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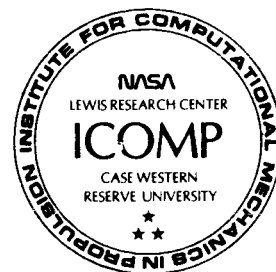
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# ON THE COALESCENCE-DISPERSION MODELING OF TURBULENT MOLECULAR MIXING

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## SUMMARY

The general coalescence-dispersion (C/D) closure provides phenomenological modeling of turbulent molecular mixing. The models of Curl (ref. 1) and Dopazo and O'Brien (ref. 2) appear as two limiting C/D models that "bracket" the range of results one can obtain by various models. This finding is used to investigate the sensitivity of the results to the choice of the model. Inert scalar mixing is found to be less model-sensitive than mixing accompanied by chemical reaction. Infinitely fast chemistry approximation is used to relate the C/D approach to Toor's earlier results (ref. 3). Pure mixing and infinite rate chemistry calculations are compared to study further a recent result of Hsieh and O'Brien (ref. 4) who found that higher concentration moments are not sensitive to chemistry.

## 1. INTRODUCTION

The modeling of molecular mixing is a known stumbling block of turbulent combustion theory (O'Brien, ref. 5). Until recently three closure models were available: the closure introduced by Dopazo and O'Brien (ref. 2), the closure of Janicka et al. (ref. 6), and the coalescence-dispersion model of Curl (ref. 1).

Since Curl's mixing model was developed within a different context its use for closure is a phenomenological step that can only be rationalized by comparison to data and some physical reasoning. In view of some encouraging results (Pope, refs. 7 and 8; Wu and O'Brien, ref. 9; Kollmann and Janicka, ref. 10; Givi et al., refs. 11 and 12; Nguyen and Pope, ref. 13; Hsieh and O'Brien, ref. 4) and of the calculational advantages of the model (Pope, ref. 14), there is a clear incentive to investigate, and possibly improve, its validity.

In the wake of the earlier publications of Pope (ref. 15), Janicka et al. (ref. 6), and Dopazo (ref. 16), Pope (ref. 17) introduced a generalized coalescence-dispersion (C/D) model. The closure models of Janicka et al. (ref. 6) and Curl (ref. 1) can be recovered from the generalized model as

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special cases. Recently Kosaly (ref. 18) showed that the Dopazo-O'Brien (ref. 2) closure recovers from the general C/D closure model.

By introducing a generalized C/D model Pope (ref. 17) introduced a family of models with the earlier models appearing as "members of the same family." This step made C/D modeling more flexible than before. The existence of numerous models allows more room for data fitting and physical reasoning.

Since we have now numerous C/D models, the problem of sensitivity is to be addressed. Are the final results sensitive to the differences between different models? Are the differences large enough to invoke experimental studies to decide between various models? What kind of experiments are to be performed to decide between the different models? This paper addresses these questions.

The present paper discusses general coalescence-dispersion modeling. The necessary background is reviewed in section 2. Section 3 discusses the mixing of an inert scalar in the spatially homogeneous case and in the strongly space dependent case of a round jet discharging into quiescent surroundings. C/D modeling in the limiting case of infinitely fast chemistry is investigated in section 4. In this section, the C/D results are compared to the noted result of Toor (refs. 3, 19, and 20) who established a relationship between the average reactant concentration and the decay of the variance without reaction.<sup>1</sup> Section 5 discusses the behavior of the higher moments of concentration in the fast chemistry case and in the case of pure mixing. The main results of the work are summarized in section 6.

## 2. BACKGROUND

We first consider the simple case of a passive scalar inhomogeneous incompressible turbulent flow and simplify the matter further by assuming that the scalar is initially homogeneously distributed in space. Let  $\varphi(\underline{x}, t)$  be the scalar and  $p(\psi, t)$  its pdf.<sup>2</sup>

The general coalescence-dispersion (C/D) model is defined by the following evolution equation (Pope, ref. 17):

$$\frac{\partial p(\psi, t)}{\partial t} = -2\beta\omega p(\psi, t) + 2\beta\omega \iint_{-\infty}^{+\infty} d\psi' d\psi'' p(\psi', t)p(\psi'', t) \\ \times \int_{-\infty}^{+\infty} d\alpha A(\alpha) \delta\left[\psi - (1 - \alpha)\psi' - \frac{1}{2}\alpha(\psi' + \psi'')\right] \quad (1a)$$

The function  $A(\alpha)$  is zero outside  $[0, 1]$ , nonnegative and normalized to unity within  $[0, 1]$ . The parameter  $\beta$  is defined by Pope (ref. 17) as

<sup>1</sup>Whereas the original derivations are contained in references 19 and 20, most of the time we will refer to reference 3 where Toor reviews both the earlier derivations and the related experimental work.

<sup>2</sup>Throughout the paper we will interchangeably refer to this case as the homogeneous (temporal) problem or the plug-flow (spatial) problem (Hill, ref. 21).

$$\beta = \frac{1}{a_1 - \frac{1}{2} a_2} \quad (1b)$$

$$a_m = \int_0^1 \alpha^m A(\alpha) d\alpha \quad (1c)$$

In the Monte-Carlo simulation of equation (1a), the random variable  $\alpha$  controls the extent of mixing which takes place when a "particle-pair" is selected for interaction.  $A(\alpha)$  is the probability density function of  $\alpha$  (Pope, ref. 17). The present paper discusses the evolution of  $p(\psi, t)$  regardless of the numerical technique applied for solution, therefore there will be no reference to "particles" in this work.

The RHS of equation (1a) represents molecular mixing in the C/D approximation. The convective term is missing from the equation because of the assumption of spatial homogeneity.

Multiplying equation (1a) by  $\psi^2$  and integrating, the time evolution of the standard deviation can be obtained. The result is

$$\sigma(t) = \sigma(0) \exp \left[ - \int_0^t \omega(t') dt' \right] \quad (2)$$

Equation (2) identifies the mixing frequency as

$$\omega(t) = \frac{3D}{\lambda_\varphi^2(t)} \quad (3)$$

where  $D$  is the diffusion-constant and  $\lambda_\varphi(t)$  is the Taylor-length associated with the mixing of the scalar. (Tennekes and Lumley, ref. 22).

The present work does not address the problems related to the determination of the Taylor-length, that is, we assume that the mixing frequency is known. Regarding the evaluation of the mixing frequency we refer to the literature (Corrsin, ref. 23; Beguier et al., ref. 24; Pope, ref. 25).

In order to complete the closure the function  $A(\alpha)$  is to be specified. different choices of  $A(\alpha)$  result in different C/D models. The original Curl-model (ref. 1) can be recovered by employing  $A(\alpha) = \alpha(\alpha - 1)$ . The closure suggested by Janicka et al. (ref. 6) is equivalent to using  $A(\alpha) = 1$  within  $0 \leq \alpha \leq 1$ . There is obviously an infinite number of choices, each leading to a different C/D model.

In a recent paper Kosaly (ref. 18) considered the case when the mixing frequency is independent of time and initial pdf is the sum of two delta functions at  $\psi = \pm 1$ .

$$p(\psi, 0) = \frac{1}{2} \delta(\psi - 1) + \frac{1}{2} \delta(\psi + 1) \quad (4)$$

From equations (1a) and (4) the short time form of the pdf was derived.

$$p(\psi, t) = \frac{1}{2} [\delta(1 - \psi) + \delta(1 + \psi)](1 - \beta\omega t) + F(\psi)\beta\omega t + O(\omega^2 t^2) \quad (5)$$

$$F(\psi) = \frac{1}{2} [A(1 - \psi) + A(1 + \psi)]$$

The first term on the RHS describes the gradual decrease of the spikes at  $\psi = \pm 1$ . The second term accounts for the build up of the pdf in the initial phase of mixing. The appearance of the  $A(\alpha)$  function in the second term shows that the behavior of this function is intimately related to the physics of mixing for  $\omega t \ll 1$ .

Physical arguments based on equation (5) led Kosaly (ref. 18) to the conclusion that any physically acceptable  $A(\alpha)$  function has to peak at  $\alpha = 0$  and decrease monotonically in  $0 < \alpha < 1$ .

Since neither  $A(\alpha) = \delta(\alpha - 1)$ , nor  $A(\alpha) = 1$  satisfy the above physical requirement, they are clearly unphysical choices. On the other hand, it is not clear, how far the end results are sensitive to the choice of  $A(\alpha)$ . Indeed Kollmann and Janicka (ref. 10) report weak sensitivity to this choice in the case of the mixing of a passive scalar in turbulent shear flow. Hsieh (ref. 26) applies the models of Janicka et al. (ref. 6) and Curl (ref. 1) to the half-heated grid problem and finds similar results, while the Curl approach is usually less demanding on computer resources. The present paper discusses the above sensitivity issue via the examples of inert scalar mixing and the conserved scalar approach to reactive turbulent flows with fast chemistry.

As further background information we review a result of Kosaly (ref. 18) who showed that if  $A(\alpha)$  has a sufficiently narrow peak at  $\alpha = 0$ , in the sense that<sup>3</sup>

$$\frac{a_2}{a_1} \ll 1 \quad (6)$$

then equations (1a) and (1b) go over into

$$\frac{\partial p(\psi, t)}{\partial t} = \omega(t) \frac{\partial}{\partial \psi} [(\psi - \langle \varphi \rangle) p(\psi, t)] + O\left(\frac{a_2}{a_1}\right) \quad (7)$$

Here  $\langle \varphi \rangle$  represents the ensemble average of the random process  $\varphi(\underline{x}, t)$ . Under the present assumptions this average is independent of time and position. The mixing frequency, however, may depend on time.

Neglecting  $O(a_2/a_1)$  on the RHS, equation (7) becomes identical to the approximate evolution equation derived by Dopazo and O'Brien (ref. 2). It turns out therefore that the Dopazo-O'Brien prediction is equivalent to C/D modeling via an  $A(\alpha)$  function with a sufficiently narrow peak at  $\alpha = 0$ ,

<sup>3</sup>Note that  $a_2 > a_3 > a_4 \dots$ , therefore for  $n > 2$ ,  $a_n/a_1 < a_2/a_1$ .

where "narrowness" is defined by equation (6). The result is indeed independent of the actual choice of  $A(\alpha)$ . For the sake of mathematical convenience we will be using

$$A(\alpha) = \delta(\alpha - \epsilon), \quad \epsilon \rightarrow +0 \quad (8)$$

to recover the Dopazo-O'Brien model from the general C/D formalism.

We are now in the position to derive general results referring to all existing closure models. It is interesting to note that the models of Curl (ref. 1) and Dopazo and O'Brien (ref. 2) appear as two opposite limiting cases.  $A(\alpha) = \delta(\alpha - 1)$  is clearly the most extreme of all the curves who peak at  $\alpha = 1$  and decrease as  $\alpha$  decreases to zero.  $A(\alpha) = \delta(\alpha - \epsilon)$ ,  $\epsilon \rightarrow +0$ , on the other hand, is the extreme representative of the physically acceptable shapes who peak at  $\alpha = 0$  and decrease with increasing values of  $\alpha$ . The  $A(\alpha) = 1$ ,  $0 \leq \alpha \leq 1$  choice (Janicka et al., ref. 6) can be considered to be halfway between the two extreme choices. In the forthcoming numerical work we will use the above three choices to investigate the sensitivity of the results to the  $A(\alpha)$  model.

The fact that the Dopazo-O'Brien model can be presented as one of the C/D models should not create the misunderstanding that it is arbitrary as, e.g., the Curl-model. While Curl's model has never been derived theoretically as a closure model for turbulent molecular mixing, the Dopazo-O'Brien closure does follow from first principles even if under rather limiting assumptions (Dopazo and O'Brien, ref. 2); Pope, ref. 27; and O'Brien ref. 5). On the other hand, using e.g., Monte-Carlo techniques, Curl's model proves to be the least demanding on computer resources, while the strict implementation of the Dopazo-O'Brien closure requires infinite computing time. This circumstance emphasizes further the need for sensitivity studies.

### 3. MIXING OF AN INERT SCALAR

In recent years several authors used C/D modeling to predict experimental data (Wu and O'Brien, ref. 9; Kollmann and Janicka, ref. 10; Givi et al., refs. 11 and 12; Nguyen and Pope, ref. 13; Hsieh and O'Brien, ref. 4). For  $A(\alpha)$  either the Curl model (ref. 1) or the model of Janicka et al. (ref. 6) were used, both of which are physically objectionable. In spite of this, the predictions turned out to be fairly encouraging indicating that the calculated end results may not be sensitive to the  $A(\alpha)$ -shape.

In order to seek insight into the sensitivity problem we first consider the case described by equation (1a), that is, the mixing of an initially homogeneously distributed inert scalar in incompressible, homogeneous turbulent flow. In this case the evolution of the pdf is governed exclusively by molecular mixing and not by the combined influence of molecular mixing and turbulent convection. The spatially homogeneous case is therefore more sensitive to the details of the molecular mixing model than the space dependent cases.

If the initial scalar field is binary, equation (5) demonstrates that immediately after the mixing starts the shape of the pdf is directly determined by the actual  $A(\alpha)$  shape.

In order to study the problem for finite times we define the moments, central moments and standardized moments of  $\varphi(\underline{x}, t)$ , in order, as

$$\langle \varphi^n \rangle_t = \int_{-\infty}^{+\infty} \psi^n p(t, \psi) d\psi \quad (9a)$$

$$\mu_n(t) = \langle (\varphi - \langle \varphi \rangle_t)^n \rangle_t = \int_{-\infty}^{+\infty} (\psi - \langle \varphi \rangle_t)^n p(\psi, t) d\psi \quad (9b)$$

$$\hat{\mu}_n(t) = \frac{\mu_n(t)}{\sigma^n(t)} \quad (n = 1, 2, \dots) \quad (9c)$$

Here  $\sigma(t) = \sqrt{\mu_2(t)}$ , is the standard deviation of the scalar.

We simplify the mathematics by assuming that the mixing frequency does not depend on time and obtain from equations (1a) to (1c) the following results:

$$\langle \varphi \rangle_t = \langle \varphi \rangle_0 \quad (10a)$$

$$\mu_2(t) = \sigma^2(t) = \sigma^2(0)e^{-2\omega t} \quad (10b)$$

$$\mu_3(t) = \mu_3(0)e^{-3\omega t} \quad (10c)$$

$$\mu_4(t) = e^{-4\omega t} \left[ \left( \langle \varphi^4 \rangle_0 - 4\langle \varphi \rangle_0 \langle \varphi^3 \rangle_0 + 3\langle \varphi^2 \rangle_0^2 \right) e^{\gamma \omega t} - 3 \left( \langle \varphi^2 \rangle_0^2 - 2\langle \varphi^2 \rangle_0 \langle \varphi \rangle_0^2 + \langle \varphi \rangle_0^4 \right) \right] \quad (10d)$$

Here  $\gamma$  is related to the moments of  $A(\alpha)$  as (Kosaly, ref. 18)

$$\gamma = \frac{a_2 + \frac{1}{4} a_4 - a_3}{a_1 - \frac{1}{2} a_2} \quad (10e)$$

The skewness ( $\hat{\mu}_3(t)$ ) and the kurtosis ( $\hat{\mu}_4(t)$ ) can be written as

$$\hat{\mu}_3(t) = \hat{\mu}_3(0) \quad (11a)$$

$$\hat{\mu}_4(t) = (3 + \hat{\mu}_4(0))e^{\gamma \omega t} - 3 \quad (11b)$$

The results refer to arbitrary initial distribution of the scalar and arbitrary  $A(\alpha)$  shape. Inserting  $A(\alpha) = \delta(\alpha - 1)$  the early results of Curl (ref. 1) can be recovered.

Equations (10a) to (10c) show that the first three moments are independent of the  $A(\alpha)$  choice. It is only the fourth moment that depends on the  $A(\alpha)$  shape through the parameter  $\gamma$ . All models with  $\gamma > 0$  yield  $\mu_4 \rightarrow \infty$  as  $t \rightarrow \infty$ .

Table I shows the  $\gamma$  values corresponding to  $A(\alpha) = \delta(\alpha - 1)$ ,  $A(\alpha) = 1$  and  $A(\alpha) = \delta(\alpha - \epsilon)$ ,  $\epsilon \rightarrow +0$ . In the third column the corresponding kurtosis ( $\mu_4$ ) values are given at  $\omega t = 1.0$ , assuming binary initial distribution. While the  $A(\alpha)$  shapes of Curl (ref. 1) and Janicka et al. (ref. 6) differ considerably, the corresponding  $\gamma$  values are relatively close, resulting in a mere 21 percent difference between the kurtosis values. This difference is indeed small compared to the corresponding difference between the Curl and the Dopazo-O'Brien values. This finding suggests that, no matter how different the models of Curl (ref. 1) and Janicka et al. (ref. 6) seem to be, one does not expect substantial differences when applying these two models (Hsieh, ref. 26). Differences can, however, be expected between Curl's model and the models with powerful peaks at  $\alpha = 0$ , represented in Table I by the extreme case of the Dopazo-O'Brien model.

Figure 1 shows the pdfs at  $\omega t = 0.96$  calculated using a Monte-Carlo routine via the models of Curl (ref. 1) and Janicka et al. (ref. 6) and a third pdf corresponding to  $A(\alpha) = \delta(\alpha - \alpha_0)$ ,  $\alpha_0 = 0.1$ .<sup>4</sup> The latter shape was chosen to represent the Dopazo-O'Brien (ref. 2) closure, while keeping the computational time within acceptable limits.<sup>5</sup> Since one knows that with the binary initial condition given in equation (4) the Dopazo-O'Brien pdf can be written as (O'Brien, ref. 5).

$$p(\psi, t) = \frac{1}{2} \delta(e^{-\omega t} - \psi) + \frac{1}{2} \delta(e^{-\omega t} + \psi) \quad (11)$$

it is easy to recognize the two peaks as approximations of the two delta functions at  $\psi = \pm e^{-\omega t} = \pm 0.38$ .

Figure 1 suggests similar conclusions as the data in Table I. Except for the sharp peak at  $\psi = 0$  in the Curl pdf, the models of Curl (ref. 1) and Janicka et al. (ref. 6) predict roughly similar behavior. They, however, both differ markedly from the Dopazo-O'Brien (ref. 2) curve, which is to be considered as an extreme representative of the models peaking at  $\alpha = 0$ .

The time evolution dictated by equation (11) is clearly nonphysical (O'Brien, ref. 5). This does not necessarily mean that the Dopazo-O'Brien model may not give correctly moments, or even the pdfs, in cases that are more involved than the mixing of two spikes in homogeneous turbulence (Dopazo, ref. 16).

We consider next a two-dimensional, stationary, incompressible, turbulent round jet discharging into quiescent air. Injected into the flow is an inert scalar whose value is taken to be unity in the nozzle and zero in the surrounding air. We present the pdf equation in its general form, although polar-cylindrical coordinates with azimuthal symmetry will be used.

Let again  $\varphi(\underline{x}, t)$  be the scalar and  $p(\psi, \underline{x})$  its pdf. The pdf does not depend on time because of stationarity. We write the pdf equation using the

<sup>4</sup>Note that for binary mixing Curl's model yields a discrete distribution (ref. 17). The continuous line in figure 1 represents the normalized histograms in the usual way.

<sup>5</sup>In the numerical simulations, the pdf is modeled by an ensemble of 20 000 Monte-Carlo elements. The slight asymmetry of the pdf around  $\psi = 0$  is due to the statistical errors associated with the finite number of computational elements.



gradient diffusion approximation to model the convective term and the generalized C/D model to account for molecular mixing (Nguyen and Pope, ref. 13).

$$\frac{\partial}{\partial x_i} (U_i p) = \frac{\partial}{\partial x_i} \left( v_T \frac{\partial p}{\partial x_i} \right) + MM(\psi, \underline{x}) \quad (12)$$

Here  $U_i (i = 1, 2, 3)$  are the components of the time averaged velocity,  $v_T$  is the eddy diffusivity of the scalar.  $MM(\psi, \underline{x})$  is defined by the RHS of equation (1a). Equation (1a) can be recovered from equation (12) by setting the eddy-diffusivity term equal to zero and writing  $U_2 = U_3 = 0$  in the convective term ( $x_1 = U_1 t$ ).

The pdf equation is supplemented by the modeled equations for the mean flow and the  $k - \epsilon$  transport equations. The eddy-diffusivity and the mixing frequency are calculated as

$$v_T = 0.09 \frac{k^2}{\epsilon}, \quad \omega = \frac{\epsilon}{k} \quad (13)$$

For more details we refer to Jones and Whitelaw (ref. 28) and Givi et al. (ref. 11).

The closure is again not complete without specifying the  $A(\alpha)$ -function within the molecular mixing term. Results will be shown using the same three models as before. The Dopazo-O'Brien model again will be represented by  $A(\alpha) = \delta(\alpha - \alpha_0)$ ,  $\alpha_0 = 0.1$ .

Figures 2(a) to (d) show the radial dependence of the first four moments of the scalar at  $x = 10D$  downstream from the nozzle for the different  $A(\alpha)$  choices. (The second, third, and fourth moments on the figures are divided by the respective powers of the first moment at the centerline of the jet.)  $R$  is the radial distance divided by the jet half-width  $r_{1,2}(x)$ . The latter quantity is defined as the radial position at which the mean axial velocity is equal to the arithmetic average of the centerline and free stream velocities.

Whereas in the spatially homogeneous case (plug-flow reactor) the time (space) dependence was determined exclusively by molecular mixing, presently convection plays an important role. We expect therefore the results be less sensitive to the choice of  $A(\alpha)$  than before.

Figures 2(a) to (c) show that the first three moments are model independent, an obvious result, since these moments were found to be independent of the  $A(\alpha)$  shape even in the homogeneous case. (cf. eqs. (10a) to (10c).) Figure 2(d) shows, however, that the fourth moment is different for the different models, as expected (cf. eqs. (10d) and (10e)). The figure again demonstrates the approximate equivalence of the Curl and Janicka et al. models and the opposite extreme nature of the Curl model vis-a-vis the Dopazo and O'Brien closure.

The influence of the difference in the fourth moments can be clearly seen in figure 3. Figures 3(a) to (c) show the pdf of the scalar across the width of the shear layer at  $x = 10D$ . Figure 3(d) presents the pdf at  $x = 10D$  and at the radial location  $R = 0.8$ . The smaller the fourth moment, the smaller the kurtosis value becomes, making thereby the Dopazo-O'Brien pdf markedly flatter than the Curl-curve.

Generally speaking, however, the difference between the Curl and the Dopazo-O'Brien pdfs is not as dramatic as seen on figure 1. This is due to the influence of the convection term which diminishes the sensitivity of the results to the details of the molecular mixing model.

By considering mixing in homogeneous flow and in a two-dimensional jet we are looking at two rather extreme cases. The two-dimensional jet case is extremely insensitive to the details of the molecular mixing model. One expects to see stronger dependence on  $A(\alpha)$  in nearly homogeneous cases like temperature mixing in incompressible flow over a half heated grid (Wu and O'Brien, ref. 9).

#### 4. REACTIVE FLOW IN THE FAST CHEMISTRY LIMIT

We consider a second-order, irreversible, one-step, isothermal reaction of the type



and assume that the turbulence is homogeneous and incompressible and ignore any dynamic or chemical role the products may play. We assume furthermore that the two species are initially homogeneously distributed in space. Let  $A(\underline{x}, t)$  and  $B(\underline{x}, t)$  be the concentrations of the species and  $p_A(c, t)$ ,  $p_B(c, t)$  and  $p_{A,B}(c', c'', t)$  their marginal and joint pdfs, respectively.

It was shown by several authors (Toor, ref. 3; O'Brien, ref. 29) that if the diffusion coefficients of the two species are equal ( $D_A = D_B = D$ ), then

$$J(\underline{x}, t) \equiv A(\underline{x}, t) - B(\underline{x}, t) \quad (15)$$

is a conserved scalar, that is, it satisfies the diffusion equation without chemical source term.

In case of infinitely fast chemistry (Toor, ref. 3; O'Brien, ref. 29, Bilger, ref. 30), the random concentration fields of the two reactants can be related to the  $J(\underline{x}, t)$  as

$$A = \begin{cases} J; & J > 0 \\ 0; & J \leq 0 \end{cases} \quad B = \begin{cases} 0; & J \geq 0 \\ -J; & J < 0 \end{cases} \quad (16)$$

Following O'Brien (ref. 29), we write

$$\begin{aligned} p_A(c, t) &= p_J(c, t) + k_A(t)\delta(c) \\ p_B(c, t) &= p_J(-c, t) + k_B(t)\delta(c) \end{aligned} \quad (17a)$$

Here  $c \geq 0$ ,  $p_J(c, t)$  is the pdf of  $J(\underline{x}, t)$  and

$$k_A(t) = \int_0^{\infty} p_J(-c, t) dc = \int_{-\infty}^0 p_J(c, t) dc \quad (17b)$$

$$k_B(t) = \int_0^{\infty} p_J(c, t) dc = 1 - k_A(t)$$

Let us assume not that at  $t = 0$  the two species are totally segregated. We write the joint pdf at the initial time,

$$p_{A,B}(c', c'', 0) = W_A \delta(c' - c_{A0}) \delta(c'') + W_B \delta(c') \delta(c'' - c_{B0}) \quad (18)$$

$$W_A + W_B = 1$$

Here  $c_{A0}$ ,  $c_{B0}$  are the respective initial concentrations of the two species. The weight factors  $W_A$ ,  $W_B$  represent area-ratios at the inlet in the plug-flow case.

Using equation (18) the initial distributions of A and B become

$$p_A(c, 0) = W_A \delta(c - c_{A0}) + W_B \delta(c) \quad (19a)$$

$$p_B(c, 0) = W_B \delta(c - c_{B0}) + W_A \delta(c) \quad c > 0 \quad (19b)$$

We compare equations (19a) and (19b) to equation (17a) and obtain

$$p_J(c, 0) = W_A \delta(c - c_{A0}) + W_B \delta(c + c_{B0}) \quad -\infty < c < \infty \quad (20)$$

Let us assume furthermore that the reacting species are present in stoichiometric proportion and write:

$$W_A = W_B = \frac{1}{2} \quad c_{A0} = c_{B0} = c_0 \quad (21)$$

Because of equation (21),  $p_J(c, t)$  is an even function of  $c$  for  $t \geq 0$ . We introduce the new variable  $\psi = c/c_0$  and obtain from equations (17a), (17b), (20), and (21):

$$p_A(\psi, t) = p_B(\psi, t) = p_J(\psi, t) \frac{1}{2} \delta(\psi) \quad (17')$$

$$p_J(\psi, 0) = \frac{1}{2} \delta(\psi - 1) + \frac{1}{2} \delta(\psi + 1) \quad (20')$$

Equation (20') is the same initial condition that was considered in equation (4). It follows from equations (17') and (20') that

$$\langle A \rangle_t = c_0 \int_0^{\infty} \psi p_J(\psi, t) d\psi \quad (22a)$$

$$\frac{\langle A \rangle_t}{\langle A \rangle_0} = 2 \int_0^{\infty} \psi p_J(\psi, t) d\psi \quad (22b)$$

where the bracket denotes ensemble averaging.

In the general C/D model of molecular mixing the pdf  $p_J(\psi, t)$  satisfies equations (1a) to (1c) and equation (20'). The mixing frequency is related to the diffusion constant and the Taylor-length scale of  $J(\underline{x}, t)$  by equation (3). The closure is to be completed by the choice of an  $A(\alpha)$ -shape.

Let us discuss now a new aspect of  $A(\alpha)$ -modeling by referring to a noted result of Toor (ref. 3), who related the average reactant concentration to the standard deviation of  $J(\underline{x},t)$  by the following relationship:

$$\frac{\langle A \rangle_t}{\langle A \rangle_0} = \frac{\sigma(t)}{\sigma(0)} \quad (23)$$

Here  $\sigma(t)$  is the standard deviation of the scalar  $J(\underline{x},t)$ .

Using equation (2) Toor's results becomes

$$\frac{\langle A \rangle_t}{\langle A \rangle_0} = \exp \left[ - \int_0^t \omega(t') dt' \right] \quad (24)$$

Let us recall that equations (23) and (24) refer to spatial homogeneity, infinite rate chemistry, stoichiometric conditions and equal diffusion constants of the two species. In the derivation Toor (ref. 3) employs Gaussian-shape for the  $p_j(\psi,t)$  but remarks that this assumption may be relaxed. Recent measurements of Bennani et al. (ref. 31), as well as extensive earlier experimental work reviewed by Toor (ref. 3) and Brodkey (ref. 32) support equation (24). It is presently under investigation whether the Toor's result (ref. 3) can be generalized for cases that are more complicated than simple second-order reactions (Brodkey and Lewalle, ref. 33).

Let us examine now whether the Toor-result can be recovered via C/D modeling. Under stoichiometric conditions  $\langle J \rangle_t = \langle A \rangle_t - \langle B \rangle_t = 0$ . Inserting this result into equation (7) and using equation (22a) we obtain the equation governing the time evolution of  $\langle A \rangle_t$ .

$$\frac{d\langle A \rangle_t}{dt} = -\omega(t)\langle A \rangle_t + 0 \left( \frac{a_2}{a_1} \right) \quad (25)$$

Equation (24) recovers if  $a_2/a_1 \rightarrow 0$ . This means that out of the family of the C/D models only the Dopazo-O'Brien approximation leads to the Toor-result.

That other C/D models do not recover equation (24) can be easily seen by considering the mixing frequency independent of time and inserting equation (5) into equation (22b). Simple calculation results in the short time expansion:

$$\frac{\langle A \rangle_t}{\langle A \rangle_0} = 1 - \frac{1}{1 - \frac{1}{2} \frac{a_2}{a_1}} \omega t + 0(\omega^2 t^2) \quad (26)$$

The linear term does not correspond to  $\exp[-\omega t]$  unless  $a_2/a_1 \rightarrow 0$ .

We conclude that -- at least in the context of the fast chemistry approximation -- the application of the Dopazo-O'Brien model is equivalent to Toor's result (ref. 3) represented by equation (24). This means that the numerous experimental studies which investigate the validity of equation (24) can be also used to investigate the Dopazo-O'Brien closure. Indeed, it is only by comparison to data that it will become possible to decide between different molecular mixing models.

The equivalence of the Dopazo-O'Brien (ref. 2) closure and Toor's result (ref. 3) is due to the long time asymptotic nature of both approaches. Theoretical discussion of the problem will be given in forthcoming paper of Kosaly (ref. 34).

It is natural, at this point, to raise again the sensitivity issue. Since we know that the Dopazo-O'Brien (ref. 2) model recovers the Toor-result, let us investigate, how far the other C/D models deviate from it. The sensitivity investigation was performed with time independent mixing frequency.

Figure 4 shows the time evolution of  $\langle A \rangle_t / \langle A \rangle_0$  for different C/D models. The  $A(\alpha) = \delta(\alpha - \alpha_0)$ ,  $\alpha_0 = 0.1$  calculation is not needed this time, since the Dopazo-O'Brien (ref. 2) result was proved to be equivalent to equation (24), and can be represented by a simple exponential. It is the Curl-prediction that deviates the most from the Toor-curve. The results coming from the model of Janicka et al. (ref. 6) are practically identical to the Curl-results (ref. 1). Figure 5 shows the deviation from the Dopazo-O'Brien (Toor) curve. The maximum deviation between the Curl-prediction and the Toor-exponential is -16 percent.

It is important to point out a basic difference between inert mixing and mixing with chemical reaction. The model independency of the first three moments in the inert mixing case limits the sensitivity of the pdf to the choice of the  $A(\alpha)$ -model. The situation is markedly different if mixing is accompanied by chemical reaction. Figures 4 and 5 demonstrate that in this case even the first moment depends on the choice of the molecular mixing closure model. We will show in the next section that model sensitivity is even stronger for the higher moments.

In order to demonstrate the influence of strong space dependence on the deviation between different models we again consider the flow case described in connection with figures 2 and 3. Reactant A is injected into the flow through the nozzle, while B is dispersed in the quiescent surroundings. Figure 6 shows the radial dependence of the average concentration of A at  $x = 10D$  downstream from the nozzle. The concentration is normalized to unity at the nozzle inlet. The radial distance is nondimensionalized as discussed earlier.

The calculation was performed in the fast chemistry approximation using different C/D models to evaluate the pdf of the conserved scalar  $J = A-B$ . The space dependence of the mixing frequency was computed via equation (13). Since equation (24) refers only to the spatially homogeneous case, the Dopazo-O'Brien (ref. 2) model was again represented by  $A(\alpha) = \delta(\alpha - \alpha_0)$ ,  $\alpha_0 = 0.1$ . Figure 6 demonstrates the equivalence of the different C/D models for calculating average concentration in this strongly space dependent case.

## 5. THE HIGHER MOMENTS OF CONCENTRATION WITH AND WITHOUT CHEMICAL REACTION

In a recent work Hsieh and O'Brien (ref. 4) investigated the mixing of two species A and B in the turbulent flow downstream of a grid. The two species were considered to be present in stoichiometric proportion. Species A was introduced on the upper half of the grid and species B on the lower half. The equation for the joint pdf of A and B was closed using a gradient diffusion model for the convection terms and Curl's approach for the

molecular mixing term. Our present interest is related to the axial dependence of their calculated mean, variance, skewness, and kurtosis values.

Hsieh and O'Brien (ref. 4) study the role of the chemical reaction in the behavior of the above quantities numerically by comparing calculated values with and without reaction. The effect of reaction reduces the mean at every axial location, whilst reduces the variance below the centerline and increases above it. The role of reaction in modifying the skewness and kurtosis values is conspicuously muted. The results without reaction compare well with experimental data referring to a half heated grid (Wu and O'Brien, ref. 9; LaRue and Libby, ref. 35). Measured results for the reacting case are not available.

The chemical reaction follows the kinetic scheme  $A + B \rightarrow P$  and is of moderate rate, of order unity, compared to the turbulent time scales in the model. In order to find out whether the interesting behavior of the skewness and the kurtosis is due to moderate rate chemistry or reflects a more general law, Hsieh and O'Brien (ref. 4) suggest studying the issue by comparing results without chemical reaction to corresponding infinite rate chemistry calculations.

In the present section, we investigate the problem in the homogeneous incompressible flow case and assume that the two species are initially segregated and homogeneously distributed in space (cf. eq. (18)). We consider stoichiometric conditions and apply equation (21) to establish the plug-flow counterpart of the geometrical arrangement by Hsieh and O'Brien (ref. 4).

In the infinite rate chemistry case the concentration pdf  $p_A(\psi, t)$  is related to the pdf of  $J$  via equation (17').  $p_J(\psi, t)$  satisfies equations (1a) to (1c) with the initial condition given in equation (20'). Let  $p_{A,m}(\psi, t)$  be the concentration pdf of  $A$  without chemical reaction. (The subscript "m" stands for "mixing only.")  $p_{A,m}(\psi, t)$  will also satisfy equations (1a) to (1c), its initial shape, however, will differ from the initial shape of  $p_J(\psi, t)$ . Using equations (19a) and (21) we write

$$p_{A,m}(\psi, 0) = \frac{1}{2} \delta(\psi - 1) + \frac{1}{2} \delta(\psi) \quad (27)$$

Let us calculate the mean and the central moments of the concentration. We write the  $n$ -th central moment in the infinitely fast reaction case using equation (17'):

$$\begin{aligned} \mu_{n,r}(t) &\equiv \langle (A - \langle A \rangle_{t,r})^n \rangle_{t,r} = \int_0^1 (\psi - \langle A \rangle_{t,r})^n p_A(\psi, t) d\psi \\ &= \int_0^1 (\psi - \langle A \rangle_{t,r})^n p_J(\psi, t) d\psi + \frac{(-1)^n}{2} \langle A \rangle_{t,r}^n \quad (28) \end{aligned}$$

Here  $\langle A \rangle_{t,r}$  is the mean as defined in equation (22a). The subscript  $r$  indicates infinitely fast reaction. (For the sake of simplicity the results are presented in  $c_0 = 1$  units.)

The  $n^{\text{th}}$  central moment without reaction can be written as

$$\begin{aligned} \mu_{n,m}(t) &= \langle (A - \langle A \rangle_{t,m})^n \rangle_{t,m} = \int_0^1 (\psi - \langle A \rangle_{t,m})^n p_{A,m}(\psi, t) d\psi \\ &= \int_0^1 \left( \psi - \frac{1}{2} \right)^n p_{A,m}(\psi, t) d\psi \end{aligned} \quad (29)$$

Writing equation (29) we used the result

$$\langle A \rangle_{t,m} = \frac{1}{2} \quad (30)$$

Since  $p_j(\psi, t)$  and  $p_{A,m}(\psi, t)$  satisfy the same equation their difference is due to their different initial shapes (cf. eqs. (20') and (27)). Simple calculation provides the known result

$$p_j(t, \psi) = \frac{1}{2} p_{A,m} \left( t, \frac{\psi' + 1}{2} \right), \quad \psi = \frac{\psi' + 1}{2}, \quad 0 \leq \psi' \leq 1 \quad (31)$$

Using equations (29) and (31) the  $n$ -th central moment without reaction can be expressed via the pdf  $p_j(\psi, t)$ .

$$\mu_{n,m}(t) = \frac{1}{2^n} \int_{-1}^{+1} \psi^n p_j(\psi, t) d\psi \quad (32)$$

We assume now that the mixing frequency is independent of time and use equation (11) to write  $p_j(\psi, t)$  in the Dopazo-O'Brien approximation (ref. 2):

$$p_j(\psi, t) = \frac{1}{2} \delta(\psi - e^{-\omega t}) + \frac{1}{2} \delta(\psi + e^{-\omega t}) \quad (11')$$

Inserting equation (11') into equations (22a), (28), and (32) straight forward calculation results in

$$\langle A \rangle_{t,r} = \frac{1}{2} e^{-\omega t} \quad (33a)$$

$$\mu_{n,r}(t) = \mu_{n,m}(t) = \frac{1 + (-1)^n}{2^{n+1}} e^{-n\omega t} \quad (33b)$$

Equation (33a) says that the mean with reaction is always smaller than without it ( $\langle A \rangle_{t,m} = 1/2$ , an obvious result. Equation (33b) is more interesting. It says that, in the Dopazo-O'Brien approximation, infinite rate chemistry and pure mixing result in the same central moments. Restricting the discussion to  $n = 2, 3, 4$  we conclude that, in the present model, the variance, skewness, and kurtosis of the concentration do not depend on chemistry.

Let us recall that equation (33b) was derived using the Dopazo-O'Brien (ref. 2) model. Figures 7 and 8 show the variance, skewness, and kurtosis versus dimensionless time for different C/D models.

For each model two curves are shown; one without chemistry, the other with infinite rate chemistry. In the Dopazo-O'Brien (ref. 2) approximation the two

curves coincide (cf. eq. (33b)). Since in the pure mixing limit the variance is model independent, the Dopazo-O'Brien variance represents the pure mixing case for the other two models as well.

Figures 7 and 8 show that both Curl's (ref. 1) model and the approximation of Janicka et al. (ref. 6) provide different results. The equality of the central moments with and without chemistry appears to be characteristic of the Dopazo-O'Brien (ref. 2) closure rather than a general property of C/D modeling.

While Hsieh and O'Brien (ref. 4) calculate radial profiles, our results refer to the axially averaged concentration. This limits the possibility of comparing the two sets of results. Keeping this limitation in mind, we conclude, nevertheless, that the Curl-results seen on figures 7(a) and (b) contradict their finding that the skewness and kurtosis do not depend on chemistry in this model. One speculates that it is the use of moderate rate chemistry that makes the skewness and kurtosis profiles similar to the profiles calculated with pure mixing. Increasing the rate of chemical reactions would probably change the results of Hsieh and O'Brien (ref. 2) considerably.

## 6. SUMMARY AND CONCLUSIONS

A generalized coalescence-dispersion (C/D) model has been introduced by Pope (ref. 17). The closures of Dopazo and O'Brien (ref. 2) and Curl (ref. 1) can be recovered from this general model as limiting cases. Results obtained by these two models "bracket" the range of results one may obtain via any other C/D model.

In case of inert scalar mixing for all C/D models studied the first three moments are the same. The fourth moment may depend quite considerably on the model. It is shown that, no matter how different the models of Curl (ref. 1) and Janicka et al. (ref. 6) seem to be one must not expect substantial difference when applying these two closures. Differences can, however, be expected between Curl's model and an opposite category of models whose extreme version is the closure of Dopazo and O'Brien (ref. 2).

Space dependence diminishes the sensitivity to the molecular mixing model. Since Curl's model is the least burdening on computer resources, once we know that the case is not sensitive to the molecular mixing model it is advisable to use Curl's approach.

In order to judge the sensitivity of the results to the molecular mixing model it is important to point out a basic difference between inert scalar mixing and mixing with chemical reaction. The model independence of the first three moments in the inert mixing case understandably limits the dependence of the pdf on the choice of the molecular mixing model. The situation is markedly different if turbulent mixing is accompanied by chemical reaction. In the present paper the limiting case of infinitely fast chemistry was used to demonstrate that even the first moment of the reactant concentration is sensitive to molecular mixing (figs. 4(a) and (b)). The model dependence of the higher moments is even stronger (figs. 7 to 8).

New insight can be gained into the nature of C/D modeling by relating this method to a result of Toor (ref. 3) who established a relationship between the



average concentration of a reactant and the mixing frequency (cf. eq. (24)). Toor's result was derived assuming spatial homogeneity, second-order, infinite rate chemistry at stoichiometric conditions and equal diffusivities of the two species. Experimental investigations reviewed by Toor (ref. 3) and Brodkey (ref. 32) seem to support Toor's result. This means that the above experimental investigations contain valuable information on the validity of the Dopazo-O'Brien closure.

Hsieh and O'Brien (ref. 4) studied the mixing of two reactants in the turbulent flow downstream of a grid. They investigated the role of chemical reaction by comparing numerically the higher moments calculated with and without reaction. They modeled the convection term by gradient diffusion and the molecular mixing term via Curl's approach (ref. 1). They employed moderate rate chemistry and found that the skewness and kurtosis of the reactant concentration are roughly identical with and without chemistry.

The possible reason behind this finding was investigated by comparing infinite rate chemistry and pure mixing results in the spatially homogeneous case. Straightforward mathematics shows that Dopazo-O'Brien (ref. 2) model results in equal central moments in the presence and in the absence of chemical reaction. The equality of the central moments, however, appears to be a characteristic of the Dopazo-O'Brien (ref. 2) closure only. It does not hold if other models, e.g. (ref. 1), are applied. Since Hsieh and O'Brien (ref. 4) used Curl's model we conclude that their skewness and Kurtosis result is due to moderate rate chemistry.

Since C/D modeling is fundamentally phenomenological in nature, comparison to data is the only way to decide its validity. The sensitivity of the first and higher moments of the reactant concentration to the molecular mixing model suggests that measured data in a plug flow reactor and simulated data inhomogeneous flow may provide useful data for model validation.

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TABLE I. - VALUES OF  $\gamma$  DEFINED IN EQUATION (10e) FOR DIFFERENT C/D MODELS  
[Initial distribution given by eq. (14).]

Model	$\gamma$	Kurtosis at $\omega t = 1$
Curl	0.5	3.59
Janicka et al.	.4	2.97
Dopazo-O'Brien	0	1.00

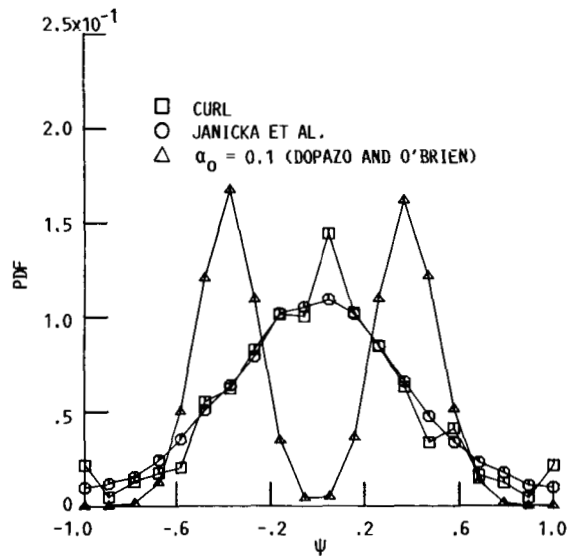


FIGURE 1.- PROBABILITY DENSITY FUNCTIONS VERSUS CONCENTRATION AT  $\omega t = 0.96$ . THE DOPAZO-O'BRIEN [2] MODEL IS APPROXIMATELY REPRESENTED BY  $A(\alpha) = \delta(\alpha - \alpha_0)$ ,  $\alpha_0 = 0.1$ . (BINARY INITIAL DISTRIBUTION).

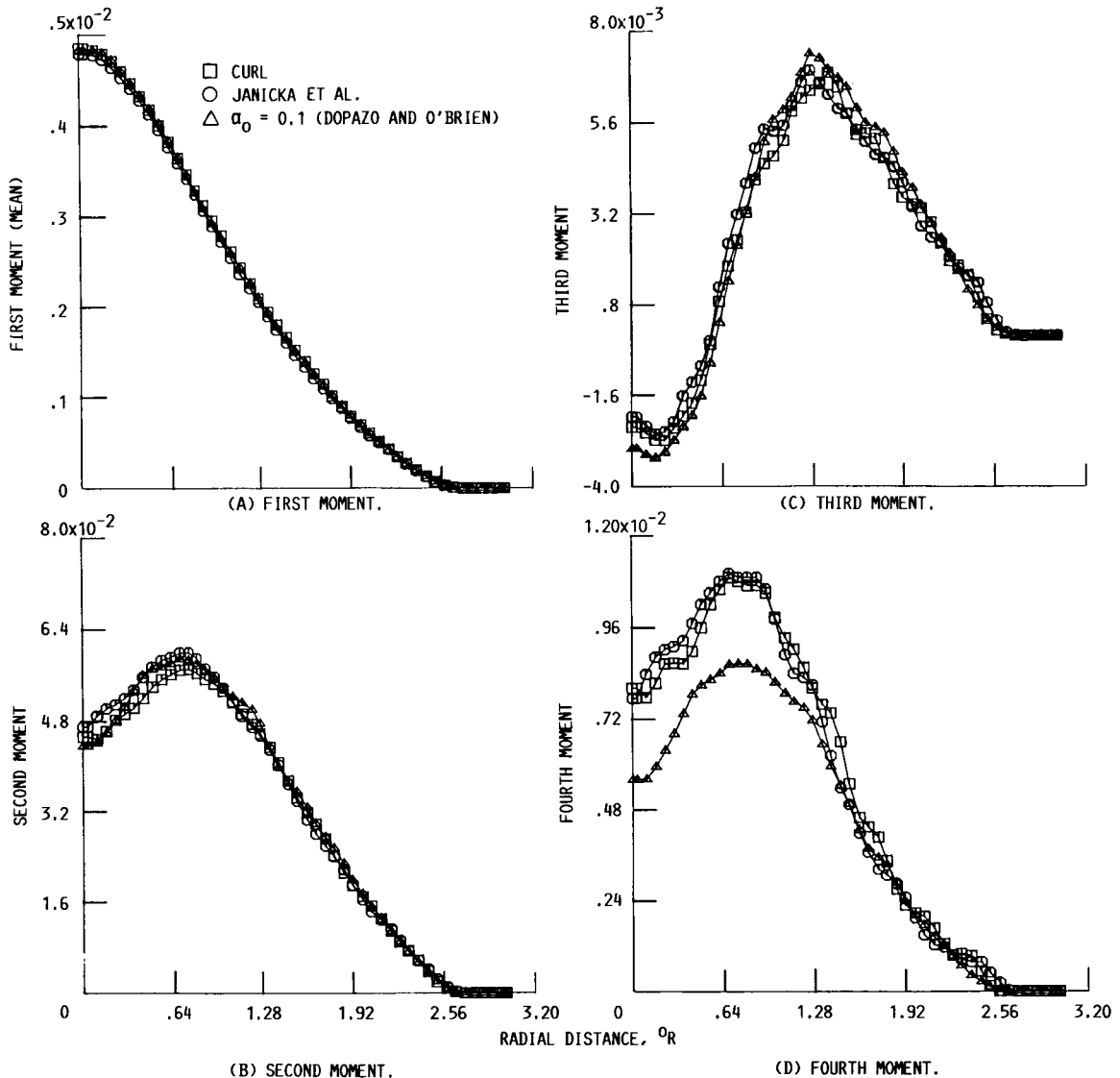
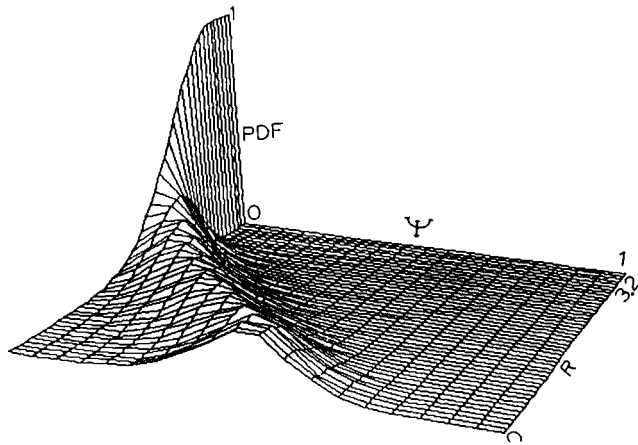
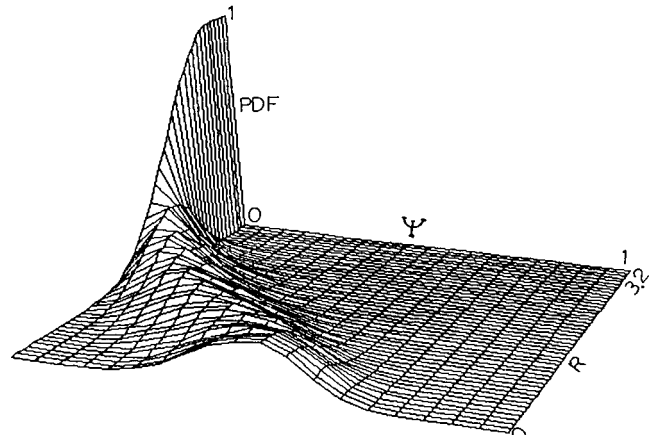


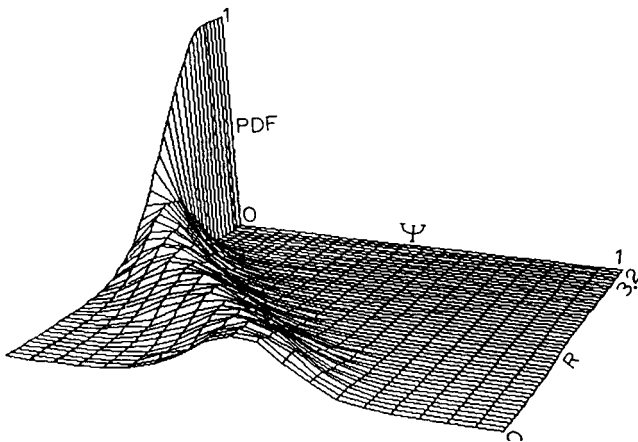
FIGURE 2. - THE FIRST FOUR MOMENTS AGAINST RADIAL DISTANCE  $x = 10D$  DOWNSTREAM FROM THE NOZZLE.



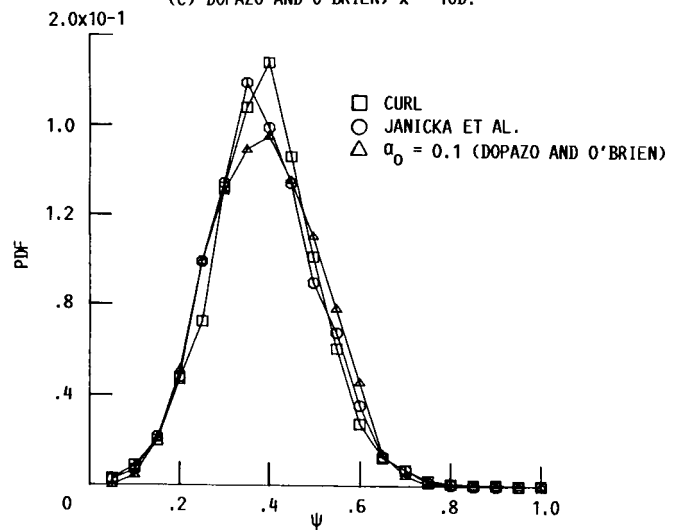
(A) CURL,  $x = 10D$ .



(C) DOPAZO AND O'BRIEN,  $x = 10D$ .



(B) JANICKA ET AL.,  $x = 10D$ .



(D) AT  $R = 0.8$ ,  $x = 10D$ .

FIGURE 3. - PROBABILITY DENSITY FUNCTIONS VERSUS CONCENTRATION AND RADIAL DISTANCE.

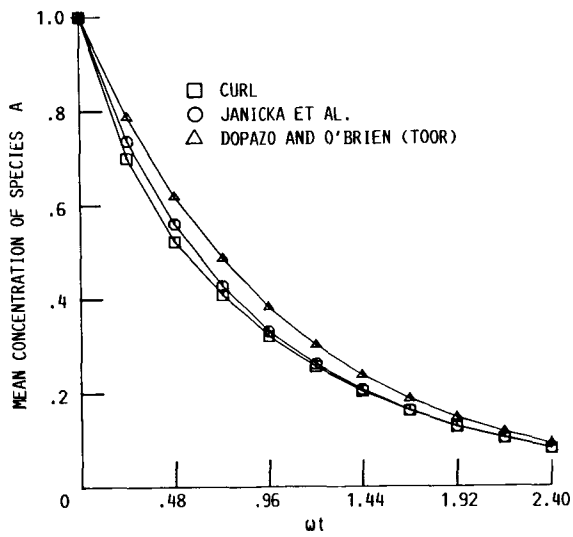


FIGURE 4. -  $\langle A \rangle / \langle A \rangle_0$  VERSUS DIMENSIONLESS TIME IN THE INFINITELY FAST CHEMISTRY LIMIT.

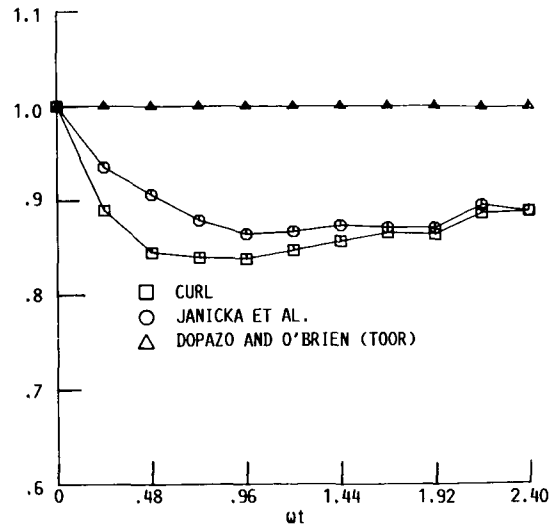


FIGURE 5. - DEVIATION FROM THE DOPAZO-O'BRIEN (TOOR) RESULTS. DATA TAKEN FROM FIGURE 4.

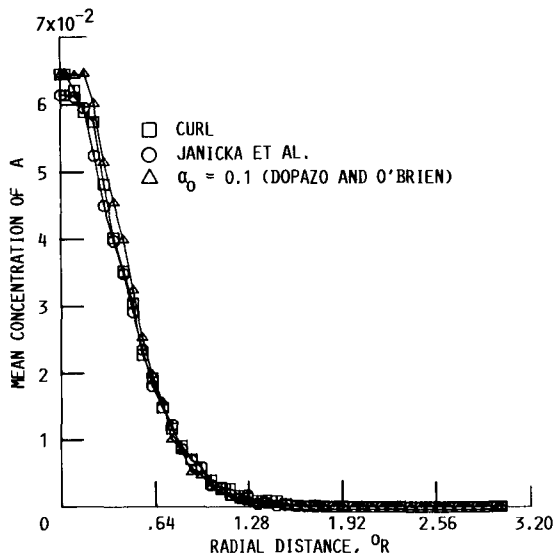


FIGURE 6. - CONCENTRATION OF A VERSUS RADIAL DISTANCE AT  $x = 10D$  DOWNSTREAM FROM THE NOZZLE.

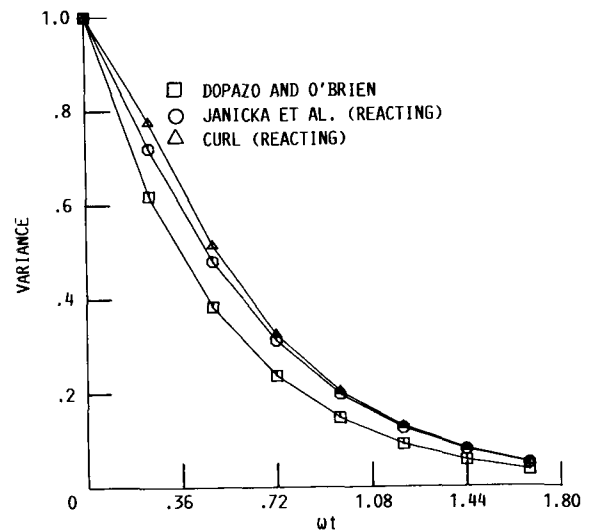


FIGURE 7. - VARIANCE OF A VERSUS DIMENSIONLESS TIME WITH INFINITE RATE CHEMISTRY AND WITH PURE MIXING. IN THE DOPAZO-O'BRIEN APPROXIMATION, THE REACTING AND NONREACTING RESULTS COINCIDE. WITHOUT CHEMISTRY, THE VARIANCE IS IDENTICAL IN THE THREE MODELS.

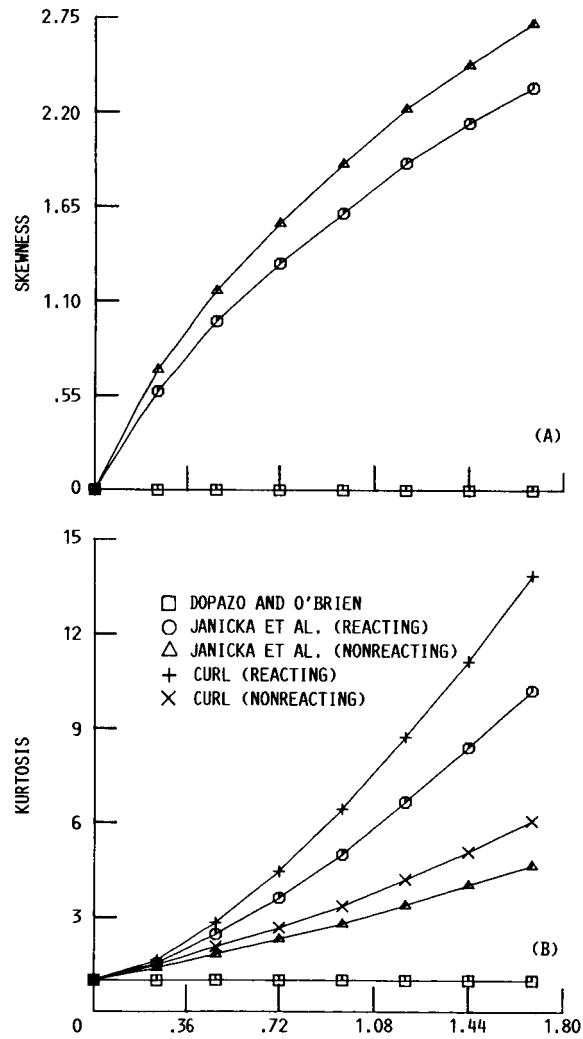


FIGURE 8. - SKEWNESS AND KURTOSIS OF A VERSUS DIMENSIONLESS TIME WITH INFINITE RATE CHEMISTRY AND WITH PURE MIXING. IN THE PURE MIXING CASE THE SKEWNESS IS ZERO.



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16. Abstract <p>The general coalescence-dispersion (C/D) closure provides phenomenological modeling of turbulent molecular mixing. The models of Curl (ref. 1) and Dopazo and O'Brien (ref. 2) appear as two limiting C/D models that "bracket" the range of results one can obtain by various models. This finding is used to investigate the sensitivity of the results to the choice of the model. Inert scalar mixing is found to be less model-sensitive than mixing accompanied by chemical reaction. Infinitely fast chemistry approximation is used to relate the C/D approach to Toor's earlier results (ref. 3). Pure mixing and infinite rate chemistry calculations are compared to study further a recent result of Hsieh and O'Brien (ref. 4) who found that higher concentration moments are not sensitive to chemistry.</p>					
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