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Dissipation-scale fluctuations and mixing transition in turbulent flows

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A small separation between reactants, not exceeding $10^{-8} - 10^{-7}$ cm, is the necessary condition for various chemical reactions. It is shown that random advection and stretching by turbulence leads to the formation of scalar-enriched sheets of strongly fluctuating thickness η_c. The molecular-level mixing is achieved by diffusion across these sheets (interfaces) separating the reactigants. Since the diffusion time scale is $\tau_d \propto \eta_c^2$, knowledge of the probability density $Q(\eta_c, Re)$ is crucial for evaluation of mixing times and chemical reaction rates. According to Kolmogorov–Batchelor phenomenology, the stretching time $\tau_{eddy} \approx L/u_{rms} = O(1)$ is independent of large-scale Reynolds number $Re = u_{rms}L/\nu$ and the diffusion time $\tau_d \approx \tau_{eddy}/\sqrt{Re} \ll \tau_{eddy}$ is very small. Therefore, in previous studies, molecular diffusion was frequently neglected as being too fast to contribute substantially to the reaction rates. In this paper, taking into account strong intermittent fluctuations of the scalar dissipation scales, this conclusion is re-examined. We derive the probability density $Q(\eta_c, Re, Sc)$, calculate the mean scalar dissipation scale and predict transition in the reaction rate behaviour from $\mathscr{R} \propto \sqrt{Re}$ ($Re \leqslant 10^3 - 10^4$) to the high-Re asymptotics $\mathscr{R} \propto Re^0$. These conclusions are compared with known experimental and numerical data.

1. Introduction

Efficiency of chemical reactions and combustion crucially depends upon the number of reactant moles mixed on a molecular scale. For a non-negligibly small reaction rate, the separation between reacting species must not exceed $r \approx 10^{-8} - 10^{-7}$ cm, which is the main reason for the immense importance of the mixing process. Slow diffusion in laminar flows leads to extremely poor mixing and, being the most common mixing accelerator, hydrodynamic turbulence plays a vital role in natural and man-made processes in heat transfer, chemical transformations, combustion, meteorology and astrophysics. The mixing process in turbulent flows involves three main steps: (i) entrainment, creating pockets of material B in a turbulent flow enriched by a substance A; (ii) advection and stretching leading to formation of thin convoluted sheets of thickness η_c separating the reactants; this process is often related to as 'mixing by random stirring'; (iii) molecular diffusion across these 'dissipation sheets' on a time scale $\tau_d \approx \eta_c^2/D$. If chemical reaction and turbulent mixing processes are fast then molecular diffusion is the reaction rate-determining step (Dimotakis 1993, 2005). Investigation of the role of scalar dissipation and dissipation sheets has become an extremely active field; see, for example, excellent reviews by Peters (2000), Bilger (2004), Sreenivasan (2004) and Dimotakis (2005) and papers by Buch & Dahm (1998), Villermaux and colleagues (2001, 2003, 2006) and Celani *et al.* (2005).

This paper is organized as follows. In the remainder of this section we discuss the Kolomogorov–Batchelor phenomenology, stressing a well-known fact that the width of the dissipation ('laminar') sheets $\eta = \alpha_v \eta_K$ is substantially larger than the Kolmogorov scale η_K. The experimentally and numerically observed factor $\alpha_v \approx 10$. As a result, the diffusion time across laminar sheets is much larger than previously thought and, at $Re \approx \alpha_v^4$, the reaction rate undergoes transition from $\mathscr{R} \propto \sqrt{Re}$ to $\mathscr{R} \approx$ const. The rest of the paper is devoted to theoretical derivation of the crucially important factor α_v. In § 2, the dissipation anomaly for the scalar field, indicating strong fluctuations of the scalar dissipation scale η_c, is derived. The probability density $Q(\eta_c)$, leading to the mean scalar dissipation scale $\overline{\eta_c} = \int \eta_c Q(\eta_c)\, \mathrm{d}\eta_c \approx \alpha_v \eta_B$ with $\eta_v \approx 7 - 10$, is evaluated in § 4. The conclusions are presented in § 5.

1.1. *Kolmogorov–Batchelor phenomenology*

Based on a classic Kolomogorov cascade concept, we illustrate the main qualitative features of the mixing of two liquids (chemical components) A and B. (A quantitative dynamic description will be developed below.) Consider a turbulent flow of a fluid A. The integral scale of turbulence, separating the energy and inertial ranges, is L and the large-scale Reynolds number is $Re = u_{rms} L/\nu$. At a time $t = 0$, a blob of the same fluid enriched with chemical component B is placed in the flow. For simplicity, we assume the linear dimension of a blob $r_0 = L$. At this point, the area of the interface, separating A and B substances, is $O(L^2)$. The life- (turnover) time of this eddy is $T_0 \equiv \tau_{eddy} \approx L/u_{rms} \approx L^{2/3} \mathscr{E}^{-1/3}$, where u_{rms} is the r.m.s. value of the turbulent velocity. The magnitude of the energy flux across scales is $\mathscr{E} = \alpha_{\mathscr{E}} u_{rms}^3/L$ with $\alpha_{\mathscr{E}} \approx 0.8$–0.9. In accord with Kolmogorov's phenomenology, after $\approx T_0$ seconds, by nonlinear interactions, this eddy is transformed into another one of linear dimensions $L_{x,1} = L/2$ and $L_{y,1} = L_{z,1} \approx \sqrt{2} L$. Since the gradient in the x-direction is largest, the characteristic time scale of this, 'daughter' structure is: $T_1 \approx (L^{2/3}/\mathscr{E}^{1/3}) 2^{-2/3}$. Then, after $n \gg 1$ steps $L_{x,n} = 2^{-n} L$; $T_n \approx 2^{2n/3} (L^{2/3}/\mathscr{E}^{1/3})$; $\tau_{v,n} \approx 2^{-2n} (L^2/\nu)$, where the viscous time is denoted as τ_v. We see that the viscous time of a structure strongly decreases with the number of 'cascade steps'. The time required to form the smallest Kolmogorov eddies on the scale $\eta_K \approx L Re^{-3/4}$ is thus:

$$T_K = T_0 + T_1 + \cdots T_n \propto T_0 \approx \frac{L^{2/3}}{\mathscr{E}^{1/3}} \equiv \tau_{eddy} \tag{1.1}$$

where $n \approx (3/4)(\ln Re/\ln 2) \approx \ln Re \gg 1$. It is important that the Reynolds number $Re = u_{rms} L/\nu$ used in the above relations be distinguished from $Re_U = UL/\nu$ where U is the mean velocity in the flow. Typically, in canonical flows used for comparison of theoretical predictions with experimental data, numerically $Re < Re_U$. The fact that the Kolmogorov scale η_K and dissipation rate $\mathscr{E}$ are formed on the time scale of a single large-scale turnover time (τ_{eddy}) has been tested in various numerical experiments.

If the Schmidt number $Sc = \nu/D \approx 1$, in accord with the cascade picture, the Kolmogorov scale is the smallest length scale created by turbulence and at times $t > T_K$, the mixing proceeds by molecular diffusion.

If however, $Sc \gg 1$ and the scalar diffusion is extremely inefficient, then, after formation of the Kolmogorov scale η_K, the stretching process by the large-scale ($r \ll \eta_K$) velocity field leads to generation of the ever thinner scalar-enriched sheets until the narrowest sheets on the length scale $\eta_c(t) \approx \eta_B \approx \eta_K/\sqrt{Sc} \ll \eta_K$ are formed. It is only after that, that the scalar diffusion takes over the mixing process. In accord with Batchelor's theory, the mean width of these sheets is $\eta_B \approx \eta_K/\sqrt{Sc}$,

where η_B is called the Batchelor (1959) scale. At this stage, the substances A and B, separated by the distance η_B, can mix only by molecular diffusion on a time scale $\tau_d \approx \eta_B^2/D \approx \eta_K^2/\nu \approx (L^2/\nu)Re^{-3/2}$.

Based on their experimental data, Villermaux *et al.* (2001) proposed a one-step process of simultaneous formation of both Batchelor and Kolmogorov scales which, being physically somewhat different, produced the logarithmically corrected Batchelor estimates of the mixing times. Their conclusions will be examined in what follows where it will be shown that the two mechanisms do not contradict each other.

We consider a simple numerical example. In gases where $Sc \approx 1$, the length scales $\eta_K \approx \eta_B$. In liquids, the situation is different and $Sc \approx 600$–3000 (Dimotakis 1993, 2005) which means that $\eta_K/\eta_B \approx 25$–50. Thus, while the scales η_B and η_K can be very different, the corresponding diffusion and viscous times scales $\tau_d \approx \eta_B^2/D \approx \eta_K^2/\nu \approx \tau_v$ are of the same order. Moreover, it has been shown in various numerical and experimental studies (see for example Monin & Yaglom 1975) that the relevant viscous scale η_v is numerically larger than the Kolmogorov scale: $\eta_v = \alpha_v \eta_K$ with $\alpha_v \approx 10$. Thus, the diffusion time is, in fact, $\tau_d = \eta_v^2/\nu \approx \tau_v$. (The origin of the large factor α_v will be discussed below.)

1.2. *Low- and large-Reynolds-number behaviour of reaction rates*

If $Sc \gg 1$, the process consists of two steps: (i) formation of structures (sheets) on the viscous scale $\eta_v \approx \alpha_v L Re^{-3/4}$; (ii) further stretching of the scalar fieled toward $\eta_c \approx \alpha_v \eta_K/\sqrt{Sc}$. It can be shown readily (see Monin & Yaglom 1975) that, after initial formation of the dissipation scale η_v, the distance between two particles across the sheets, stretched by the large-scale velocity field decreases with time as $r(t) = \eta_v \exp(-\gamma t)$. Thus, the scalar dissipation scale is formed on a time scale τ_c given by the relation:

$$\eta_v e^{-\gamma \tau_c} \approx \eta_v/\sqrt{Sc},$$

where $\gamma = a_\gamma \sqrt{\mathscr{E}/\nu}$ with $1/\sqrt{3} > a_\gamma > 1/2\sqrt{3}$ (Monin & Yaglom 1975). This gives for the total two-step time of formation of the scalar dissipation scale $\tau_{eddy} + \tau_c$ where:

$$\tau_c = \frac{L}{2a_\gamma u_{rms}} Re^{-1/2} \ln Sc \ll \tau_{eddy}.$$

Since τ_c is very small, the contribution of the second (Batchelor's) step to the total mixing time is negligible which can serve as a justification for the one-step process advocated by Villermaux *et al.* (2001). The diffusion time across scalar dissipation sheets is:

$$\tau_d \approx \frac{\alpha_v^2 L^2 Re^{-3/2}}{\nu} \approx \alpha_v^2 \frac{L}{u_{rms}} Re^{-1/2}.$$

Comparing the above relations, we come to a non-trivial conclusion: although both characteristic times τ_d and τ_c scale with the Reynolds number as $\tau_c \propto \tau_d \propto 1/\sqrt{Re}$, owing to the large magnitude of the numerical factor $\alpha_v \approx 5-10$, the process of sheet-thinning by large-scale stretching is numerically much faster than diffusion across the sheets. This means that during the stretching stage, the scalar diffusion across the interfaces can be neglected. The factor $\alpha_v \approx 5-10$ will be calculated below and it will become clear that its large magnitude is a consequence of complex small-scale dynamics of intermittent turbulence.

Thus, the ratio of the mixing (inviscid) and diffusion times can be estimated as:

$$\frac{\tau_{eddy}}{\tau_d} \approx \frac{\sqrt{Re}}{\alpha_v^4}$$

and $\tau_{eddy}/\tau_d \approx 1$ for $Re_{tr} \approx \alpha_\nu^4 \approx 10^3 - 10^4$.

In the flows with $Re < Re_{tr}$, molecular diffusion across the dissipation sheets is the longest rate-determining process. In the wall flows, this Reynolds number corresponds to $Re_U \leqslant 10^5$. It is only when $Re_U \gg 10^5$ that the rapid diffusion through extremely thin interfaces is dynamically irrelevant for the mixing process and inviscid mixing time τ_{eddy} is the rate-determining step. Based on the above considerations, we can expect a transition from the diffusion- to advection-dominated mixing at

$$Re_{tr} \approx \alpha_\nu^4. \tag{1.2}$$

If mixing is the reaction rate ($\mathscr{R}$)-determining process, we expect

$$\mathscr{R} \propto \frac{\nu}{\eta_\nu^2} \approx \frac{\nu}{\alpha_\nu^2 L^2} Re^{3/2} = \frac{\sqrt{Re}}{\alpha_\nu^2 \tau_{eddy}} \qquad (Re < Re_{tr}), \tag{1.3a}$$

$$\mathscr{R} \propto \frac{u_{rms}}{L} \approx \frac{\nu}{L^2} Re^1 = \frac{Re^0}{\tau_{eddy}} \qquad (Re > Re_{tr}). \tag{1.3b}$$

In accord with (1.3), the transitional Reynolds number depends only upon coefficient α_ν, characterizing small-scale properties of turbulence. Thus, based on experimental and numerical data, we can conclude that, since the small-scale property of turbulence $\alpha_\nu \approx 5$–10 is a more or less universal number, independent of type of the flow, the derived $Re_{tr} \approx 10^3 - 10^4$ must be approximately universal. A mixing transition leading to the Reynolds-number-independent reaction rate at approximately universal Reynolds number $Re_{tr} \approx 10^3$, has been observed in experiments on various flows by Dimotakis (1993, 2005). In studies of the low-Reynolds-number scalar mixing by a single vortex, Villermaux & Duplat (2003) demonstrated strong dependence of the mixing time upon vortex circulation which is proportional to Reynolds number. This conclusion qualitatively agrees with the above predictions.

2. Statistical description of disipation structures

Below we present a quantitative theory leading to numerical coefficient $\alpha_\nu \approx 10$. It will be shown that the transition in the mixing rate $\mathscr{R}$, described by relations (1.3), is a consequence of strong anomalous fluctuations of the scalar dissipation rate and corresponding dissipation scale. According to (1.3), the predicted transitional Reynolds number is equal to $Re_{tr} \approx \alpha_\nu^4 \approx 10^3 - 10^4$ ($R_\lambda \approx 100 - 200$). It is clear that no substantial inertial range can be observed in this 'low'-Reynolds-number flow (see Schumacher, Sreenivasan & Yakhot 2007); however, the situation is much more interesting. Schumacher *et al.* (2007) have shown that even at Reynolds numbers $R_\lambda \geqslant 20$, the moments of the the dissipation rate and dissipation scales are characterized by the anomalous exponents identical to those observed in high-Reynolds-number flows ($R \to \infty$). In other words, the small-scale dynamics of the flows with $R_\lambda \approx 100$ is identical to that of a flow at $R_\lambda \to \infty$. Therefore, all conclusions, based on the multifractal theory of intermittent turbulence, are valid for the mixing process considered below.

The Kolmogorov theory (K41), treating the uv cutoff $\eta_K \approx L Re^{-3/4} = \text{const}$, completely disregarded the non-trivial dynamics of the dissipation range fluctuations. It became clear recently that the dissipation scale is not a constant number, but a random field defined as:

$$\eta \approx \frac{\nu}{\delta_\eta u} \equiv \frac{\nu}{|(u(x+\eta) - u(x))|}, \tag{2.1}$$

where $\delta_y u_i = u_i(x+y) - u_i(x)$. In this form, the relation for η was derived (Yakhot 2003, 2006; Yakhot & Sreenivasan 2004, 2005) from the dissipation anomaly, first introduced by Polyakov (1995) for the case of Burgers turbulence and later generalized to the Navier–Stokes turbulence by Duchon & Robert (2000) and Eyink (2003). Even earlier, the fluctuating dissipation scale was used by Paladin & Vulpiani (1987) in the context of multifractal theory. The local value of the Reynolds number $Re \approx u_\eta \eta/\nu \approx 1$, with $\delta_\eta u \equiv u_\eta$ as a typical speed of an eddy of linear dimension η, was mentioned in Landau & Lifshitz (1959) as a criterion for the dominance of viscous dissipation over nonlinearity.

The physical meaning of the dissipation scale η is understood as follows. As $r \to 0$, the velocity field is analytic, so that $\delta_r u \approx \partial_x u(x) r$. Thus, in this limit $S_n = \overline{(\delta_r u)^n} \propto r^n$. On the other hand, in accord with the Kolmogorov 4/5-law, in the inertial ('rough') range, $S_3 \propto r$ which, unless the limiting process is well-defined, cannot be valid in the limit $r \to 0$. We define the dissipation scale η in a following way. In the analytic range, the Taylor expansion is accurate and, owing to continuity of velocity field: $u(\eta+) = u(\eta-) \approx u(0) + \eta \partial_x u(0)$, where $u(\eta\pm)$ is equal to $u(\eta)$ evaluated from the rough and analytic ranges, respectively (for illustration, see Schumacher *et al.* 2007). Subtracting $u(0)$ from both sides of this relation, we obtain

$$\eta = \frac{\delta_\eta u}{\partial_x u(0)}$$

where $\delta_\eta u = u(\eta+) - u(0)$. Thus, we expressed the derivative, proportional to the velocity difference in the limit of infinitesimal displacement, in terms of velocity difference evaluated in the rough anomalous range, but on a particular length scale $r = \eta$.

Equation (2.1) is an order of magnitude estimate of the 'dissipation scale' and, in general, $\eta \approx \gamma \nu/\delta_\eta u$ where γ is a velocity-field-independent factor which was investigated in numerical simulations by Schumacher *et al.* (2007). Interested in the qualitative aspects of the mixing process, we, for now, set $\gamma = 1$ and use (2.1).

According to the analytic theory (Theorem 2, Yakhot 2003), there exist an infinite number of 'dissipation scales' η_n varying in the interval

$$LRe^{-\beta} \leqslant \eta_n \leqslant LRe^{-1/2}$$

($\beta \approx 1$) separating smooth $S_n \propto r^n$ ($r \ll \eta_n$) and singular $S_n \propto r^{\xi_n}$ ($r \gg \eta_n$) ranges of the moments $S_n = \overline{(\delta_r u)^n}$. This fact has been decisively demonstrated in numerical experiments by Schumacher *et al.* (2007). The above relation tells us that $\eta_2 = O(\eta_K)$ (with the *Re*-dependent correction derived below), is only one of many dissipation scales which must be accounted for in the analysis of the scalar mixing. This expression, which sets an upper bound $\eta \leqslant O(L/\sqrt{Re})$ on possible magnitudes of dissipation scales, showing that the inertial range is even smaller than previously thought, is extremely important (see § 4.)

2.1. *Scalar dissipation scales*

Let us consider the equation for concentration c of a passive scalar advected by velocity $\boldsymbol{u}$:

$$\frac{\partial c}{\partial t} + \boldsymbol{u} \cdot \nabla c = D \nabla^2 c, \qquad \nabla \cdot \boldsymbol{u} = 0 \tag{2.2}$$

We assume that the velocity field $\boldsymbol{u}$ is governed by the Navier–Stokes equations and $\boldsymbol{u} = \boldsymbol{U} + \boldsymbol{u}'$ with $\boldsymbol{U}$ and $\boldsymbol{u}'$ corresponding to quasi-regular (sometimes time-dependent) and chaotic (turbulent) contributions, respectively. The fluctuations of the scalar

dissipation field $N = D(\nabla c)^2$, governed by (2.2), have been investigated in detail for the case of the δ-correlated in time, large-scale velocity field (Kraichnan–Batchelor problem), where the stretched exponential tail of the distribution function $P(N)$ was derived in the range $N \gg \overline{N}$ (Chertkov, Falkovich & Kolokolov 1998; Gamba & Kolokolov 1999). In this paper, we are interested in statistical properties of 'molecular diffusion sheets', which are not directly related to the tails of the scalar dissipation rate distribution.

The scalar field is analytic, so that for $r \to 0$, $c(x+r) - c(x) \approx (\partial c(x)/\partial x)r$. In addition, in the scalar 'inertial range' $r \gg \eta_K$, we can derive the Yaglom relation (see Monin & Yaglom 1975):

$$S_3^{u,c} = \overline{(u(x+r) - u(x))(c(x+r) - c(x))^2} = -\tfrac{4}{3}\overline{N}r,$$

where $N = D\overline{(\partial c/\partial x_i)^2} = O(1)$. In the case $Sc \gg 1$, there exists an additional scalar 'rough' range $\eta_B \leqslant r \leqslant \eta_K$, where $S_{2,c}(r) = \overline{(c(x+r) - c(x))^2} \propto \ln r$. It is only at the scales $r \ll \eta_B$ that the scalar field is smooth. By our definition, the length scale $r \approx \eta_c$ is the scale separating analytic and singular contributions to the scalar field $c(\boldsymbol{r}, t)$. As follows from the Yaglom relation, in the inertial range $r \gg \eta_c \to 0$, the scalar field $c(\boldsymbol{x})$ is not differentiable and in the limit $D \to 0$, we must be careful with evaluation of spatial derivatives of the scalar field.

From (2.2) we have

$$\frac{\partial c^2}{\partial t} + \boldsymbol{u} \cdot \nabla c^2 = 2Dc\nabla^2 c \tag{2.3}$$

and introducing the 'point-splitting' $c(\pm) = c(x \pm y)$ and $\boldsymbol{u}(\pm) = \boldsymbol{u}(x \pm y)$, derive:

$$\frac{\partial (c(+)c(-))}{\partial t} + (\nabla_+ \boldsymbol{u}(+) + \nabla_- \boldsymbol{u}(-))c(+)c(-) = D(\nabla_+^2 + \nabla_-^2)c(+)c(-). \tag{2.4}$$

Equation (2.4) involves derivatives of singular (in the inertial range) functions. Thus, in the limit $y \to \eta_c \to 0$, the exact equation (2.3) for the scalar variance can appear from (2.4) only if singular and regular contributions balance separately. Indeed, in the limit $y \to \eta_c \to 0$, taking into account that $\partial/\partial y = \partial/\partial x_+ = -\partial/\partial x_-$ and repeating all steps presented in detail in Yakhot (2006), we derive:

$$\frac{\partial}{\partial y_i}(\delta_y u_i (\delta_y c)^2) + 2\nabla_+ \boldsymbol{u}(+)c^2(-) + 2\nabla_- \boldsymbol{u}(-)c^2(+) = -4D\delta_y c \frac{\partial^2}{\partial y^2}\delta_y c. \tag{2.5}$$

Equation (2.5), though resembling the Yaglom relation, is exact locally in space and time. Since in incompressible isotropic turbulence the velocity–scalar correlation function $\overline{u_i(x)c^2(x')} = 0$ (Landau & Lifshitz 1959; Monin & Yaglom 1975), it is clear that averaging over a 'ball' of radius η_c (Duchon & Robert 2000; Eyink 2003), the second and third terms in the left-hand side of (2.5) disappear giving : $(\partial/\partial y_i)(\delta_y \overline{u_i (\delta_y c)^2})|_{\eta_c} = -4D\delta_y c(\partial^2/\partial y^2)\delta_y c|_{\eta_c}$ where $\overline{A}|_{\eta_c}$ stands for the averaging over a 'ball' of radius η_c with the centre at an arbitrary point $\boldsymbol{r}$. Since $y \to \eta_c$, this expression gives the locally valid estimate for the scalar dissipation scale, independent of the specific model of turbulence:

$$\eta_c \approx \frac{D}{\delta_{\eta_c} u}. \tag{2.6}$$

We see that the random variable η_c depends upon local time-dependent values of velocity fluctuations and, being a dynamic relation, cannot be obtained on dimensional grounds from the globally averaged expressions for various correlation functions. In

the most interesting and important case $Sc = \nu/D \gg 1$, on the scale $\eta_c \ll \eta$ the velocity field is analytic, giving:

$$\eta_c^2 \approx \frac{D}{\partial u(x)/\partial x} \approx \frac{D\eta}{\delta_\eta u} \approx \frac{D\eta^2}{\nu} = \eta^2/Sc. \tag{2.7}$$

Thus, the probability of the scalar dissipation scale $Q(\eta_c)$ is evaluated readily from the PDF $Q(\eta)$ calculated in Yakhot (2006). This result leads to some important consequences. If

$$\tau_d \approx \eta_v^2/\nu = \alpha_v^2 \eta_K^2/\nu \approx \alpha_v^2 \tau_{eddy}/\sqrt{Re}, \tag{2.8}$$

to evaluate the reaction rate and the proportionality coefficient α_v, we need the probability density function $Q(\eta_c) = \sqrt{Sc}\,Q(\eta\sqrt{Sc})$.

The fluctuations of the dissipation scale, defined as the width of the scalar dissipation isosurfaces $N = D(\partial c/\partial x_i)^2 \approx \text{const}$, have been numerically investigated in Kushnir, Schumacher & Brandt (2006) and Schumacher, Sreenivasan & Yeung (2005). Strong fluctuations have been detected and characterized by their probability density. It will become clear below that the dynamic definition (2.6), (2.7), though qualitatively similar to the one used in the above-mentioned works, is quantitatively very different. In experiments on both cold and reacting (burning) jets, J. Frank (personal communication, 2007) observed strong fluctuations of quasi-laminar scalar patches which can be associated with the entrained scalar dissipation scales. His experiments also showed strong fluctuations characterized by broad probability densities. Similar features were observed by Wang & Peters (2006) in the analysis of the 'dissipation elements' in reacting flows. All these examples show that the theory of PDFs of the scalar dissipation scale $Q(\eta_c)$ is essential for description of mixing, reactions and heat-mass transfer.

3. Probability density of the scalar dissipation scale

In what follows, we set $L = 1$, so that $r/L \equiv r < 1$ and if the moments of velocity increments $S_{n,0} = \overline{(\delta_r u)^n} = A(n) r^{\xi_n}$, then the probability density function can be found from the Mellin transform:

$$P(\delta_r u, r) = \frac{1}{\delta_r u} \int_{-i\infty}^{i\infty} A(n) r^{\xi(n)} (\delta_r u)^{-n} \mathrm{d}n, \tag{3.1}$$

where we set the integral scale L and the dissipation rate $\mathscr{E}$ equal to unity. Multiplying (3.1) by $(\delta_r u)^k$ and evaluating a simple integral, gives $S_{k,0} = A(k) r^{\xi_k}$. Under a different name, this transformation has been used in Tcheou *et al.* (1999) in the context of the multifractal theory of turbulence. With the Gaussian large-scale boundary condition for the probability density at $r = 1$, the amplitudes $A(n) = (2n-1)!!$ and, for the values of $n < 1/b$, we can use Taylor expansion of the function ξ_n giving $\xi_n \approx (an - bn^2)$. Substituting this into (3.1) and evaluating the integral using the steepest descent method, gives (Yakhot 2006) the relation for the probability density ($\delta_\eta u \equiv u_\eta$):

$$P(u_\eta, \eta) = \frac{2}{\pi u_\eta \sqrt{4b|\ln \eta|}} \int_{-\infty}^{\infty} \exp(-x^2) \exp\left[-\frac{(\ln u_\eta/\eta^a \sqrt{2}x)^2}{4b|\ln \eta|} \right] \mathrm{d}x, \tag{3.2}$$

where $\eta/L = \nu/(L\delta_\eta u) = (u_{rms}/\delta_\eta u)(1/Re)$. The parameters $a \approx 0.38$ and $b = 0.017$, leading to $\xi_3 = 1$, give $\xi(n)$ in close agreement with experimental data in the range $-1 < n < 10$–15 (see for example Yakhot 2003, 2006).

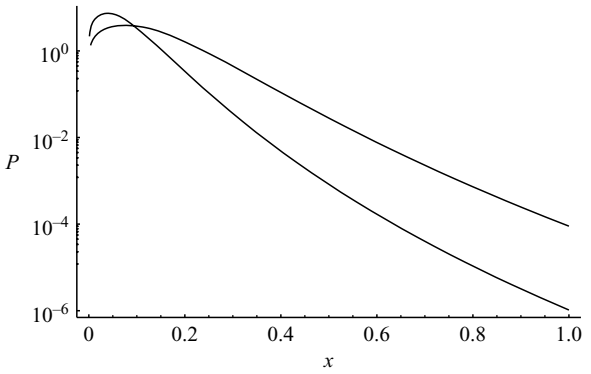

FIGURE 1. Probability density $P(u_\eta/u_{rms}) = P(x)$. $Re_\eta = 1000$. The curve shifted to the left corresponds to $Re_\eta = 10\,000$.

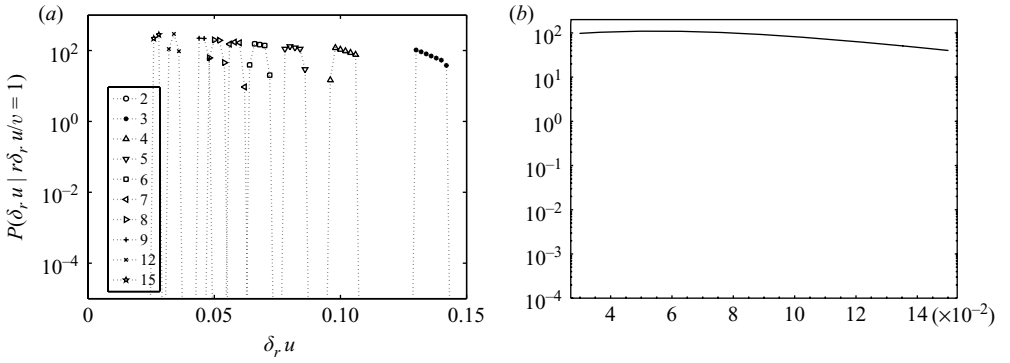

FIGURE 2. (*a*) Numerical simulations (Schumacher 2007). Conditional probability density $P(u_\eta/u_{rms})$. $Re = 3300$; ($R_\lambda = 65$). The displacement $r = n\Delta$, where Δ is the computational mesh size and n are given in the left-hand column. The Kolmogorov dissipation scale $\eta_K = 3\Delta$. (*b*) Theoretically predicted (equation 3.2) probability density $P(x) \equiv P(u_\eta/u_{rms})$. The PDF is supported only in a relatively narrow range of variation of $\delta_\eta u$ and η.

We would like to stress that the probability density function $P(u_\eta, Re)$ is a conditional PDF $P(u_r, Re|Re_r = 1)$, with $Re_r = ru_r/\nu$, and not the PDF of velocity differences at an arbitrary value of the displacement r. The difference between the two is huge: while the PDF $P(u_r) \neq 0$ at $u_r = \delta_r u = u(x+r) - u(x) = 0$, the conditional PDF $P(u_\eta|Re_\eta = 1)$ is not supported at $u_\eta = 0$ (see figures 1 and 2), and thus, both the moments of u_η and those of $\eta = \nu/u_\eta$ are finite.

Numerical simulations by Schumacher (2007)

To investigate this important point, J. Schumacher (personal communication, 2007) conducted detailed high-resolution ($N = 1024^3$; 2048^3 grid points) numerical simulations of the large-scale-driven isotropic and homogeneous turbulence. The set-up is described in detail in Schumacher *et al.* (2007). Setting the displacement $r = n\Delta$ where Δ is the size of computational mesh, he counted only the events with $r\delta_r u/\nu = 1$. The chosen mesh size corresponded to $\eta_K = 3\Delta$. It is clear that the imposed constraint led to a substantially reduced data set with the resulting larger than usual statistical uncertainty. Moreover, the discreteness of the grid (the numbers n are given in the left-hand column of figure 2*a*) contributed to the discontinuity of the curve. Still,

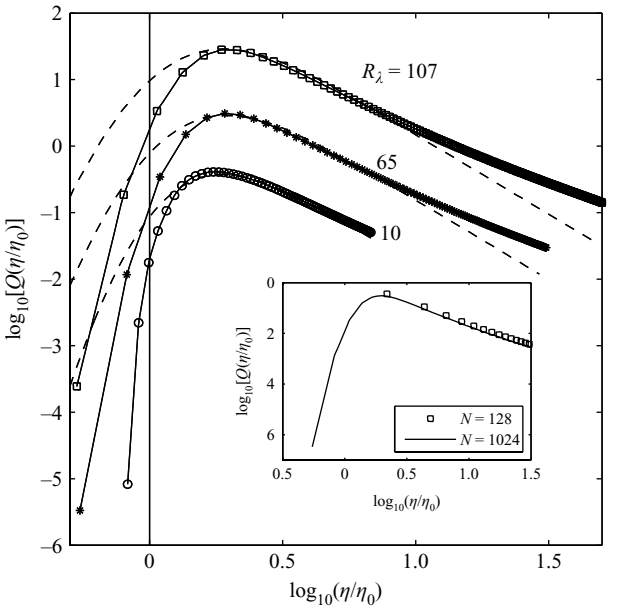

FIGURE 3. Probability density of velocity dissipation scale $Q(\eta/\eta_0, Re)$ for $10 < R_\lambda < 107$. Numerical simulations ($N = 128^3$ and $N = 1024^3$) by Schumacher (2007). Dotted lines, theoretical prediction (3.3). Inset: dependence of the results upon computational resolution. The curves corresponding to different Reynolds numbers are shifted for clarity of presentatyion.

the results for $R_\lambda = 65$, presented in figure 2, are most illustrative. We see that in accord with theoretical prediction, the PDF $P(u_\eta)$ is supported in a very narrow interval of the velocity variation $0.03 < u_\eta/u_{rms} < 0.15$ and $\eta_K \leqslant r \leqslant 5\eta_K$. Thus, both the moments of u_η and those of $\eta \propto 1/u_\eta$ are finite. This is most important for what follows. We also see that the very large (and very small) variations of velocity difference across the dissipation sheets are of very low probability (if possible at all), which justifies the Taylor expansion $\xi_n \approx an - bn^2$ used in the derivation of (3.2). The results of numerical simulations compare well with the theoretical predictions derived from (3.2) (figure 2*b*). To make a more quantitative comparison with theoretical predictions given by (3.2), we need a much larger data set.

Substituting $u_\eta/u_{rms} = (1/Re)(L/\eta)$ into (3.2), gives the desired probability density of dissipation scales. Fixing $L = \mathscr{E} = u_{rms} = 1$, $\nu = 1/Re$ and taking into account that, by virtue of (2.1) , $u_\eta Re/u_{rms} = L\delta_\eta u/\nu \approx L/\eta$ gives for the probability density $Q(\eta)$ (in what follows we denote $\eta/L \equiv \eta$):

$$Q(\eta, Re) = \frac{1}{\eta\sqrt{4b\ln\eta}} \int_{-\infty}^{\infty} \exp(-x^2)\,\mathrm{d}x \exp\left(-\frac{\ln^2(\eta^{a+1}\sqrt{2}x\,Re)}{4b\ln\eta}\right)$$
$$= \frac{1}{\eta\sqrt{4b|\ln\eta|}} \int_{-\infty}^{\infty} \exp(-x^2)\,\mathrm{d}x \exp\left(-\frac{\ln^2((\eta/\eta_0)^{a+1}\sqrt{2}x)}{4b|\ln\eta|}\right) \qquad (3.3)$$

The theoretically derived PDFs of dissipation scales (equation (3.3)) are compared with the outcome of the most detailed numerical simulations by Schumacher (2007) in figure 3 showing a very close agreement between theoretical predictions and simulations in the range $\eta \leqslant \eta \leqslant 10-20\eta_0$, where $\eta_0 \approx LR^{-1/(1+a)} \approx LRe^{-0.72} \approx \eta_K Re^{0.03}$ (see Schumacher *et al.* 2007). Since $\eta \propto 1/\delta_\eta u$, the disagreement at the smaller scales

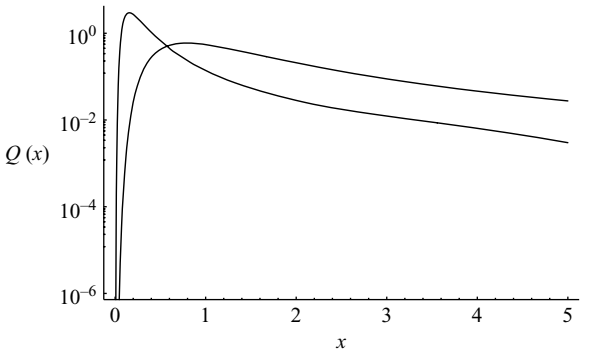

FIGURE 4. $Q(\eta_c/\eta_0, Re = 10^5) = Q(x)$: PDFs of dissipation scale from (3.3) for $Sc = 1$. The shifted to the left curve corresponds to $Sc = 25$.

$\eta < \eta_K$ is related to the upper bound on the velocity difference $\delta_\eta u \ll u_{rms}$, not accounted for in the theory. In the range $\eta > 10$–$20\eta_K$, the theoretical curve is somewhat higher than the 'numerical curve', which is definitely related to the cutoff in the dissipation scale distribution $\eta < \sqrt{L}$ (see § 2). Both ranges of very small and very large dissipation scales scale and are irrelevant for the mixing processes that we are interested in this paper.

In the most interesting case $Sc \gg 1$, we, using the relation (2.7), obtain the PDF of the scalar dissipation scales by a simple substitution $\eta \to \eta_c\sqrt{Sc}$. The result is presented in figure 4, where $\eta_0 = LRe^{-(1/1+a)} \approx \eta_K Re^{0.03}$. As expected, the probability density of dissipation scales is expressed in terms of the ratio $\eta' = \eta/\eta_0$ and the width of the distribution is the weak function of the Reynolds number.

4. Mean dissipation scale and diffusion time. Evaluation of α_v

Now we can evaluate the moments of the dissipation scale:

$$\overline{\left(\frac{\eta}{\eta_0}\right)^n} = \int_0^\infty \left(\frac{\eta}{\eta_0}\right)^n Q(\eta, \eta_0)\,\mathrm{d}\eta \tag{4.1}$$

so that $\overline{\eta} \equiv \alpha_v\eta_0 = \alpha_v\eta_K Re^{0.03}$ corresponds to $n = 1$. The mean diffusion time can also be calculated readily:

$$\overline{\tau_d} = \overline{\eta^2}/\nu = \int_0^\infty \eta^2 Q(\eta, \eta_0, Re)\,\mathrm{d}\eta/\nu. \tag{4.2}$$

It is easy to see that if the above integrals are evaluated over the interval $0 \leqslant \eta < \infty$, the high-order moments with $n \geqslant 2$ diverge, i.e. do not exist. However, in accord with the theorem (Yakhot 2003), $\eta \leqslant O(\sqrt{Re})$, which sets up the finite upper limit of integration rendering all moments of the dissipation scale finite. It is worth mentioning that the moment with $n = 1$, leading to $\overline{\eta} \equiv \alpha_v\eta_K$, does exist and can readily be evaluated. The numerical results vary slightly with the position of the maximum of PDF $Q(\eta/\eta_0)$, which depends upon the magnitude of parameter γ in the expression $\gamma\eta u_\eta/\nu = 1$. If we choose $\gamma \approx 1$, so that, in accord with the numerical simulation of Schumacher (2007), the maximum of the curve is set at $\eta_{max}/\eta_0 \approx 2$, then numerical integration (4.1) yields $\overline{\eta} = \alpha_v\eta_0 \approx 7\eta_0$ and $\overline{\eta^2} \approx 120\eta_0$. A similar result was obtained by Schumacher (2007) who reported a slight variation of $\overline{\eta}/\eta_0$ with Reynolds number in the interval $6 < \overline{\eta}/\eta_0 < 10$. This result also agrees with the conclusion $\overline{\eta}/\eta_K \approx 6 - 7$ obtained from

numerical simulations by A. Pumir & G. Falkovich (personal communication, 2007) as an input parameter to their theory of rain. All these results seem to agree with each other. We must bear in mind that the theoretical estimate derived in this paper involves dimensionless length scale η/η_0 where $\eta_0 = \eta_K Re^{0.03}$ and not η/η_K. This may introduce some slight modifications to the above estimates.

4.1. *Mixing reactants having finite lifetime*

To characterize chemical reactions, we can define the dimensionless Damköhler number $Da = (\tau_c + \tau_d)/\tau_{reaction}$, which is the ratio of the time scale of hydrodynamic mixing to $\tau_{reaction}$, where $\tau_{reaction}$ is the reaction time between perfectly mixed reactants A and B. In what follows, we consider a simple example of a model photochemical reaction $A^* + B = AB + h\nu$ where A^* is a component A in an initially prepared electronically excited state characterized by the lifetime τ_e. By definition, the finite lifetime τ_e implies time-dependence of concentration of excited states $c_{A^*} = c(t=0)\exp(-t/\tau_e)$ meaning that τ_e seconds after the initial laser pulse (excitation), the concentration of an excited reagent decreases by factor e.

First, we are interested in a diffusion-dominated limit $Re \leqslant 10^3 - 10^4$. A chemical reaction is possible only if diffusion time $\tau_d \approx \alpha_\nu^2 \eta_B^2/D \leqslant \tau_e$, $\alpha_\nu \eta_B \leqslant \sqrt{\tau_e D}$ and $\eta_\nu \leqslant \sqrt{\tau_e \nu}$. This gives, in addition to the Damköhler number, a dimensionless reaction criterion:

$$Y = \alpha_\nu \sqrt{\frac{\tau_{eddy}}{\tau_e}} Re^{-1/4} = \alpha_\nu \sqrt{Da} \sqrt{\frac{\tau_{reaction}}{\tau_e}} Re^{-1/4} \leqslant 1. \tag{4.3}$$

This relation means that the reaction is possible only if

$$\frac{\tau_{eddy}}{\tau_e} \leqslant \frac{\sqrt{Re}}{\alpha_{nu}^2}. \tag{4.4}$$

We see that if the lifetime is short, the reaction is possible only if

$$Re \geqslant \alpha_\nu^4 \left(\frac{\tau_{eddy}}{\tau_e}\right)^2 \approx (10^3 - 10^4)\left(\frac{\tau_{eddy}}{\tau_e}\right)^2, \tag{4.5}$$

which is often impossible in the relatively low-Re flow. Thus, we conclude that it is only when $Re > Re_{tr}$ that we can expect $L/u_{rms} \leqslant \tau_e$ with a non-zero reaction rate.

5. Conclusions

1. The scalar and velocity dissipation scales η and η_c in turbulent flows are not constant numbers, but describe random fields with $\eta_c \approx D/(\partial u/\partial x)$ and $\eta \approx \nu/\delta_\eta u$, respectively. In an important case $Sc \gg 1$, these scales are related as $\eta_c \approx \eta/\sqrt{Sc}$. This has been observed in numerical and physical experiments by Schumacher & Sreenivasan (2003), Kushnir *et al.* (2006), Wang & Peters (2006), J. Frank (personal communication, 2007), A. Pumir & G. Falkovich (personal communication, 2007), Schumacher *et al.* (2007).

2. Based on the Mellin transform and Taylor expansion of the scaling exponents of velocity structure functions, the probability density of the scalar dissipation scale has been derived. Two main results of this paper are: (i) Owing to strong small-scale intermittency, the calculated mean thickness of a dissipation sheet is $\alpha_\nu \eta_0 \approx \alpha_\nu \eta_K Re^{0.03}$ where $\alpha_\nu \approx 5$–10; (*b*) extremely strong intermittency leads to a long mean scalar-transport time $\overline{\tau_d}$ across the sheets and, in the flows with $Re \leqslant \alpha_\nu^4 \approx 10^3 - 10^4$, to molecular diffusion as a reaction-rate-determining step .

3. Therefore, the chemical reaction rate is: $\mathscr{R} \propto \sqrt{Re}$ for $Re < Re_{tr}$, and $\mathscr{R} \propto Re^0$ in the interval $Re > Re_{tr}$. The transition in the reaction rates was derived without any assumptions about sharp flow modifications at $Re = Re_{tr}$, assumed by Dimotakis (2005) for interpretation of experimental data on mixing in jets, wakes, mixing layers, etc. To demonstrate the $\mathscr{R} \propto \sqrt{Re}$ range of the Reynolds-number variation of the reaction rate, we must design a special set of experiments. All that can be said today is: the theory presented in this paper led to a transition from the Re-dependent to the Re-independent reaction rate happening at $Re_{tr} \approx 10^3 - 10^4$. Qualitatively, the predicted transition is similar to that observed experimentally by Dimotakis and his group. Our results seem to agree with Villermaux & Meunier (2003) experimental data on a mixing by a single vortex demonstrating that in this low-Re flow, the mixing time depended upon the vortex circulation which is proportional to the Reynolds number. The mixing time, independent of Reynolds number, has been demonstrated in Villermaux & Duplat (2006) on an example of a cold jet flow in the interval $10^3 \leqslant Re \leqslant 10^7$. These experimental results seem to agree with the theoretical conclusions of this paper. It remains to be seen whether the conclusions of Villermaux & Meunier (2003) hold in the more complex turbulent flows that we are interested in that.

4. The total time of the two-step formation of the Batchelor scale $T \approx \tau_{eddy} + \tau_c \approx \tau_{eddy}$, with $\tau_c \ll \tau_{eddy}$, justifies the Villermaux *et al.* (2001) suggestion of a short-cut in the Corrsin–Obukhov cascade or one-step Batchelor–scale formation.

5. We may think that the diffusion-dominated range $Re < Re_{tr} \approx 10^3 - 10^4$ is not relevant for real-life scientific and engineering applications. This may be so for cold flows; however, taking into account that kinematic viscosity of air varies with temperature as $\nu \propto T^{1.7}$, we come to the conclusion that at $T = 1500\,^{\circ}\mathrm{K}$, typical of combustion, the viscosity is twenty times that of air at room temperature. Therefore, often, the Reynolds numbers of reacting flows are relatively small with the mixing governed by the relations derived above. Similar effects may explain the poor performance of turbulence models in the description of heat transfer in very hot gases. This will be the subject of a future paper.

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