2\mathscr{D}\Delta Z/\chi_\xi}$, where ΔZ is the mixture fraction space thickness over which the heat release rate drops to 10% of its maximum. This kind of scaling is yet to be explored to study the scalar spectra in non-premixed combustion. However, Knaus and Pantano[42] showed that the Kolmogorov scaling holds well for the inertial-convective range if one uses density weighted quantities for the scaling.

The classical works by Corrsin[18], O'Brien[45] and Pao[34] showed the influence of first-order chemical reaction on the scalar spectrum under dilute conditions (heat release effects are small). Furthermore, Pao showed[34] the need for the numerical solutions when introducing reversible reaction and, series and parallel reactions in the analysis but those reactions did not change the spectral characteristics. In the following analysis, we shall revisit those analyses in the light of recent understandings of reactive scalar spectra deduced using DNS data of premixed combustion with realistic finite rate chemical reactions (multi-step chemical kinetics) and large heat release rates. These DNS studies showed that the pressure-dilation work, denoted by $\overline{p'\nabla \cdot \mathbf{u}''}$, played leading order role for the turbulent kinetic energy budget in premixed flames.[46,47] Thus, the velocity spectrum depicted the due influence of this effect at the wavenumber corresponding to a laminar flame thickness along with a corresponding influence in scalar spectra also.[33] However, the shape of the spectrum, specifically for wavenumbers sufficiently smaller than the wavenumber corresponding

to the laminar flame scale (chemical length scale), remained similar to those observed in constant density flows.

For a reaction $F + vO \rightarrow (1+v)P$, where v is the oxidiser mass per unit mass of fuel, with a reaction rate expression $\dot{\Omega}_F = -K_r[F][O]$, one can write $\dot{\Omega}_F = -[F]/\tau_c$ for fuel-lean combustion systems and τ_c is a chemical timescale for the reaction and $[F]$ is the molar density of the species F. Hence, approximating the combustion reaction as a first-order reaction (with respect to the species of interest) is acceptable although its overall order is two. Even if one considers the Arrhenius rate expression for K_r then the exponential term involving temperature or τ_c can be estimated for the temperature, T, corresponding to the maximum reaction rate (or heat release rate). If one considers the full exponential dependence on temperature for K_r then one must consider the cross-correlation between F and T and the corresponding spectra under the above approximation for the reaction rate, otherwise the cross-correlations of F-O, F-T and O-T, and their spectra must be included in the analysis. If one considers a multi-step chemical kinetics then additional cross-correlations among scalars will arise as demonstrated by Pao.[34] These additional cross-spectra and the exponential non-linear term for temperature will severely limit the analytical treatment to deduce further understanding of reactive scalar spectra. Also, it is common to express reaction rate per unit volume using a reaction progress or a regress variable in premixed combustion. Thus, the subscript F used in the discussion below implies the reaction regress variable, which takes 1 in the unburnt mixture and 0 in the burnt mixture. This variable is related to the progress variable through $c = 1 - F$. With the above points noted, the spectral density of the reaction related term becomes $\mathscr{F}(\widehat{\dot{\omega}}_F) = -E_F/\tau_c$. Now, Eq. (2) gives

$$\frac{dsE_F}{dk} = -2\mathscr{D}k^2 E_F - 2\frac{E_F}{\tau_c} \tag{4}$$

for the stationary spectral density of F. It is to be noted that the dissipation contribution is also included along with the reaction related term since the coupling between reaction rate and molecular diffusion is strong in premixed combustion. We have $s \sim \varepsilon^{1/3}k^{5/3}$ for the case of our interest and thus one gets

$$E_F(k) = B\varepsilon^{-1/3}k^{-5/3}\exp\left[\frac{-3A_2}{2}\left(\frac{k}{k_{oc}}\right)^{4/3} + 3A_2\left(\frac{k}{k_{oc}}\right)^{-2/3}\left(\frac{k_{ch}}{k_{oc}}\right)^2\right], \tag{5}$$

where $k_{ch} = (\mathscr{D}\tau_c)^{-1/2}$ is the wavenumber corresponding to the chemical length scale, which is the Zeldovich thickness. It is also worth noting that $k_{ch}/k_{oc} = Ka^{-1/2}Sc^{-3/4}$, where $Ka = (k_k/k_{ch})^2$

is the Karlovitz number. The dimensional constant B can be evaluated to be $B = \chi_F/(3I)$ using the identity $\chi_F = 2\mathscr{D} \int k^2 E_F \, dk$, where I is given by the integral

$$I = \int_{x_l}^{\infty} x \, \exp\left[\frac{-3A_2}{2}x^2 + \frac{3A_2}{xKa}\right] \, dx, \tag{6}$$

with $x = (k/k_{oc})^{2/3}$. Hence, E_F given in Eq. (5) becomes

$$E_F(k) = A_F \chi_F \varepsilon^{-1/3} k^{-5/3} \exp\left[\frac{-3A_2}{2Sc}\left(\frac{k}{k_k}\right)^{4/3} + \frac{3A_2}{KaSc}\left(\frac{k}{k_k}\right)^{-2/3}\right], \tag{7}$$

with $A_F = 1/(3I)$, which is expected to be of the order one. One observes that when Ka becomes very large (passive scalar situation) the passive scalar spectrum, E_ξ, given earlier for $Sc = 1$ is recovered. It is understood that the statistical quantities appearing in Eq. (7) are density weighted averages. We shall discuss the need to have the lower limit as x_l rather than as zero after studying the characteristics of E_F.
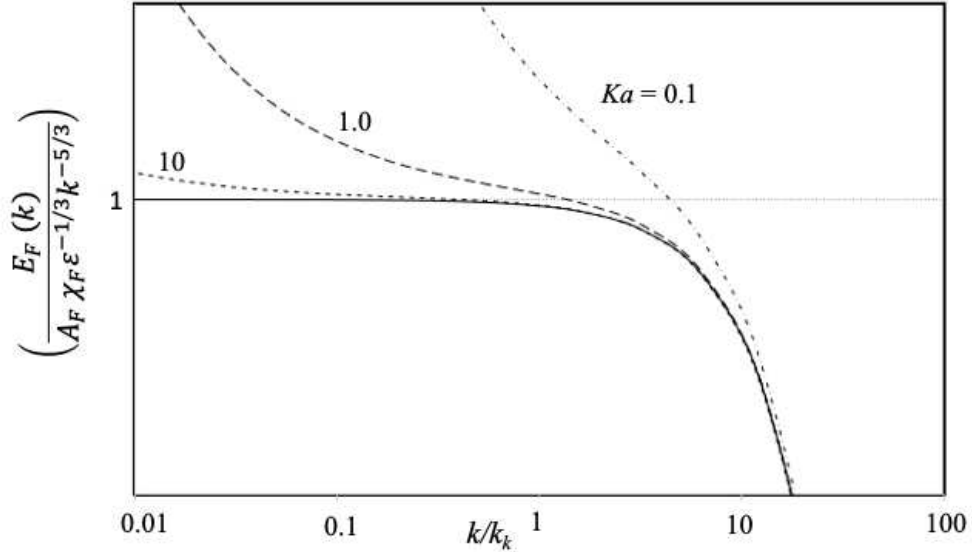

FIG. 2. Typical variation of normalised E_F with k/k_k in log-log scale. The solid line is for non-reactive case ($Ka \rightarrow \infty$) and the other three lines are for specific Ka values given above and $Sc = A_2 = 1$.

Figure 2 shows variation of $E_F(k)$ normalised using the quantities premultiplying the exponential function in Eq. (7) (also known as the compensated spectrum when A_F is excluded from the denominator) with k/k_k, where k_k is the Kolmogorov wavenumber. Hence, one shall see a flat line for $k/k_k \ll 1$ when there are no effects of molecular diffusion or combustion as shown by the solid line in Fig. 2. The diffusion effects are seen for $k/k_k \geq 1$ as expected for $Sc = 1$. Any

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deviation from this solid line is because of the exponential term arising from combustion and its effect on the scalar fluctuations at a scale 100 times larger than the Kolmogorov scale is felt quite strongly for the Ka values considered here when Sc and A_2 are unity. This influence diminishes when $k/k_k \geq 0.3$ for $Ka = 10$ and the combustion effects are felt even at a scale which is nearly 1/6th of the Kolmogorov scale for $Ka = 1$. This effect is extended up to $k/k_k \simeq 15$ for the smallest Ka considered here. These Ka values are chosen to be representative of wrinkled and corrugated flamelets, and thin reaction zones regimes of turbulent premixed combustion.[48] The collapse of these three curves into a single curve depends on how the dissipation rate, χ_F, depends on Ka and also the Damköhler number, Da, defined as the ratio of large-scale turbulence timescale to τ_c. This turbulence timescale is given by $\tau_t = \int E(k)\, dk / \left(2\nu \int k^2 E(k)\, dk\right) = K/\varepsilon$, where E is the turbulent kinetic energy spectrum. Furthermore, the integral I may depend on Ka and thus one can expect A_F to have some dependence on Ka. Since this integral is related to the density weighted mean scalar dissipation rate, one needs to understand the leading order processes influencing the scalar dissipation rate in turbulent premixed combustion before discussing Ka dependency of A_F. This discussion is provided in Section IV.

It is also noted that the scalar dissipation spectrum is related to the scalar spectrum through $D_F(k) = 2\mathscr{D}k^2 E_F(k)$ and the above comments on E_F will carry forward to D_F also. It has been shown in recent studies that χ_F has dependence on Da and Ka and these studies are reviewed briefly in the next section to bring out the key aspects in them to further understand the reactive scalar spectrum behaviour in premixed combustion. Also, it clear from the foregoing discussion that the statistical quantities such as the scalar dissipation rate in turbulent premixed combustion cannot be modeled by simply extending the passive scalar models because the combustion effects on the scalar fluctuation are felt strongly on a wide range of scales, and is driven by chemical reactions which are small scale phenomena. Indeed, this point will become clearer in Section IV.

III. SCALAR DISSIPATION RATE MODELING

The scalar dissipation rate, χ_F, appearing in Eq. (7) denotes the dissipation rate of Favre variance of F, $\sigma_F^2 = \overline{\rho F'' F''}/\overline{\rho}$, and it is defined as $\chi_F = 2\left(\overline{\rho \mathscr{D} \nabla F'' \cdot \nabla F''}\right)/\overline{\rho} = 2\varepsilon_F$. A transport equation for ε_F was derived by Swaminathan and Bray[7] and it is written as

$$\frac{D\overline{\rho}\varepsilon_F}{Dt} = D_1 - D_2 + T_1 + T_2 + T_3 + T_4 \tag{8}$$

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after ignoring the contributions coming from temperature dependence of $\mathscr{D}$. The operator D/Dt is the substantial derivative, the molecular diffusion and dissipation are denoted by $D_1 = \nabla \cdot \left(\overline{\rho \mathscr{D} \nabla \varepsilon_F} \right)$ and $D_2 = 2 \overline{\rho \mathscr{D}^2 \left(\nabla \cdot \nabla F'' \right)^2}$ respectively. The turbulent transport of the dissipation rate is represented by T_1. The effect of dilatation, when $Le = 1$, is represented by $T_2 = 2 \overline{\rho \varepsilon_F \nabla \cdot \mathbf{u}''}$ which results from the density change across the flame front and this term generally acts as a source term for ε_F evolution. The symbol Le is the Lewis number which is defined as the ratio of thermal diffusivity to mass diffusivity. The interaction of turbulence and scalar fields denoted by T_3 has three contributions and the inner product between the fluctuating scalar gradient and turbulent strain rate, given by $T_{32} = -2 \overline{\rho \mathscr{D} \left(\nabla F'' \cdot \nabla u'' \cdot \nabla F'' \right)}$, is the most dominant term of T_3. The influence of reaction rate is represented by $T_4 = 2 \left(\overline{\mathscr{D} \nabla F'' \cdot \nabla \dot{\omega}_F''} \right)$, where $\dot{\omega}_F''$ is the fluctuating mass-based reaction rate per unit volume. An order of magnitude analysis of Eq. (8) gave

$$T_2 + T_{32} + T_4 - D_2 \simeq 0 \tag{9}$$

as the leading order balance for high Da combustion[7]. This was verified through DNS data analyses for both high and low Da combustion and typical results are shown in Fig. 3.[16] These results are normalised using the respective laminar flame thermal thickness, δ_{th}, unburnt mixture density ρ_u and laminar burning velocity S_L. For fuel lean mixtures, the thermal thickness is a multiple of Zeldovich thickness, $\sqrt{\mathscr{D} \tau_c}$, defined earlier. The major contribution to T_3 was shown to come from the inner product term, T_{32}.[16] In non-reacting turbulent flows, the scalar gradient is known to align with the most compressive principal strain rate,[13–15] which is also the case for mixture fraction in non-premixed combustion,[12] and thus T_{32} acts as a source term for χ_ξ. On the other hand, T_{32} acts as a sink for χ_F in high Da premixed combustion because the scalar gradient aligns with the most extensive principal strain rate[8,9,11,16] and this difference cannot be ignored.

Simple models for the four terms in Eq. (9) were also proposed and verified using DNS data in past studies.[16,49,50] These models were also used to obtain an algebraic expression for ε_F,[17] which was also validated using experimental data for turbulent burning velocities.[51] This model can be written as

$$\chi_F = 2\varepsilon_F \simeq G \frac{\left(2K_c^* - \tau C_4 \right) Da + C_3}{\tau_t} = G \frac{\left(2K_c^* - \tau C_4 \right) + C_3/Da}{\tau_c} \tag{10}$$

for a location with significant dissipation rate (mean reaction rate) inside the flame brush, which would be the middle of the flame brush. The symbol G is a factor of the order one, τ is the heat release parameter, defined as the temperature rise, $T_b - T_u$, across the flame front divided by the unburnt mixture temperature, T_u, which has a value of 6 to 8 for typical combustion condi-
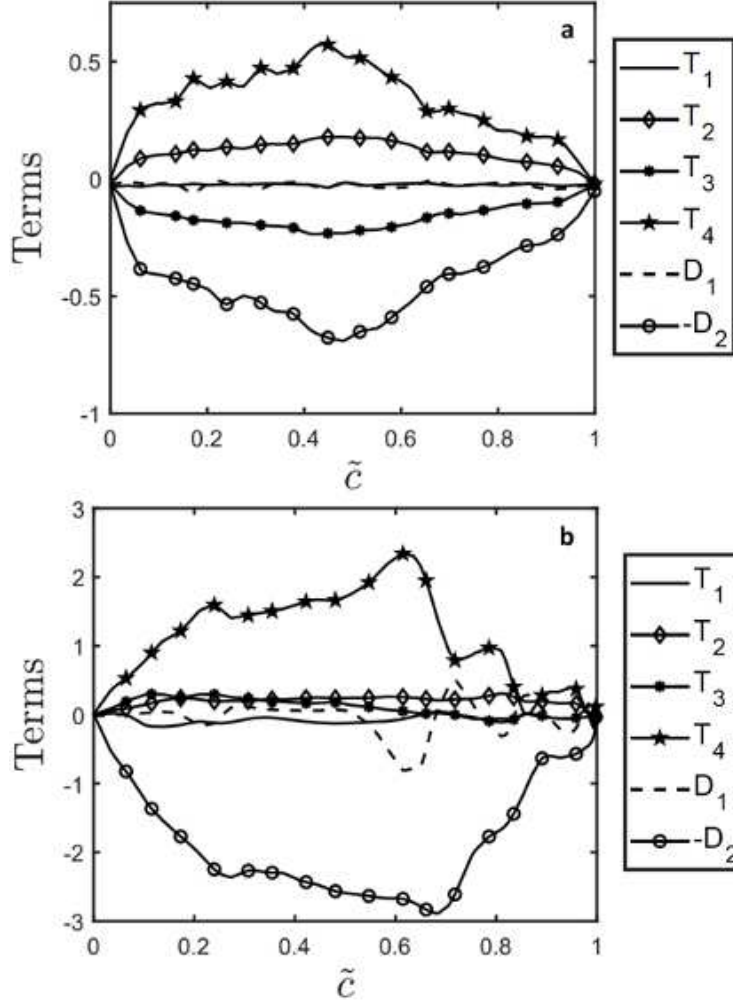
FIG. 3. Variations of $D_1, D_2, T_1, T_2, T_3, T_4$ across the flame brush, $\widetilde{C} = 1 - \widetilde{F}$. These terms are normalised using $\delta_{th} / (\rho_u S_L^2)$ and the results are shown for two cases (a) $Da = 6.8, Ka = 0.3$ and (b) 0.3, 11.[16] Reproduced from N. Chakraborty, J. W. Rogerson, and N. Swaminathan, "A-priori assessment of closures for scalar dissipation rate transport in turbulent premixed flames using direct numerical simulation," Phys. Fluids 20, 045106, 2008, with the permission of AIP Publishing.

tion, K_c^*/τ varying from 0.7 to 0.8 signify the contribution of T_2, and $\tau_t = K/\varepsilon$ is the timescale for large scale turbulence. The Damköhler number is related to Ka through $Da \simeq Ka^{-1}\sqrt{Re_t}$, where Re_t is the turbulence Reynolds number. The parameters $C_3 = 1.5\sqrt{Ka}/\left(1+\sqrt{Ka}\right)$ and $C_4 = 1.1\left(1+Ka\right)^{-0.4}$ have dependencies on the Karlovitz number.[17] These relations arise because the scalar gradient generation is driven by chemical reactions, turbulence, and turbulence-scalar interaction in premixed combustion, see Eq. (9). Indeed, the functional dependencies discussed above have come from DNS data analyses and seem to work quite well for a range of combustion

and turbulence conditions spanning from corrugated flamelets to thin reaction zones regimes of premixed combustion.[22–24] However, some minor changes to these functional dependencies were also reported in past studies when *Le* is not unity.[52,53] It is also to be noted that there are alternative approaches to model the scalar dissipation rate in premixed combustion which are reviewed by Chakraborty et al.[54]

IV. DISCUSSION

By substituting Eq. (10) into Eq. (7) one gets

$$\frac{E_F \tau_t}{\widehat{A}_F \varepsilon^{-1/3} k^{-5/3}} = f_1(Ka) \exp\left[\frac{-3A_2}{2Sc}\left(\frac{k}{k_k}\right)^{4/3} + \frac{3A_2}{Ka\,Sc}\left(\frac{k}{k_k}\right)^{-2/3}\right], \tag{11}$$

or

$$\frac{E_F \tau_c}{\widehat{A}_F \varepsilon^{-1/3} k^{-5/3}} = f_2(Ka) \exp\left[\frac{-3A_2}{2Sc}\left(\frac{k}{k_k}\right)^{4/3} + \frac{3A_2}{Ka\,Sc}\left(\frac{k}{k_k}\right)^{-2/3}\right], \tag{12}$$

where $\widehat{A}_F = A_F G$. Equation (10) suggests that $f_2(Ka)$ is $\left(0.5\tau + C_5 Ka^{3/2}/\left(1+\sqrt{Ka}\right)\right)$ and $f_1(Ka) = Da\,f_2(Ka)$. Now, it is becoming clear that the full scaling of scalar spectrum in turbulent premixed combustion involves both turbulence and chemical scales as contemplated by Kolla et al.[33] The above scaling on *Ka* is based on the analysis of turbulent premixed flames of hydrocarbon– and hydrogen–air mixtures in past studies which considered statistically 1D propagating flame in spatially decaying turbulence,[8,16,17,52,55] oblique flames,[56–59] spherically expanding flames,[60,61], slot-jet flames,[62] and swirling flames.[63,64] These studies used both single and multi-step chemical kinetics, and also unity and non-unity Lewis numbers. Hence, the above scaling may be considered to be quite robust. However, the ranges of *Da* and *Ka* spans a narrow region in the corrugated flamelets and thin reaction zones regimes of turbulent combustion diagram, and hence it may be prudent to consider the above scaling as tentative because of the limited ranges of *Da* and *Ka* values.

The constant A_F in Eq. (7) involves the integral in Eq. (6) and this integral will not converge if $x_l = 0$ because of the $1/x$ in the exponent. This becomes severe for low *Ka* (combustion in wrinkled flamelets regime). In his seminal analysis, Corrsin[18] suggested to consider from the 'energy-bearing' wavenumber to the end of the inertial-convective range to evaluate the parameter in the scalar spectrum. Furthermore, the reaction was taken to occur in the inertial-convective range (see his Eq. 19), which is possible only if the reaction is a unimolecular reaction. Combus-

tion reaction is generally bi-molecular and it can be approximated to be first-order with respect to the scalar of interest as discussed in Section II.

Numerical analysis of the integral in Eq. (6) suggests the following. To keep A_F to be of the order one, x_l must be kept as $1.26 Ka^{-1/3}$ which comes out to be the solution of $x_l - \exp\left[-1.5x_l^2 - 3/(x_l Ka)\right] = 0$, where A_2 and Sc are takend to be 1 for simplicity and without the loss of generality. This variation of x_l shown in Fig. 4 suggests that k_l/k_k varies from 4.46 for $Ka = 0.1$ to 0.06 for $Ka = 500$ for the integral in Eq. (6) to converge (have a finite value) and to have A_F of the order one. The values of A_F depends weakly on Ka as shown in Fig. 4. If one takes the lower limit to be 100 times larger than the Kolmogorov scale, see Fig. 2, then the integral in Eq. (6) become so large implying that the scalar dissipation rate, χ_F, becomes unphysical and A_F becomes 0 for Ka below 3 and this variation of A_F with Ka is also shown in Fig. 4.
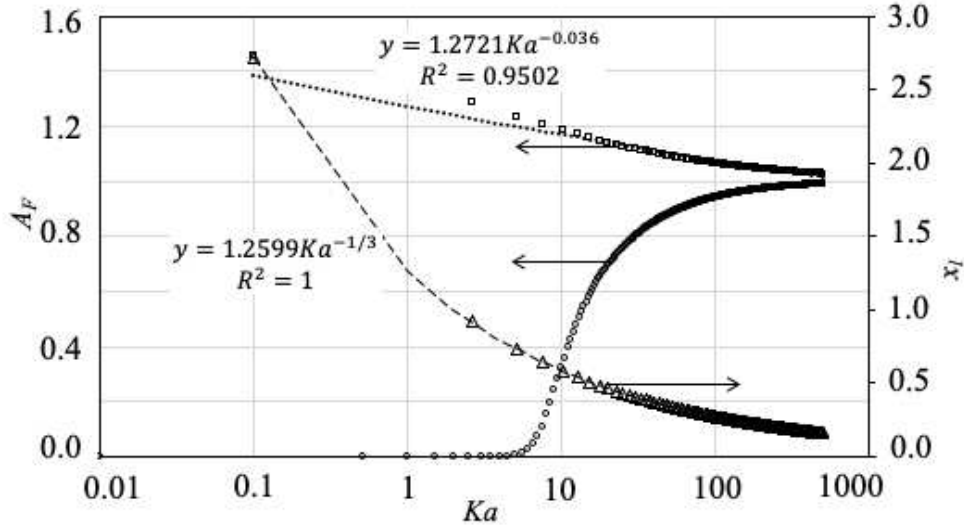

FIG. 4. Variation of A_F and x_l with Ka. Symbols are from numerical computations and lines show the best fits.

The choice of having $x_l \sim Ka^{-1/3}$ seem to be physically meaningful and this brings out an important point. If one translates this in terms of chemical length scale then the lower wavenumber becomes $k_l/k_{ch} = \sqrt{Ka}\,(k_l/k_k) = 1.41$, suggesting that the relevant largest length scale is of the order of the chemical length scale for the dissipation field. Kolla et al.[43] also suggested that a chemical scale is the relevant scale for the dissipation rate by analysing the dissipation spectra deduced using DNS data of premixed flames. Furthermore, the scalar fluctuations produced by the reactions around the chemical length scale cascade exponentially to larger scales (see Eq. 11 or 12)

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as seen in Fig. 2. The rate of this cascade is larger if the combustion is stronger (small Ka or large Da). The length scales in the inertial-convective range and below do not influence the dissipation rate in premixed combustion. This is also signified by the leading order balance in Eq. (9) involving only the reaction rate related terms, except T_{32}. Furthermore, this T_{32} term involves gradients of scalar and velocity fluctuations and thus it is also related to small scales. This observation holds on a physical basis also because the reactant mixture is homogeneous everywhere except in reacting regions of premixed combustion and hence turbulence alone cannot produce scalar gradients. Chemical reactions, which are small-scale phenomena, produce scalar gradients and the turbulence either enhances or hinders this production. Hence, using the passive scalar mixing based models for turbulent premixed combustion is questionable on physical grounds. This is also clear from the spectral considerations discussed in this study. This observation is highly relevant for LES of premixed combustion since SGS modeling involves small scale quantities.

V. SUMMARY AND CONCLUSION

The spectral characteristics of scalar fluctuations and their dissipation in turbulent combustion is of long standing interest. The seminal work by Corrsin, O′Brien, and Pao gave useful insights and raised many intriguing questions, for which there are no convincing answers yet. Spectral analysis using DNS data of non-premixed combustion showed that the Kolmogorov scaling worked quite well if the density weighted quantities are used in the scaling.[42] The pressure dilatation work was shown to have a due influence on the turbulent kinetic energy spectrum in premixed combustion along with a spike appearing in scalar spectrum due to reaction rate.[33] However, the full scaling of the scalar spectrum couldn't be achieved and it may involve Kolmogorov scale, laminar flame thickness, Karlovitz, Ka, and Damköhler, Da, numbers.

This study shows that the scalar spectrum, E_F, in premixed combustion involves Karlovitz and Damköhler numbers which are related to both chemical and turbulence scales. This is deduced using spectral analysis following the classical work of Corrsin.[18] The dependence of E_F on Ka and Da is observed to arise through the mean scalar dissipation rate. It is also shown that the relevant scale for the dissipation rate is of the order of the chemical length scale and the dissipation rate in premixed combustion is not influenced by the scales in the inertial-convective range, which is contrastingly different from the case of passive scalar mixing in turbulence. Also, scalar fluctuations produced near the chemical scales cascade exponentially to larger scales. Although a

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tentative functional dependence on Ka and Da is obtained in this study, which may hold in general, a suitable form of this scaling to collapse the entire scalar spectral density over a broad range of Da and Ka values is still to be found and further analysis would be helpful in that regard.

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DATA AVAILABILITY

Data sharing is not applicable to this article as it involves theoretical analysis and no new data was created or analyzed in this study.

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