



THE INFLUENCE OF VISCOSITY ON MIXING IN JET REACTORS

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Abstract—A free turbulent jet discharging into a tank is a simple device and can develop local turbulent energy dissipation rates on the order of 10^3 – 10^4 W kg⁻¹, which are suitable for fast reactions and drop dispersion. Parameter estimation for an earlier model of micromixing and mesomixing has been updated to include further information on turbulence in the initial developing region of the jet. Measured product distributions for fast chemical reactions, employing various initial jet velocities, viscosities and feed times for semi-batch operation, showed two limiting regimes where either micromixing or mesomixing was controlling. The updated model described these measurements satisfactorily, as well as some earlier drop breakup experiments.

INTRODUCTION

A simple circular jet discharging into a tank, which is operated as a semi-batch reactor, has several attractive features, e.g., inexpensive, easy cleaning and high local rates of turbulent energy dissipation. Values of order 10^3 – 10^4 W kg⁻¹ have been found by Baldyga *et al.* (1994), which are suitable for attaining small drop sizes in dispersions and high selectivities in some fast, mixing-controlled reactions. A model of mesomixing and micro-mixing in such a jet reactor has been developed (Baldyga *et al.*, 1994), which necessitated several approximations about the incompletely characterised initial jet region. Depending upon the criteria chosen, this extends from the outlet of the nozzle (diameter = d ; axial position $x = 0$) to roughly $x = 8d$. Its outer edge is a shear layer whose turbulence spreads radially outwards with the moving jet, but also inwards destroying the initial potential core until the fluid on the jet axis is also fully turbulent. Quantitative information, including turbulence parameters, in this initial region is however incomplete and not fully consistent.

The present contribution aims

(a) to improve the estimation of model parameters, especially for the initial developing region of the jet, without however changing the structure of the earlier model (Baldyga *et al.*, 1994), and

(b) to compare the updated model with some of the earlier experimental results, but also with new ones whereby the viscosity is increased by a factor of around seven, whilst maintaining turbulent flow.

MODEL PARAMETERS

Energy dissipation rate model

The rate of fine-scale engulfment, which is commonly the limiting mechanism in micromixing, de-

pends upon the kinematic viscosity (ν) and the rate of turbulent energy dissipation per unit mass (ϵ). Jet turbulence is highly inhomogeneous and the distribution of its properties is two-dimensional, although almost nothing is known about the radial dependence of ϵ in the initial region. The axial dependence was shown in Fig. 2 of Baldyga *et al.* (1994) and will also be accepted here. In the fully turbulent region, measurements where x exceeds roughly $8d$ have been correlated by eq. (1),

$$\epsilon = Cd^3 u_0^3 / x^4 \quad (1)$$

C was previously determined by fitting to be 110 (Baldyga *et al.*, 1994). Experimentally determined values of C are however around half this value, e.g., 48 or somewhat higher (Friehe *et al.*, 1972), 48 (Antonia *et al.*, 1980) and 39 (Panchapakesan and Lumley, 1993) and it is more realistic to set $C = 50$. Description of dissipation rates should also be improved by including a virtual origin, so that a revised eq. (1), namely eq. (2), will be employed here.

$$\epsilon = 50d^3 u_0^3 / (x - 2d)^4 \quad (2)$$

Flow field

The field used earlier in the initial region is plausible (Boguslawski and Popiel, 1979; Hill, 1972). However, the critical Reynolds number (du_0/ν) for full development of the vortex pairs, which form in the initial region and contribute to macro-scale engulfment, is 5000–8000 (Liepmann and Gharib, 1992). The value given before (Baldyga *et al.*, 1994), namely 10^3 , is too low.

Time constant for mesomixing, t_m

Mesomixing refers to turbulent mass exchange between the jet and its surroundings, which is inertial-convective in character. Starting from eq. (32) of the earlier publication and Corrsin's original eq. (70)

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(Corrsin, 1964), its time constant is

$$t_m = 2.04L_c^{2/3}/\varepsilon^{1/3} \quad (3)$$

where, using Corrsin's notation, L_c is the integral scale of the concentration fluctuations, when $Sc \gg 1$. The time constant t_m characterises the rate of reduction of concentration variance in the inertial-convective sub-range. From Corrsin's eqs (20a) and (27) the energy dissipation rate may be eliminated from eq. (3) to give

$$t_m = 0.098(\lambda^2/\nu)(L_c/L)^{2/3} \quad (4)$$

where, λ and L are, respectively, the Taylor microscale and the integral scale of the turbulence. The Taylor microscale grows linearly along a jet, a suitable correlation being (Friehe *et al.*, 1972)

$$\lambda = 0.88Re^{-0.5}x. \quad (5)$$

The constant 0.88 is quite possibly too small (Antonia *et al.*, 1980), a value of 1.07 being also consistent with some experiments (Friehe *et al.*, 1972). Moreover, a virtual origin probably improves the correlation of λ for short distances from the nozzle [Fig. 3 of Antonia *et al.*, 1980]. Equation (5) will be modified here to eq. (6)

$$\lambda = c_1(x+a)Re^{-0.5} \quad (6)$$

where c_1 is of order unity and a is likely to be in the range of d to $2d$ (Antonia *et al.*, 1980). Substituting into eq. (4) then gives

$$t_m = 0.098 [c_1(x+a)]^2 (\nu u_0)^{-1} (L_c/L)^{2/3}. \quad (7)$$

As reported earlier the reagent feed stream, denoted in Baldyga *et al.* (1994) by B and shown there in Fig. 5, was quickly dispersed across the jet, suggesting that $L_c \approx L$. The time constant t_m will therefore be evaluated here from eq. (8) where c_2 should be close to 0.1.

$$t_m = c_2(x+a)^2 (\nu u_0)^{-1}. \quad (8)$$

EXPERIMENTAL RESULTS

The apparatus, the chemical reactions and the experimental method have already been described (Baldyga *et al.*, 1994). In some runs, the viscosity of both the jet and bulk solutions was increased to 6.2 mPa s at 298 K by the addition of 0.5 wt% hydroxyethyl cellulose (Natrosol-GR) (Gholap *et al.*, 1994). Semi-batch operation was employed whereby the B-rich solution forming the jet was added during a feed time (t_f) to the bulk solution, containing the reagents A and AA, within the tank. With nozzle velocities (u_0) in the range 4 to 10 m s^{-1} , the jet Reynolds number varied from 5200 to 90,000. The length of straight pipe (i.d. 0.008 m) terminating in the nozzle ($d = 0.008$ m) was 0.64 m, so that flow through the nozzle was fully developed. The internal and external diameters of the B-feed pipe were 0.001 and 0.0018 m. Feed times were so long that the momentum of the feed was negligible compared to that of the jet: due to lack of information a possible enhancement of the jet's turbulence caused by the feed pipe was neglected.

The experimental conditions used may be summarized as

$$\begin{aligned} V_A &= 0.037 \text{ m}^3, V_B = 2.11 \times 10^{-4} \text{ m}^3, \\ C_{A0} = C_{AA0} &= 0.4113 \text{ mol m}^{-3}, N_{A0}/N_{B0} = 1.2, \\ \xi &= 1, \alpha = 175, C_{B0} = 60 \text{ mol m}^{-3}, \\ w &= 1.75d = 0.014 \text{ m}, t_f = 42\text{--}4280 \text{ s}, \\ \nu &= 8.9 \times 10^{-7} \text{ m}^2 \text{ s}^{-1} \text{ and } 6.2 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}, \\ u_0 &= 4, 6 \text{ and } 10 \text{ m s}^{-1}. \end{aligned}$$

The simultaneous diazo coupling of 1-naphthol(A) and 2-naphthol(AA) with diazotized sulfanilic acid(B) has a product distribution(X_Q) which is sensitive to micromixing at turbulent energy dissipation rates up to 10^5 W kg^{-1} (Bourne *et al.*, 1992). X_Q is the yield of the dye formed from 2-naphthol (Baldyga *et al.*, 1994; Bourne *et al.*, 1992).

Figures 1–3 show the variation of X_Q with the feed time (t_f) in aqueous and viscous solutions when the jet velocities (u_0) were 4, 6 and 10 m s^{-1} , respectively. With longer feed times, the product distribution

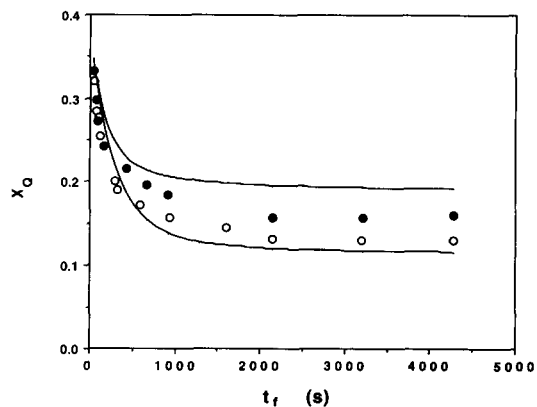


Fig. 1. Product distribution as a function of feed time for nozzle velocity 4 m s^{-1} : (○) aqueous; (●) viscous; (—) model.

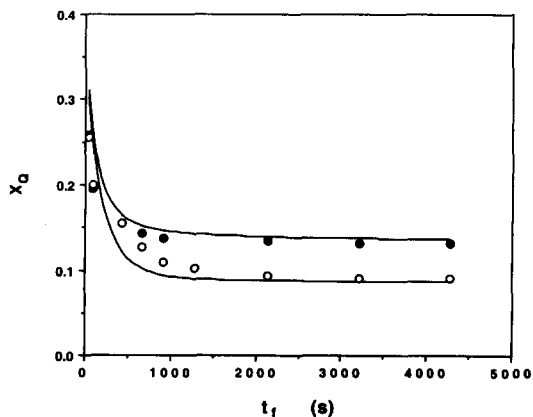


Fig. 2. Product distribution as a function of feed time for nozzle velocity 6 m s^{-1} : (○) aqueous; (●) viscous; (—) model.

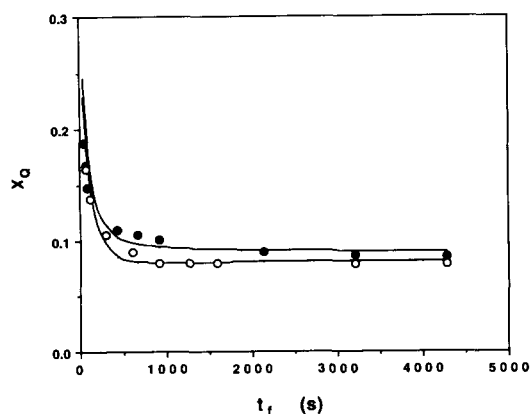


Fig. 3. Product distribution as a function of feed time for nozzle velocity 10 m s^{-1} : (○) aqueous; (●) viscous; (—) model.

reaches an asymptotic value, which decreases with increasing power input to the jet. Even when $u_0 = 10 \text{ m s}^{-1}$, this asymptote (0.078 aqueous and 0.086 viscous) exceeds the yield in the chemical or kinetically controlled regime [0.019 Bourne *et al.*, (1992)]. The viscosity also influences X_Q in fully turbulent flow. It may be concluded that the product distribution of these fast reactions is determined by micromixing as the sole mixing mechanism, when feed times are long.

Figures 1–3 also show that for shorter feed times X_Q progressively exceeds its asymptotes as the feed rate increases, whilst all other quantities are held constant. Similar results for aqueous solutions were given earlier [Fig. 11 of Baldyga *et al.* (1994)]. It can now be seen that X_Q becomes increasingly less dependent on viscosity as the feed time decreases. For example, when $u_0 = 6 \text{ m s}^{-1}$, using $t_f = 93 \text{ s}$, X_Q was 0.200 in aqueous solution and 0.195 in viscous solution. Reducing t_f to 42 s gave $X_Q = 0.255$ (aqueous) and 0.259 (viscous). The analytical error in measuring X_Q is ± 0.003 (Bourne *et al.*, 1992). At the highest feed rates represented in Figs 1–3, viscosity has no significant effect on product distribution. Equation (41) of Baldyga *et al.* (1994) gave a bulk blending time of 9 s under the least favourable conditions ($u_0 = 4 \text{ m s}^{-1}$) and 6 s when $u_0 = 6 \text{ m s}^{-1}$. Comparing these macro-mixing times with t_f , it is most likely that the mixing and reacting jet fluid was engulfing fluid of uniform composition from the bulk of the tank. It may be concluded that the product distribution at short feed times depended essentially on mesomixing which signifies transverse turbulent exchange between the jet and its surroundings as well as inertial-convective disintegration of large eddies.

COMPARISON WITH MODEL

As was shown in the last section, the measured product distributions in Figs 1–3 cover both the micromixing- and mesomixing-controlled regimes and

should therefore offer a good test of the model and its parameters. With reference to the time constant for mesomixing, t_m , the values of c_2 and a in eq. (8) are expected to be close to 0.1 and d , respectively. The curves in Figs 1–3 were computed from the model using $c_2 = 0.12$ and $a = d$. The agreement with the measurements is satisfactory, particularly with increasing jet velocity. As noted earlier, the jet Reynolds number ranged from 5200 to 90,000 and with reference to Fig. 1 ($u_0 = 4 \text{ m s}^{-1}$) it is possible that turbulence was not fully developed particularly in the viscous solution.

Once the turbulence in a device has been characterised, it should be possible to predict the maximum stable drop size when a fluid, having known physical properties, is dispersed. Figure 12 of Baldyga *et al.* (1994) illustrated this for the dispersion of a silicone oil in the jet used for the fast chemical reactions. The present study in the micromixing-controlled regime (long feed times) supports the energy dissipation profile given in eq. (2). Peak values of the dissipation rate occur when $x = 7d$ (Baldyga *et al.*, 1994) and are 640 W kg^{-1} ($u_0 = 4 \text{ m s}^{-1}$), 2160 W kg^{-1} ($u_0 = 6 \text{ m s}^{-1}$), 5120 W kg^{-1} ($u_0 = 8 \text{ m s}^{-1}$) and $10,000 \text{ W kg}^{-1}$ ($u_0 = 10 \text{ m s}^{-1}$). The maximum stable drop sizes, measured earlier, corresponded at these four jet velocities to 896, 3084, 7401 and $13,530 \text{ W kg}^{-1}$ (Baldyga *et al.*, 1994). Equation (2) is superior to the earlier ε -profile with respect to drop breakup in the jet. Radial variations of ε could explain (Baldyga *et al.*, 1994) why values obtained from drop dispersion exceed the radially averaged dissipation rates which are consistent with product distributions.

CONCLUSIONS

The model structure employed earlier to describe micromixing and mesomixing in a turbulent jet (Baldyga *et al.*, 1994) has been retained, but parameter estimation was reviewed, especially for the initial region of rapidly developing flow. Virtual origins were introduced into the axial profiles of the energy dissipation rate and the time constant for mesomixing: their constants (C , c_2 and a) were estimated from approximately known properties of turbulent jets. Radial profiles of the dissipation rates in the initial region cannot be specified in the present state of knowledge: more information would allow modelling reactive mixing and drop breakup to be refined.

Measured product distributions for a set of fast chemical reactions, conducted at room temperature in aqueous (0.89 mPa s) and viscous (6.2 mPa s) solutions using semi-batch operation, allowed two mixing regimes to be identified. With long feed times, micromixing controlled X_Q , whereas with short feed times, mesomixing was relevant. Both mechanisms were important with intermediate feed times.

The updated jet reactor model described well the product distributions when operating with various feed times, jet velocities and viscosities. The peak energy dissipation rates agreed quite well with values deduced earlier from drop breakup measurements

(Baldyga *et al.*, 1994). The updated model is thought to be more accurate than the earlier version.

NOTATION

a	virtual origin of jet, m
A	reagent 1-naphthol
AA	reagent 2-naphthol
B	reagent diazotized sulfanilic acid
c_1, c_2, C	dimensionless constants, dimensionless
C_{i0}	initial concentration of reagent i , mol m ⁻³
d	nozzle diameter, m
L	integral scale of velocity fluctuations, m
L_c	integral scale of concentration fluctuations, m
N_{i0}	initial quantity of reagent i , mol
Re	Reynolds number, dimensionless
Sc	Schmidt number, dimensionless
t_f	feed time, s
t_m	time constant for mesomixing, s
u_0	velocity of jet at nozzle exit, m s ⁻¹
V_A	volume of A and AA-rich solution in reactor, m ³
V_B	volume of B-rich solution added to reactor, m ³
w	axial distance between nozzle and B-feed tube, m
x	axial distance from the nozzle, m
X_Q	yield of dye formed from AA, dimensionless
<i>Greek letters</i>	
α	volume ratio V_A/V_B , dimensionless
ε	rate of dissipation of turbulent kinetic energy per unit mass, W kg ⁻¹

λ	Taylor microscale, m
ν	kinematic viscosity, m ² s ⁻¹
ξ	stoichiometric ratio N_{AA0}/N_{A0} , dimensionless

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