Non-Premixed Simple Reaction in Homogeneous Turbulence

In turbulent flow the equations governing the development of conversion vs. distance are not closed because of the contribution of the concentration fluctuations to the average reaction rate. Toor's hypothesis permits closure from data obtained for mixing without reaction. This paper presents a critical review of the derivation of the hypothesis and discusses its validity.

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Introduction

The study of chemical reaction in spatially homogeneous turbulent flow is of practical importance for plug flow type reactors. If variations in the cross-stream direction are negligible in the reactor, time averages taken at a given axial position z ($\langle \ldots \rangle_z$) can be modeled via ensemble averages taken at time t ($\langle \ldots \rangle_t$) in homogeneous turbulence (Bilger, 1980).

The prediction of average concentration vs. time in spatially homogeneous turbulence is an important task. The equations satisfied by the average concentrations are unclosed. If the reaction is of the type $A + nB \rightarrow$ product the unclosed term can be written as

$$\psi^2(t) = \frac{\langle ab \rangle_t}{\langle ab \rangle_o} \tag{1}$$

where a and b are the concentration fluctuations of the reacting species and zero refers to the initial time.

Toor (1969, 1975) characterized turbulent mixing by the ratio of unmixedness, defined as

$$d^2(t) = \frac{\langle a_m^2 \rangle_t}{\langle a_m^2 \rangle_o} \tag{2}$$

and proved that $\psi(t)$ and $d(t)$ are equal in the slow chemistry limit. In Eq. 2 subscript m refers to pure mixing; $\langle a_m^2 \rangle_t$ is the mean square fluctuation of species A in the absence of chemical reaction.

In the infinitely fast chemistry limit Toor (1969, 1975) assumes stoichiometric conditions and equal diffusivities of the two reactants and relates the average concentration of A to $d(t)$ by the simple equation:

$$\frac{\langle A \rangle_t^f}{\langle A \rangle_o} = d(t) \tag{3}$$

The index f indicates infinitely fast reaction. Since in this limit

$$\psi_f^2(t) = \left(\frac{\langle A \rangle_t^f}{\langle A \rangle_o} \right)^2 \tag{4}$$

Toor concludes that

$$\psi_f(t) = d(t) \tag{5}$$

that is, $\psi(t)$ and $d(t)$ are equal in the fast chemistry limit as well. It was hypothesized therefore that

$$\psi(t) = d(t) \tag{6}$$

is valid for arbitrary reaction rates; this is Toor's hypothesis.

If Eq. 3 is valid, then Eq. 6 can be written equivalently as (Toor, 1969, 1975):

$$\psi(t) = \frac{\langle A \rangle_t^f}{\langle A \rangle_o} \tag{7}$$

Toor's hypothesis found general acceptance in the chemical engineering literature (Hill, 1976; Brodkey, 1981). It is presently under investigation whether Toor's hypothesis can be generalized for more complicated cases than second-order reactions (Brodkey and Lewalle, 1985).

If Eqs. 6 and 7 are valid, either of them can be applied to close the equations satisfied by the average concentrations. The present paper investigates the validity of estimating $\psi^2(t)$ via Eq. 6 or Eq. 7.

Our rapid reaction results differ from Eqs. 3 and 5. The results show that Eq. 6 becomes valid in the limit when the reaction rate is much smaller than the rate of mixing. It is concluded that Eqs. 6 and 7 are not equivalent but represent different estimates of $\psi(t)$. While we refer to Eq. 6 as Toor's hypothesis, we emphasize the difference between Eqs. 6 and 7 by referring to Eq. 7 as the fast conversion estimate of $\psi(t)$. This estimate becomes valid in the limit when the reaction rate is much larger than the rate of mixing.

The derivations refer to stoichiometric conditions. It is suggested that further progress requires more detailed plug flow measurements and direct numerical simulation work in homogeneous turbulence.

Background

We consider a second-order, irreversible isothermal reaction of the type $A + nB \longrightarrow$ product, and ignore any dynamic or chemical role the products may play. Assuming Fickian diffusion, the concentrations of the two species obey the following equations:

$$\frac{\partial A}{\partial t} + \underline{u} \cdot \nabla A - D_A \nabla^2 A = -kAB \tag{8a}$$

$$\frac{\partial B}{\partial t} + \underline{u} \cdot \nabla B - D_B \nabla^2 B = -kAB \tag{8b}$$

Here the quantity nA has been replaced by the symbol A (O'Brien, 1971), $\underline{u}(t, \underline{x})$ is the incompressible, turbulent velocity field, $A(t, \underline{x})$ and $B(t, \underline{x})$ are random concentration fields, k is the kinetic reaction rate assumed constant, and D_A and D_B are the respective diffusivities of the two species.

Assume that the turbulent field is spatially homogeneous, the initial spatial distributions of the two species are independent of space, and the two species are initially segregated. The equations satisfied by the average conversions can be written as:

$$\frac{d}{dt}\frac{\langle A\rangle_t}{\langle A\rangle_o} = -k\langle B\rangle_o\left[\frac{\langle A\rangle_t}{\langle A\rangle_o}\frac{\langle B\rangle_t}{\langle B\rangle_o} - \psi^2(t)\right] \tag{9a}$$

$$\frac{d}{dt}\frac{\langle B\rangle_t}{\langle B\rangle_o} = -k\langle A\rangle_o\left[\frac{\langle A\rangle_t}{\langle A\rangle_o}\frac{\langle B\rangle_t}{\langle B\rangle_o} - \psi^2(t)\right] \tag{9b}$$

$\psi^2(t)$ has been defined in Eq. 1. Equations 9a and 9b are unclosed, $\psi^2(t)$ is to be modeled.

Slow and Fast Reaction Limits

Toor's hypothesis resulted from discussion of the extreme cases of infinitely slow and infinitely fast chemistry. For the definition of these limits and the different nondimensional numbers characterizing them, refer to the review article of Bilger (1980).

In the slow chemistry limit Toor (1969, 1975) proves that

$$\psi_s(t) = d(t) \tag{10}$$

where the index s indicates the slow chemistry limit and $d(t)$ was defined in Eq. (2).

In the infinitely fast chemistry case Toor (1962, 1975) assumes equal diffusivities and introduces the conserved scalar

$$J(t, \underline{x}) = A(t, \underline{x}) - B(t, \underline{x}) \tag{11}$$

Let $p_A(c, t)$, $p_B(c, t)$, and $p_J(c, t)$ be the respective probability density functions (PDF's) of $A(t, \underline{x})$, $B(t, \underline{x})$, and $J(t, \underline{x})$. In the fast chemistry limit (O'Brien, 1971).

$$p_A(c, t) = p_J(c, t) + k_A(t)\delta(c) \tag{12a}$$

$$p_B(c, t) = p_J(-c, t) + k_B(t)\delta(c) \tag{12b}$$

Here $c > 0$ and

$$k_A(t) = \int_0^\infty p_J(-c, t)dc = \int_{-\infty}^0 p_J(c, t)dc \tag{13a}$$

$$k_B(t) = \int_0^\infty p_J(c, t)dc = 1 - k_A(t) \tag{13b}$$

From Eq. 12a,

$$\langle A\rangle_t^f = \int_0^\infty c p_J(c, t)dc \tag{14}$$

follows (Toor 1962, 1975).

Let us assume stoichiometric conditions with equal concentrations c_o of the two species in the inlet feeds. These assumptions make $p_J(c, t)$ an even function of c with zero mean (O'Brien, 1971).

Toor (1962) evaluates Eq. 14 with the assumption that $p_J(c, t)$ is Gaussian and obtains

$$\langle A\rangle_t^f = \frac{\langle j^2\rangle_t^{1/2}}{\sqrt{2\pi}} \tag{15}$$

Here $\langle j^2\rangle_t^{1/2}$ is the standard deviation of the conserved scalar J. If the PDF of J is Gaussian both for $t > 0$ and at $t = 0$, then

$$\frac{\langle A\rangle_t^f}{\langle A\rangle_o} = \frac{\langle j^2\rangle_t^{1/2}}{\langle j^2\rangle_o^{1/2}} = d(t) \tag{16}$$

follows from Eq. 15.

Using Eq. 16 together with Eq. 4, Toor (1969, 1975) finds that

$$\psi_f^2(t) = d^2(t) \tag{17}$$

Equations 16 and 17 are identical to Eqs. 3 and 5 given in the Introduction.

While it is generally expected (O'Brien, 1980) that the PDF of a conserved scalar relaxes asymptotically to Gaussian form, for short times the PDF is not Gaussian, but rather of bimodal shape determined by the initial conditions (Miyawaki et al., 1974). The use of the Gaussian assumption at $t = 0$ is not justified.

That $p_J(c, t)$ is bimodal for short times can be shown mathematically and understood physically. Since we are considering stoichiometric conditions with equal concentrations c_o of the reactants in the inlet feeds, the initial PDF's of the reactants become

$$p_A(c, 0) = p_B(c, 0) = \frac{1}{2}\delta(c - c_o) + \frac{1}{2}\delta(c) \tag{18}$$

which via Eqs. 12a and 12b, in turn results in

$$p_J(c,0) = \frac{1}{2}\delta(c - c_o) + \frac{1}{2}\delta(c + c_o) \tag{19}$$

In physical terms Eq. 19 says that $J = c_o$ in the inlet feed containing A and $J = -c_o$ in the inlet feed containing B.

Since $p_J(c, t)$ becomes Gaussian only for long times, Eq. 15 is an asymptotic result that must not be applied at $t = 0$. We divide it by $\langle A \rangle_o$ and use Eq. 2 to obtain

$$\frac{\langle A \rangle_t^f}{\langle A \rangle_o} \approx \left(\frac{\langle j^2 \rangle_o^{1/2}}{\langle A \rangle_o \sqrt{2\pi}} \right) d(t) \tag{20}$$

From Eqs. 18 and 19,

$$\langle A \rangle_o = \frac{c_o}{2}, \quad \langle j^2 \rangle_o^{1/2} = c_o \tag{21}$$

follows. Upon insertion into Eq. 20, the average conversion can be written asymptotically as

$$\frac{\langle A \rangle_t^f}{\langle A \rangle_o} \approx \sqrt{\frac{2}{\pi}}\, d(t) \tag{22}$$

From Eqs. 4 and 22,

$$\psi_f^2(t) \approx \frac{2}{\pi} d^2(t) \tag{23}$$

follows.

Equations 22 and 23 differ from Eqs. 3 and 5. Indeed, Eqs. 3 and 5 do not apply in experimental cases when $\langle A \rangle_o$ is evaluated from the concentration and flow rate values at the reactor inlet. The majority of the experimental investigations belong to this category.

Equations 22 and 23 have been derived using the assumption that the PDF of a conserved scalar becomes asymptotically Gaussian. While this assumption is corroborated by some experimental evidence (Miyawaki et al., 1974; Tavoularis and Corrsin, 1981), the statement has never been proved formally. Since asymptotically Gaussian behavior is a likely appropriate assumption, in the rest of this paper Eqs. 22 and 23 will be employed.

Short Time Behavior

The quantities $\psi^2(t)$ and $d^2(t)$ have been defined in Eqs. 1 and 2. The definitions show that

$$\lim_{t \to 0} \frac{\psi^2(t)}{d^2(t)} = 1 \tag{24}$$

The short time behavior of $\psi(t)$ and $d(t)$ is investigated further in the Appendix. It is shown there that

$$\lim_{t \to 0} \frac{d}{dt} \frac{\psi^2(t)}{d^2(t)} = 0 \tag{25}$$

Equations 24 and 25 are valid independently of the value of the reaction speed.

Chemical Reactions of Arbitrary Rate

According to Eqs. 5 and 10, $\psi(t)$ is equal to $d(t)$ in the limiting cases of fast and slow chemistry. This led Toor (1969, 1975) to hypothesize Eq. 6, which says that $\psi(t)$ and $d(t)$ are equal independently of the reaction speed. Since we found that Eq. 5 should be replaced by Eq. 23, it is necessary to reinvestigate the nature of the relationship between $\psi(t)$ and $d(t)$.

Figure 1 shows $\psi^2(t)/d^2(t)$ vs. time. The two solid curves represent the infinitely slow and fast chemistry limits. The slow chemistry behavior is given in Eq. 10. The information on the fast chemistry limit is given by Eqs. 23, 24, and 25. Accordingly, the fast chemistry curve ψ_f^2/d^2 starts out at unity with zero derivative and tends to the $2/\pi = 0.64$ asymptote for long times.

The fast chemistry curve in Figure 1 is to be viewed with caution; since it is based on asymptotic information only, its behavior for intermediate times cannot be predicted theoretically. Further experimental and theoretical investigations are needed to clarify this behavior.

In spite of this qualitative nature of the fast chemistry curve, Figure 1 helps to replace Toor's hypothesis with a new one. Infi-

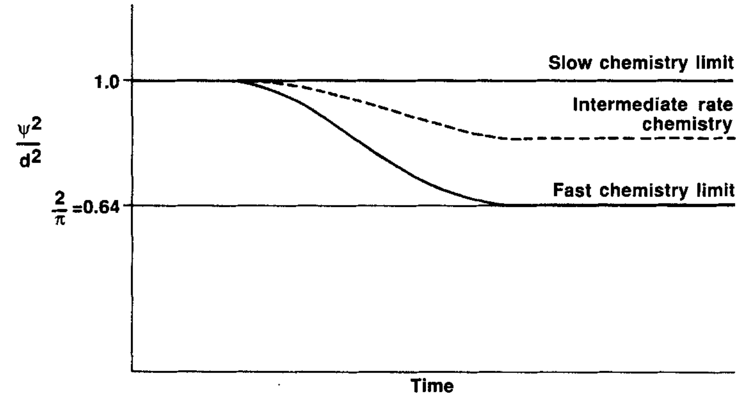

Figure 1. ψ^2/d^2 vs. time

The figure shows that Eq. 6 becomes valid in the slow chemistry limit. The fast chemistry curve is based on asymptotic information. The intermediate rate chemistry curve illustrates the hypothesis that any finite rate chemistry curve will lie between the limiting curves.

nitely slow and fast chemistry are limiting cases. It is plausible to hypothesize that any finite rate chemistry curve will lie between the two limiting curves. The dashed line in the figure serves to illustrate the new hypothesis pictorially.

The recent numerical simulations of Leonard and Hill (1986) and J. C. Hill (private communication, 1987) support Figure 1. Some further support comes from the early experiments of Mao and Toor (1971), who measured $\psi^2(t)$ for intermediate rate chemistry and compared it to a measured $\psi_f^2(z)$ curve. As expected from Figure 1, the measured $\psi^2(t)$ values were consistently above the $\psi_f^2(z)$ curve and the difference increased with decreasing Damköhler number. This finding supports Figure 1 but a more detailed study of the experimental conditions and results is desirable.

The present philosophy differs from Toor's original argument (1969, 1975) but the quantitative difference is not necessarily large. In the asymptotic region $\psi^2(t)$ differs from $d^2(t)$ at most by a factor of 0.64; this means that the $\psi^2(t)/d^2(t)$ ratio may be much closer to unity in actual finite rate cases. The difference between $\psi^2(t)$ and $d^2(t)$ is influenced also by the details of the behavior of the limiting fast chemistry curve for intermediate times.

However, let us point out an inherently unfavorable tendency in estimating $\psi^2(t)$ by $d^2(t)$ via Eq. 6. The aim of the procedure is to close Eqs. 9a and 9b. Figure 1 indicates that the estimate is the more valid as the chemistry is slower. In the slow chemistry limit, however, $\psi^2(t)$ becomes negligible in Eqs. 9a and 9b. This means that in the limit when Eq. 6 becomes strictly valid, the equations become closed; closure becomes unnecessary. As the unclosed term becomes more important in the equation, the estimate deteriorates.

We turn now to discussion of Eq. 7. The use of this equation for closure was suggested by Toor (1969, 1975) as an equivalent and experimentally more convenient version of Eq. 6. The equivalence of Eqs. 6 and 7 was based on Eq. 3. The present work replaces Eq. 3 by Eq. 22. It turns out, therefore, that Eq. 7 is a separate statement that does not follow from Toor's hypothesis. Since Eq. 7 suggests estimating $\psi^2(t)$ from fast conversion data, we refer to it as the fast conversion estimate. Let us investigate its validity in the various limiting cases.

Since Eq. 7 can be rewritten via Eq. 4 as

$$\psi(t) = \psi_f(t) \tag{26}$$

it follows that Eq. 7 becomes strictly valid in the infinitely fast chemistry limit.

$\psi_s(t) = d(t)$ from Eq. 10. We use Eqs. 4, 24, 25, and 23 to evaluate the validity of the fast conversion estimate in the slow chemistry limit. It turns out that for short times $\psi_f^2/\psi_s^2 \longrightarrow 1$ and $d/dt(\psi_f^2/\psi_s^2) \longrightarrow 0$, whereas for long times ψ_f^2 is lower than ψ_s^2 by a factor of $2/\pi$. The behavior for intermediate times cannot be determined mathematically.

It seems plausible to hypothesize that the error in using Eq. 7 in any finite rate chemistry case will lie between its fast and slow chemistry limits.

The ψ^2 term is most important in Eqs. 9a and 9b in the fast chemistry limit. As mentioned before, it becomes negligible in the slow chemistry limit. Since Eq. 7 is valid for fast chemistry and deteriorates for slow chemistry, its application for closure involves an inherently favorable tendency.

McKelvey et al. (1975) measure conversion vs. distance in intermediate and slow rate chemistry cases. They evaluate ψ^2 from rapid reaction data using Eq. 7 and solve Eqs. 9a and 9b numerically. The predictions show excellent agreement with the intermediate and slow rate chemistry measurements.

Let us point out that this finding does not corroborate Toor's hypothesis but supports rather the application of the fast conversion estimate for closure. One speculates that the agreement between data and prediction is related to the advantageous tendency involved in the application of Eq. 7. Since closure via Eq. 6 involves the opposite tendency, it is questionable whether it would lead to equally satisfactory results. Experimental and numerical investigations are needed to clarify this problem.

Conclusions

In turbulent flow Eqs. 9a and 9b are unclosed due to the appearance of a second-order fluctuation term. Toor (1969, 1975) suggested closing the equations either by using pure mixing experiments, Eq. 6, or by using fast chemistry conversion data, Eq. 7.

While in the original discussion the two procedures were presented as equivalent, we conclude that they are different. Equation 6 becomes valid in the slow chemistry limit, Eq. 7 in the opposite limit of fast chemistry. New hypotheses regarding the use of Eqs. 6 and 7, for intermediate rate chemistry, are illustrated in Figure 1 and discussed after Eq. 26.

It is concluded that the measurements of McKelvey et al. (1975) support the application of Eq. 7 for closure. Whether the use of Eq. 6 would result in equally satisfying results is an open question.

The deviation from Toor's original results is due to the difference between the present fast chemistry calculations and Toor's corresponding results (1969, 1975). Equations 23, 24 and 25 provide only asymptotic information on the ψ_f^2/d^2 vs. time curve. The behavior for intermediate times is not known, however. This information, together with examination of intermediate chemistry cases, is essential for utilizing the idea of closure via Eq. 6 or Eq. 7 in problems of engineering importance.

It is concluded that detailed measurements in plug flow reactors and numerical simulations in homogeneous flow are essential for futher progress. The recent numerical work of Leonard and Hill (1986, 1987) is an important step in this direction.

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Notation

A, B = instantaneous concentration of reacting species
a, b = instantaneous concentration fluctuation of reacting species
c = concentration variable
$d^2(t) = \langle a_m^2 \rangle_t / \langle a_m^2 \rangle_o$
D = coefficient of molecular diffusivity
$J = A - B$
j = fluctuation of J
k = kinetic reaction rate
$p(c, t)$ = concentration PDF at time t
$\underline{u}$ = turbulent velocity
t = time

z = axial distance from reactor inlet
$\underline{x}$ = spatial coordinate
$\psi^2(\bar{t}) = \langle ab \rangle_t / \langle ab \rangle_o$

Indices

f = fast chemistry limit
o = initial
m = mixing in absence of chemical reaction

Other notation

$\langle \ldots \rangle_t$ = ensemble average at t
$\langle \ldots \rangle_z$ = time average at z

Appendix

Let us investigate the short time behavior of $\psi(t)$ and $d(t)$. To this end, we introduce the random concentration fields $A_m(t, \underline{x})$ and $B_m(t, \underline{x})$, where subscript m indicates the absence of reaction between the species. Since the concentration fields at an ideal inlet cannot be affected by the reaction (Toor, 1975), it is correct to write

$$A(0, \underline{x}) = A_m(0, \underline{x}), \quad B(0, \underline{x}) = B_m(0, \underline{x}) \qquad \text{(A1)}$$

The concentration fields A_m and B_m satisfy Eqs. 8a and 8b with the chemical source term set equal to zero. Due to initial segregation, this term is zero at $t = 0$ in the reacting case as well. It follows from Eqs. 8a, 8b, and A1 that

$$\left.\frac{\partial A}{\partial t}\right)_{t=0} = \left.\frac{\partial A_m}{\partial t}\right)_{t=0}, \quad \left.\frac{\partial B}{\partial t}\right)_{t=0} = \left.\frac{\partial B_m}{\partial t}\right)_{t=0} \qquad \text{(A2)}$$

therefore

$$A(t, \underline{x}) = A_m(t, \underline{x}) + 0(t^2), \quad B(t, \underline{x}) = B_m(t, \underline{x}) + 0(t^2) \qquad \text{(A3)}$$

and

$$a(t, \underline{x}) = a_m(t, \underline{x}) + 0(t^2), \quad b(t, \underline{x}) = b_m(t, \underline{x}) + 0(t^2) \qquad \text{(A4)}$$

In deriving Eqs. A3 and A4 only initial segregation had to be assumed. Considering again spatially homogeneous, incompressible flow and space-independent initial concentration distributions, we may write:

$$\langle ab \rangle_t = \langle a_m b_m \rangle_t + 0(t^2) \qquad \text{(A5)}$$

We assume furthermore that the two species have equal diffusivities and are present in equal concentration in the inlet feeds, and refer to a result of Toor (1969, 1975), who proved that

$$a_m(t, \underline{x}) = -b_m(t, \underline{x}) \qquad \text{(A6)}$$

Hence

$$\langle ab \rangle_t = -\langle a_m^2 \rangle_t + 0(t^2)$$

that is,

$$\psi^2(t) = \frac{\langle a_m^2 \rangle_t}{\langle a_m^2 \rangle_o} + 0(t^2) \qquad \text{(A7)}$$

The first term on the righthand side of Eq. A7 is identical to the righthand side of Eq. 2. We conclude that

$$\psi^2(t) = d^2(t) + 0(t^2) \qquad \text{(A8)}$$

therefore,

$$\lim_{t \to 0} \frac{d}{dt} \frac{\psi^2(t)}{d^2(t)} = 0 \qquad \text{(A9)}$$

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