PDF MODELLING OF TURBULENT REACTING FLOWS: REYNOLDS AND DAMKOEHLER NUMBER EFFECTS

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ABSTRACT

Turbulent reacting flows with moderate or high Reynolds (Re), Schmidt (Sc) and Damkoehler (Da) numbers can be calculated only approximately. Lagrangian probability density function (PDF) methods are especially attractive for computing such flows, because approximations are restricted to the simulation of mixing processes. For the calculation of these processes, a new Lagrangian multi-scale scalar mixing model is presented here. In contrast to previously applied methods, this model describes for high-Sc flows mixing also at the Kolmogorov- and Batchelor-scales. This is relevant, because the characteristic scalar mixing time may be much larger than for flows without extended Kolmogorov- and Batchelor-scale. The significant effect of this delay of the onset of chemical reactions is illustrated by means of the simulation of mixing and parallel chemical reactions of species in a turbulent pipe flow. It is shown, that the neglect of such effects by conventional techniques results in errors of about 50% for the case considered. This application reveals the remarkable effects of Re and Da, which are closely related to each other.

INTRODUCTION

Turbulent reacting flows may be distinguished by means of their characteristic numbers, Re, Sc and Da: they determine the competition between the most important processes, mixing and reaction. In flows of industrial or environmental relevance, Re, Sc and Da may vary over orders of magnitudes. Hence, approximation methods are needed for many flow calculations, because direct numerical simulation (DNS) can only be applied to flows with small Re, Sc and Da.

The approximated calculation of turbulent reacting flows by Eulerian techniques as Reynolds-averaged Navier Stokes (RANS) equations or large eddy simulation (LES) requires closure models for the mean or filtered chemical conversion rate, which have to reflect the influence of mixing on this conversion rate. Usually applied closures are justified only for very high or small Da. The attraction of Lagrangian PDF methods, which calculate the joint PDF of velocity and composition using particle methods, results from two facts: First, in contrast to DNS, these methods can be applied to flows with Re, Sc and Da that vary from very small to very high values. Second, in contrast to conventional Eulerian methods, PDF methods do not have the problem to close the reaction rate, i.e., the competition between mixing and reaction is simulated only as a consequence of the mixing simulation (Pope, 1985, Fox, 1996).

To improve existing mixing models and to illustrate simultaneously the relevance of considering Re-, Sc- and Da-effects, we present a new Lagrangian multi-scale scalar mixing model. In contrast to methods used previously (Fox, 1995, 1997), this model is shown to be applicable to high-Sc flows (liquids). Due to the fact that the characteristic scalar mixing time may be much larger, the mixing in these flows differs remarkably from that in low-Sc flows (gases).

First, usual Lagrangian models for the velocity and mass fractions of species are presented as a frame for the following development. Then, the derivation of the new mixing model and its application to mixing and reaction of species in a turbulent pipe flow are described.

LAGRANGIAN STOCHASTIC MODELS

By Lagrangian stochastic models, the turbulent flow can be described according to a variety of turbulence models (Pope, 1994a, Durbin and Speziale, 1994, Wouters et al., 1996, Heinz, 1997). To keep the development of the methodology as simple as possible, we describe the flow by means of the simplified Langevin model (Pope, 1985). In that way, the change in time t of positions $\mathbf{x}^* = (x_1^*, x_2^*, x_3^*)$ and velocities $\mathbf{U}^* = (U_1^*, U_2^*, U_3^*)$ of a fluid particle moving with the flow is given by (i = 1, 2, 3)

$$\frac{d}{dt}x_i^*(t) = U_i^*, \tag{1a}$$

$$\frac{d}{dt}U_i^*(t) = -\rho^{-1}\frac{\partial p}{\partial x_i} - \frac{1}{\tau}\left(\frac{3}{4}C_0 + \frac{1}{2}\right)\left(U_i^* - \langle U_i\rangle\right) + \sqrt{C_0\frac{q^2}{2\tau}}\frac{dW_i}{dt}. \tag{1b}$$

Here, p is the Reynolds-averaged pressure, ρ the averaged fluid density, $q^2 = \langle u_i u_i\rangle$ twice the turbulent kinetic energy (TKE) and $\tau = q^2 / (2\varepsilon)$ the dissipation time scale of turbulence, where ε is the mean dissipation rate of TKE. Eulerian velocity fluctuations are denoted by $u_i = U_i - \langle U_i\rangle$, and summation is assumed for repeated subscripts. The Reynolds-averaged Eulerian velocity $\langle\mathbf{U}\rangle(\mathbf{x}^*, t)$ is written without star in contrast to Lagrangian quantities. The last term in (1b) describes the influence of random accelerations. This term is characterized by the white noise dW_i/dt, which is a Gaussian process with vanishing mean values, $\langle dW_i / dt\rangle = 0$, and with uncorrelated values at different times, $\langle dW_i / dt(t) \cdot dW_j / dt'(t')\rangle = \delta_{ij}\,\delta(t - t')$. The symbol δ_{ij} is the Kronecker delta and $\delta(t - t')$ the delta function. For the parameter C_0 we apply $C_0 = 3.5$ (Dreeben and Pope, 1997, Heinz, 1998).

Analogously, scalar transport is modelled by means of the often applied 'interaction by exchange with the mean' (IEM) model (Pope, 1985). The mixing of the mass fraction Φ_α^* of a scalar α is described as

$$\frac{d}{dt}\Phi_\alpha^*(t) = -G_\alpha\left(\Phi_\alpha^* - \langle\Phi_\alpha\rangle\right) + \rho^{-1}r_\alpha, \tag{1c}$$

where G_α is assumed to be an unknown coefficient. The first term on the right-hand side of (1c) models the scalar micromixing in a formal correspondence to that of momentum in (1b). $\langle\Phi_\alpha\rangle$ denotes the mean Eulerian mass fraction of the α^{th} scalar. The reaction rate $\rho^{-1}\,r_\alpha(\Phi)$ describes chemical transformations exactly.

Equations (1a-c) determine the one-point joint velocity-composition PDF, from which transport equations can be derived for all their moments. These transport equations for the mean velocities and mass fractions correspond to the exact RANS equation for these quantities if the acceleration due to gravity and molecular effects are neglected. The transport equations for the variances of the velocity fields correspond with Rotta's (1951) turbulence model, if Kolmogorov's (1942) theory for the dissipation is adopted. A corresponding comparison between the composition covariance transport equation derived from (1c) with equations applied in the Eulerian framework is used in the next section for the calculation of G_α.

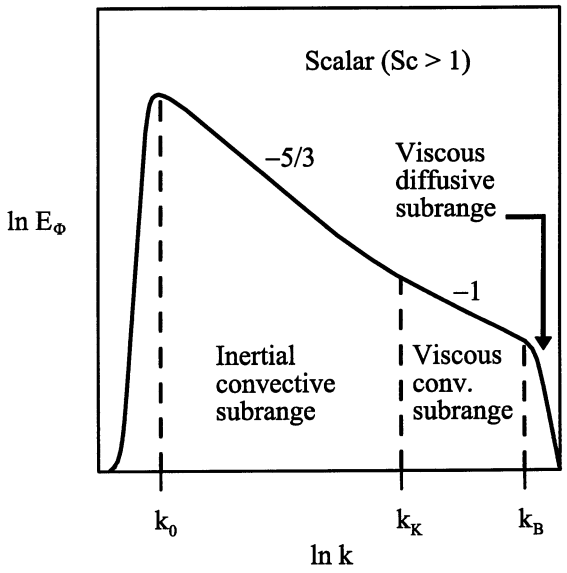

Figure 1. Idealized scalar energy spectrum in fully developed homogeneous, isotropic turbulence. There exists a large inertial convective subrange between the integral-scale and the Kolmogorov-scale wavenumbers, k_0 and $k_K = Re_t^{3/2}\,k_0$. Re_t is the turbulent Reynolds number. For Schmidt numbers Sc > 1, the scalar energy spectrum has additional viscous convective (extending to the Batchelor-scale wavenumber $k_B = Sc^{1/2}\,k_K$) and diffusive subranges. The slopes − 5 / 3 and − 1 denote the scaling laws $E_\Phi(k) \sim k^{-5/3}$ and $E_\Phi(k) \sim k^{-1}$ in the corresponding subranges, where E_Φ is the spectral scalar density functions.

Provided G_α, boundary and initial conditions are given, the equations (1a-c) provide a complete description of the turbulent flow and scalar mixing and reaction, because the Eulerian mean velocities, compositions, the mean pressure gradient and the TKE can be evaluated from particle properties (Pope, 1994b).

COMPOSITION FREQUENCY MODEL

The coefficient G_α in (1c) is often modelled by $G_\alpha = C_\varphi / (2\tau)$, i.e., it is considered as determined by the characteristic turbulence frequency τ^{-1} of the inertial subrange. C_φ is a constant with a standard value $C_\varphi = 2.0$. However, in particular for high-Sc reacting flows, which are of special interest for many applications in the chemical process industry, the consideration of mixing processes at the

Kolmogorov- and Batchelor-scales may be very important, see Figure 1. The Kolmogorov-scale wavenumber k_K and the Batchelor-scale wavenumber $k_B = Sc^{1/2} k_K$ are well-separated in that case, i.e., the characteristic transport time from k_0 to k_B is significantly larger than for small-Sc flows so that there is a remarkable delay of the onset of chemical conversion processes. In dependence on the reaction scheme, this fact may lead to phenomena that cannot be predicted by simple approximations.

By adopting guidance of an Eulerian multi-scale mixing model, the consideration of multi-scale mixing in Lagrangian simulations of the evolution of scalar fields recently has been considered by Fox (1995, 1997). From a methodological point of view, this approach represents a substantial progress due to its systematic nature of describing the transport of scalar energy as a cascade process from large to small scales. This Lagrangian spectral relaxation (LSR) model describes the multi-scale transport of scalars in gases in a very good agreement with DNS data, but there are some questions with reference to its direct applicability to liquid-phase reacting flows. Presently, comparisons of predictions of the LSR model with experimental data are not available for flows with Sc > 1.

Therefore, we use another Eulerian multi-scale mixing model (Baldyga, 1989) as a guideline to extend the Lagrangian description of mixing processes at the Kolmogorov- and Batchelor-scale to flows with Sc >> 1. Baldyga's model is applicable to the description of the mixing of different scalars in inhomogeneous and instationary flows, relatively simple and proved for a variety of problems (Baldyga, 1994, Kruis and Falk, 1996, Baldyga and Henczka, 1995, 1997). We apply this model by constructing the composition frequency G_α in (1c) so that the composition covariance transport equations that follow from (1c) are fully consistent with the corresponding equations of Baldyga. The model derived in that way is denoted below as our general model.

The G_α obtained is given through a complicated system of partial differential equations. For applications, an algebraic expression for G_α is of special interest in order to reduce the computational effort. This approximation corresponds to the consideration of the mixing frequency G_α as a property of the flow field, which is asymptotically little influenced by details of scalar initial distributions or boundary conditions. By neglecting gradient terms, one obtains an algebraic equation of third-order in G_α. Only one of these three solution of the cubic equation will be realized, which reads

$$G_\alpha = \frac{1}{\sqrt{3}} \sqrt{\frac{r^2}{3} - s} \cdot \cos\left(\frac{\varphi + 2\pi}{3}\right) - \frac{r}{3}. \qquad (2)$$

Here, the abbreviations r, s and φ are given by by $r = -(E + G + C_\varphi / \tau + 2\gamma_\alpha)$, $s = E C_\varphi / \tau + (G + \gamma_\alpha)(E + C_\varphi / \tau + \gamma_\alpha)$ and $\varphi = \mathrm{Arccos}([E G C_\varphi / \tau - 2 r^3 / 27 + r s / 3] / (2 [(r^2 / 3 - s)^3 / 27]^{1/2}))$. The influence of Kolmogorov- and Batchelor-scale mixing is reflected through the frequencies $E = 0.058 / \tau_\eta$ and $G = (0.303 + 17050 / Sc) E$. Here, τ_η is the Kolmogorov time scale that is related to the kinematic viscosity ν by $\tau_\eta = (\nu / \varepsilon)^{1/2}$. Additionally, $\gamma_\alpha = -2 \langle u_j c_\alpha \rangle / \sigma_\alpha^2 \cdot \partial \langle C_\alpha \rangle / \partial x_j$ is introduced ($\langle u_j c_\alpha \rangle$ is the turbulent scalar flux), which represents the influence of scalar gradients on the composition frequency. With reference to its IEM-structure, the model (2) is denoted as algebraic multi-scale interaction by exchange with the mean (AMSIEM) model.

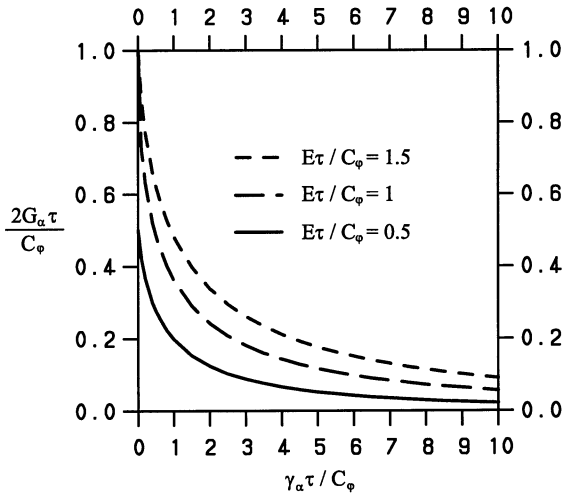

Figure 2. The composition mixing frequency G_α according to (2) in dependence on E, G and γ_α, where all quantities are normalized to C_φ / τ. G is related to E by $G = (0.303 + 17050 / Sc) E$, where Sc = 800.

Analysis of (2) reveals the relation $G_\alpha \leq 0.5$ Min $(G, E, C_\varphi / \tau)$. The equal sign applies to $\gamma_\alpha = 0$, i.e., in that case G_α is given by the minimum of the available frequencies G, E and (C_φ / τ). Hence, the consideration of E and G may lead to an increase of the characteristic mixing time G_α^{-1} due to the consideration of scalar fluctuations beyond the inertial convective subrange in the viscous convective and diffusive subranges. For $\gamma_\alpha \neq 0$, we find $G_\alpha < 0.5$ Min $(G, E, C_\varphi / \tau)$, i.e., the appearance of scalar gradients always leads to a reduction of the mixing frequency, which may be remarkable. This is illustrated in Figure 2, where the dependence of G_α on C_φ / τ, E, G and γ_α is illustrated. At $\gamma_\alpha = 0$, $2 G_\alpha / (C_\varphi / \tau)$ is equal to $E / (C_\varphi / \tau)$, provided that $E / (C_\varphi / \tau) \leq 1$. For $E / (C_\varphi / \tau) \geq 1$, we find $2 G_\alpha / (C_\varphi / \tau) = 1$. For $\gamma_\alpha \neq 0$, the composition frequency decreases in both cases: The appearance of scalar gradients γ_α leads to a greater amount of organized motion, so that the amount of unordered motion (the molecular diffusion represented by G_α) decreases.

The required calculation of γ_α can be simplified by relating $\langle u_j c_\alpha \rangle$ algebraically to the scalar gradient according to (1a-c). This leads to $\gamma_\alpha = \lambda \tau (q^2 / \sigma_\alpha^2) [\partial \langle C_\alpha \rangle / \partial x_j]^2$, where $\lambda = 8 C_0 / [(3 C_0 + 2)(3 C_0 + 2 + 4 G_\alpha \tau)]$. This expression for γ_α is applied in the simulations presented next.

APPLICATION TO MIXING AND PARALLEL CHEMICAL REACTIONS IN A PIPE

The general mixing model derived as described in the previous section represents a Lagrangian stochastic model that satisfies exactly Baldyga's Eulerian composition covariance transport equations, which are tested for different flows. This fact provides evidence for its good performance. We illustrate this performance in respect to the Baldyga's (1994) pipe flow experiments for two reasons: first, to demonstrate the applicability of the algebraic version AMSIEM of the general mixing model, and second, to show the remarkable effect of the consideration of mixing processes at the Kolmogorov- and Batchelor-scale.

Experimental investigations were carried out in a tubular reactor with an inner diameter of D = 32 mm equipped with a concentrically located tube with an inner diameter of 1.81 mm and an outer diameter of 2.52 mm, see Figure 3. The mean pipe velocity $\langle U_1 \rangle$ varied from 0.469 m s^{-1} to 2.19 m s^{-1}, which corresponds to a change of the pipe Reynolds number $Re = \langle U_1 \rangle D / \nu$ from 15000 to 70000, where the kinematic viscosity $\nu = 10^{-6}$ m^2 s^{-1}. The investigation of the mixing and reaction of species were performed by introducing a premixture of hydrochloric acid (B = HCl) and ethyl chloroacetate (C = CH2ClCOOC2H5) over the reactor cross-sectional area. A solution of sodium hydroxide (A = NaOH) was fed through the concentrically located injector. These species react according to

$$A + B \xrightarrow{k_1} R, \qquad A + C \xrightarrow{k_2} S, \tag{3}$$

where $k_1 \to \infty$, k_2 = 23 dm^3 / (mol s) at a temperature T = 293 K. R and S are the reaction products. This reaction scheme represents the structure of many important chemical process engineering or environmental applications. As shown below, the efficiency of chemical conversions according to (3) may depend very sensitively on mixing. This efficiency can be evaluated through the final selectivity X_s of forming S, which is proportional to the decrease of the C-concentration along the reactor

$$X_s = \frac{\overline{C}_{C0} - \overline{C}_{C,out}}{\overline{C}_{A0}}, \tag{4}$$

where $\overline{C}_{C0}$, $\overline{C}_{A0}$ and $\overline{C}_{C,out}$ are the inlet concentrations of C and A and the outlet concentration of C, respectively, averaged over the reactor diameter. These quantities were measured chromatographically.

To describe this pipe flow, the Lagrangian equation (1a-c) were closed by $\tau = C_\mu D (q^2 / 2)^{-1/2}$ and $\tau_\eta = (C_\mu D \nu)^{1/2} (q^2 / 2)^{-3/4}$, where C_μ = 0.09. In that way, the ratio of these time scales $Re_1 = \tau / \tau_\eta$ is found as a robust function of the TKE, which is advantageous for these simulations. Boundary conditions are provided according to the approach of Dreeben and Pope (1997). The simulations were carried out by means of the code PDF2DV (Pope, 1994b). The domain is discretized into (radial) 18 and (streamwise axial) 75 cells.

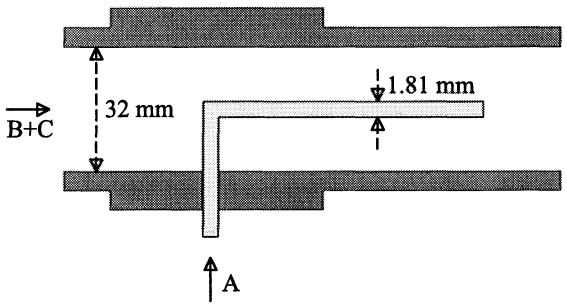

Figure 3. Illustration of the pipe flow geometry and feed streams A, B and C.

A nonuniform grid spacing was applied in the streamwise direction, so that the length of the first cell was five times smaller than that of the last cell and approximately of the same size as the radial cell length. In that grid, the radial extent of the source corresponds to the radius of the first grid cell. The extent of the computational domain was 10 D in the streamwise direction. The mean particle streamwise velocity at the inlet region was derived from the Reynolds number by $\langle U_1 \rangle = Re\ \nu / D$, and the radial mean particle velocity was set to zero. For the turbulence intensity we applied, independent of Re, 0.16 at the inlet region. The influence of variations of this value on the results is lower than 0.2%. The initial values for τ and τ_η were determined according to the parametrizations given above. It was proved that the flow field quantities were obtained in accord with the features found in measurements and DNS (Eggels, 1994, Eggels et al., 1994, Toonder and Nieuwstadt, 1997). A detailed description of the performance of the same code for similar conditions can be found elsewhere (Dreeben and Pope, 1997).

Calculations of X_s according to (4) were performed for Re-variations between 15000 and 70000 and various initial concentrations for A, B and C. The mixing was calculated by the AMSIEM (2). The results of these simulations are presented in Figure 4. The fact that in this experiment the selectivity X_s depends on the Reynolds number Re can be explained as follows. Close to the injector, there is a lot of A compared to B + C. After consumption of part of A by the reaction with B there remains A to participate in the second reaction. The higher $Re = \langle U_1 \rangle D / \nu$ (or the inlet flow velocity, respectively), the smaller is the selectivity X_s due to the slower chemistry between A and C. This dependence of the chemistry on the Reynolds number can be made explicit by means of the Damkoehler number $Da_C = k_2 \overline{C}_A \tau = \xi\ k_2 \overline{C}_A D^2 / (Re \nu)$ for the reaction between A and C, where $\xi = C_\mu (q^2 / 2 \langle U_1 \rangle^2)^{-1/2}$ is introduced. This quantity is proportional to the inverse turbulence intensity. At the centreline, we find by means of (1a-b) in an algebraic approximation $\xi = x_1 / (2D) + \xi_0$. The value of ξ at the streamwise position $x_1 = 0$ is denoted by ξ_0, which is independent of Re. Hence, we find that Da_C is inversely

proportional to Re,

$$Da_C = \frac{k_2 \overline{C}_A D^2}{Re\,\nu}\left[\frac{x_1}{2D} + \xi_0\right]. \qquad (5)$$

The effect of Da_C can also be observed by comparing Figure 4a and 4b. Due to the chosen initial concentration of A, Da_C related to Figure 4b is twice of the value of Da_C related to Figure 4a. At Re = 15000, the inlet values of these Damkoehler numbers are Da_C = 0.1 and Da_C = 0.05, respectively.

The comparison with the measurements reveals the good performance of the AMSIEM model. The calculation of the mixing with the usually applied mixing rate C_φ / τ instead of G_α, leads to a remarkable underestimate of the selectivity X_s, which amounts to approximately 50% for Re = 15000. The reason for this difference is given by the fact that C_φ / τ provides a too strong mixing between A and B + C. In that case, X_s decreases due to the infinitely fast destruction of A through reaction with B. The overprediction of mixing through C_φ / τ is illustrated in Figure 5, where the ratios $E / (C_\varphi / \tau) = 0.029\ Re_\lambda$ and $G_\alpha / (C_\varphi / \tau)$ are shown along the centreline for Re = 15000 and Re = 70000. The normalized composition frequency of the model that applies C_φ / τ as mixing rate is given by 1. The values of $E / (C_\varphi / \tau)$ smaller than 1 indicate for the different Reynolds numbers the relevance of Re-effects. For $\gamma_\alpha = 0$, the calculated mixing frequency $2\,G_\alpha$ would be equal to E, but the appearance of a streamwise scalar gradient in conjunction with small values for the scalar variance results in values of $\gamma_\alpha / (C_\varphi / \tau)$ near 0.5 in the reaction zone, which leads to a smaller mixing frequency G_α, see Figure 2.

SUMMARY

In contrast to previously applied Lagrangian PDF methods, the mixing model presented here is shown to be applicable to the calculation of the multi-scale turbulent mixing in inhomogeneous liquid-phase reacting flows. This is of relevance for both chemical engineering applications and the further development of models that describe multi-scale mixing in multi-phase flows. The methodology used here to construct the mixing model can be applied in consistency with other Eulerian variance transport equations and Lagrangian (frame) models than the simple models used here to illustrate the approach. Additionally, the presented description of micromixing can be adopted in computationally less-demanding PDF methods (Fox, 1998).

The mixing model in its general formulation can be seen as proved for different flows through its full consistency with the well-tested equations of Baldyga. The good performance of its algebraic version, i.e., the AMSIEM model, was illustrated. An interesting result from a theoretical point of view is the fact that scalar gradients may lead to a drastic reduction of the composition frequency. Figure 4 in conjunction with relation (5) reveals, first, the relevance of considering Re- and Da-effects, and, second,

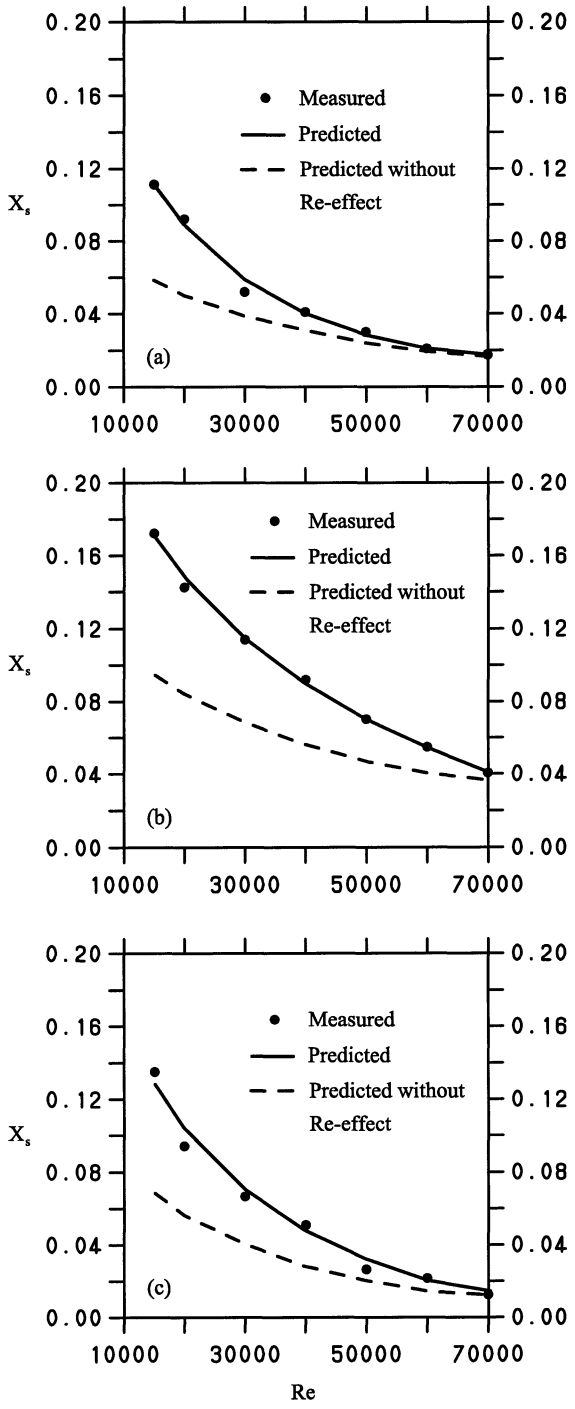

Figure 4. Product distribution X_s vs. Re. The initial feed concentrations of A, B and C are: (a) C_{A0} = 0.45 mol / dm^3, $C_{B0} = C_{C0}$ = 0.009 mol / dm^3, (b) C_{A0} = 0.9 mol / dm^3, $C_{B0} = C_{C0}$ = 0.018 mol / dm^3, (c) C_{A0} = 0.45 mol / dm^3, $C_{B0} = C_{C0}$ = 0.014 mol / dm^3. The dashed line gives the result of the model that applies C_φ / τ as mixing rate.

that the difference to methods that neglect such effects may be larger for flows with lower Reynolds numbers than considered here.

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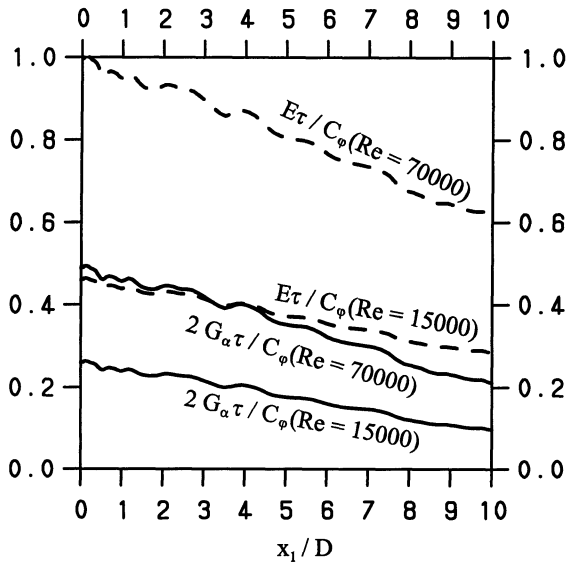

Figure 5. The composition frequency G_α and the Kolmogorov-scale rate E normalized to C_φ / τ along the pipe centreline for Re = 15000 and Re = 70000.

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