Mesomixing in Semi-Batch Reaction Crystallization and Influence of Reactor Size

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Experiments on semibatch reaction crystallization of benzoic acid are reported, in which hydrochloric acid was fed into an agitated solution of sodium benzoate. The influence of mixing and the influence of reactor size are examined on the product crystal mean size. The product mean size increases with increasing stirring rate and with decreasing feed rate. At low feed rates, the mean size increases at decreasing feed pipe diameter. At high feed rates the influence of the feed pipe diameter is more complex. Micromixing is of some importance in most experiments, but the rate of mesomixing especially governs the process. Mesomixing seems to be adequately described by the inertial-convective disintegration mechanism. In many aspects experimental results cannot be described by the turbulent-dispersion mechanism. The product mean size does not exhibit a clear dependence on reactor size, but depends more strongly on other parameters. Results from experiments from 1 L scale to 200 L scale can be correlated fairly well against a dimensionless number defined as the ratio of the total time of reactant feeding to the time constant of mixing. The best representation of the mixing time constant is obtained by making it directly proportional to the ratio of the feed pipe diameter and the linear velocity of the bulk flow passing the feed pipe. The proportionality constant can be calculated from turbulence data over the bulk flow at the feed point. © 2004 American Institute of Chemical Engineers *AIChE J,* 50: 3107–3119, 2004

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Introduction

Reaction crystallization is commonly used in the production of pharmaceuticals and organic fine chemicals. The usual procedure is to feed one reactant solution into an agitated solution of the other reactant in a semi-batch, also called fed-batch, operation. The chemical reaction is often fast and the solubility of the product compound is low or very low compared to reactant concentrations. Hence, the supersaturation at the feed point becomes high which results in rapid nucleation and fast crystal growth. The process proceeds under conditions of partial segregation, and the product size distribution becomes significantly influenced by the mixing conditions. Results published on the influence of different process variables in semi-batch reaction crystallization are reviewed by Torbacke and Rasmuson (2001).

The hydrodynamic situation in a turbulently stirred tank is complex, and varies considerably from location to location. The mixing is due to the transport of the energy provided by the agitator through a cascade of vortices to the smallest scales where the energy is dissipated as heat. Mixing takes place on all levels from the largest scales of the flow to the molecular scale, but, in particular, three levels are distinguished: macromixing, mesomixing, and micromixing. Macromixing describes mixing at the scale of the tank; that is, how fluid lumps are distributed over the whole tank volume. Micromixing includes mixing in the smallest vortices and the ultimate molecular diffusion. Mesomixing is defined as the mixing interme-

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diate between micromixing and macromixing, and mesomixing is of particular interest when reactant feed pipes are used, in which case it accounts for the early mixing of the feed at scales comparable to the feed pipe diameter.

In industry, there are three common scale-up procedures for agitated tank processes (Tatterson, 1994; Uhl and von Essen, 1987). These are (1) constant power input per unit volume, (2) constant impeller rotational speed, and (3) constant torque (power divided by speed) per unit volume, which is the same as constant impeller tip speed. Constant power per unit volume is assumed to keep the rate of micromixing constant, the rate of macromixing is inversely proportional to the agitation rate, and scale-up at constant tip speed is used when simple blending or maximum shear rate is to be kept constant (Leng, 1991). However, scale-up is complicated since it is impossible to keep all conditions equal in different scales of processing, and sometimes the process is controlled by more than one dominating mechanism, or is governed by different mechanisms at different scales. In addition, spatial variations are stronger in large reactors (Leng, 1991; Tatterson, 1994), and the isoenergetic curves may be different in vessels of different volume, even at geometric similarity (Bourne and Dell′ava, 1987). Spatial gradients are particularly strong in the impeller region, and the exact location of the feed pipe can be critical, especially in reaction crystallization, since crystallization kinetics depends strongly on the supersaturation and spatial variations in supersaturation may be strong.

Competing chemical reactions in a semibatch stirred-tank reactor have been successfully reproduced at larger scale by scaling up at constant power per unit volume (Bourne and Dell′ava, 1987). The measured product distribution became independent of scale when feeding in the impeller region. However, when feeding in the bulk region, the product distribution was not reproduced and it was shown that the feed plume is more quickly drawn into the impeller region in the small tank reactor than in the large stirred tank. In this sense the level of mixing is higher in the small tank reactor. Rice and Baud (1990) also found scale-up at equal power input per unit volume to be successful for agitated tanks where the feed is supplied through three feed pipes, simultaneously. Rice and Baud (1990) suggest changing the impeller to tank diameter ratio in order to further improve the correlation. On the other hand, Tipnis et al. (1994) found for competing chemical reactions in semibatch stirred-tank reactors, that experimental data agreed well at equal stirring rate, while scaling-up at constant power per unit volume failed, as well as at constant tip speed. Scaling-up of semibatch processes with fast chemical reactions at constant tip speed was not successful in the works of Wang and Mann (1992) and of Mann and El-Hamouz (1995).

There are only few studies on scale-up in the field of reaction crystallization. Vrhunec et al. (1999) performed semi-batch experiments on sodium perborate in industrial scale and in an 80 L vessel, and obtained equal size distributions at equal stirring rate. In crystallization of calcium oxalate (Zauner and Jones, 2000a) in crystallizers of a different size, the experimental results could not be correlated to the stirring rate, and results on calcium carbonate (Zauner and Jones, 2000b) cannot be scaled up by keeping the local energy dissipation rate constant. However, calcium oxalate crystallizes as either single crystals or agglomerates and calcium carbonate forms different polymorphs, and these facts complicate the interpretation of the results.

Torbacke and Rasmuson (2001) studied semi-batch reaction crystallization of benzoic acid in a loop reactor. The experiments were arranged so that the circulation time, the level of the feed point mixing intensity, and the level of mesomixing could be changed independently of each other. It was concluded that especially mesomixing was important to the product weight mean size, but existing theories could not provide a satisfactory explanation and correlation of the results. However, the results could be fairly well correlated by a dimensionless number describing the ratio of the need for mixing to the rate of mixing provided. In this work, experiments are performed in agitated tanks of various sizes. The influence of process variables, especially the feed pipe diameter is evaluated. Using the experimental results, and by comparison with data from previous work, the influence of reactor size is examined. Results are explained and discussed with reference to current mixing theories, and the previously proposed dimensionless number is examined within the framework of the inertial-convective theory.

Theory

Because of the complexity, a complete theory over turbulent mixing in an agitated tank cannot be given. However, specific features of certain parts of the mixing process can be treated in a semiempirical manner, and the rate determining step can be elucidated if time constants are defined and compared.

The rate of macromixing can be related to the macroscale circulation time and, hence, to the agitation rate. The mean circulation time is

$$\tau_{circ} = \frac{V}{Q_{bulk}} = \frac{V}{N_q N D^3} \tag{1}$$

where V is the tank volume, Q_{bulk} is the volumetric bulk flow, N is the agitation rate, N_q is the flow number, and D is the impeller diameter.

In modern theoretical treatment, micromixing accounts for viscous-convective mixing, below the Kolmogoroff microscale, and viscous-diffusive mixing, below the Batchelor microscale and includes the ultimate molecular diffusion. The rate of micromixing is directly related to the rate of dissipation of the turbulent energy, ε (Wu and Patterson, 1989). The most frequent definition of the micromixing time constant appears to be

$$\tau_{micro} = C \sqrt{\frac{\nu}{\varepsilon}} \tag{2}$$

where ν is the kinematic viscosity. The constant C varies from 3.5–17.3 in the literature for the physical properties of water (Corrsin, 1964; Baldyga and Bourne, 1999; Geisler et al., 1988). The most common value seems to be 12.7 (Baldyga and Bourne, 1986), which is used in this study if nothing else is stated.

Mesomixing has been much less studied, but two mechanisms have been proposed: the turbulent dispersion mechanism and the inertial-convective disintegration mechanism (Baldyga

and Bourne, 1999). For turbulent-dispersive mesomixing, Baldyga et al. (1993), defined a characteristic length scale, d_D for the initial dispersion of the feed

$$d_D = \sqrt{\frac{Q_{feed}}{u_{bulk}}} \tag{3}$$

Q_{feed} is the volumetric flow rate of the feed, and u_{bulk} is the mean velocity of the bulk flow passing the feed point. The feed flow is assumed to rapidly adopt the velocity of the bulk flow passing the feed pipe. If the linear bulk flow velocity is higher than the linear feed flow velocity in the pipe, the feed is strained to a reduced cross-sectional dimension when it enters into the bulk. Equation 3 is essentially the continuity equation applied to the feed under these conditions.

If values of d_D are smaller than the feed pipe diameter, the feed pipe represents a local finite source (Baldyga et al., 1993), and the feed plume mixing according to the turbulent dispersion theory is described by

$$X^0_{feed}(t) = \frac{4Q_{feed}}{\pi d^2 u_{bulk}} \left(1 - \exp\left(-\frac{d^2}{16 D_t t}\right)\right) \tag{4}$$

The higher the mixing is, the lower is the value of X^0_{feed} describing the concentration of the feed fluid along the axis of the feed plume at a distance from the feed point corresponding to the time t. The initial condition, that is, when the feed fluid just enters into the tank, is given by

$$X^0_{feed} \rightarrow \frac{4Q_{feed}}{\pi d^2 u_{bulk}} = \frac{u_{feed}}{u_{bulk}} = U \quad \text{when} \quad t \rightarrow 0. \tag{5}$$

At very long times, Eq. 4 simplifies into

$$X^0_{feed} \rightarrow \frac{Q_{feed}}{4\pi u_{bulk} D_t t} \quad \text{when} \quad t \rightarrow \infty, \tag{6}$$

The time constant for the turbulent dispersive mechanism can be estimated by (Baldyga and Bourne, 1992)

$$\tau_{TD} = \frac{Q_{feed}}{u_{bulk} D_t} = \frac{d_D^2}{D_t} \tag{7}$$

which relates to that d in Eq. 4 is replaced by d_D. The turbulent diffusivity D_t can be estimated by (Baldyga and Bourne, 1999; Versteeg and Malalasekera, 1995)

$$D_t = 0.1 \frac{k^2}{\varepsilon} \tag{8}$$

where k is the turbulent kinetic energy and ε is the local energy dissipation rate. The turbulent kinetic energy k is

$$k = \frac{1}{2}(u^2_{rms} + v^2_{rms} + w^2_{rms}) = \frac{3}{2}(u^2_{rms}) \tag{9}$$

where u_{rms}, v_{rms} and w_{rms} are the root-mean-squares of the fluctuating velocities in three different directions. Often the turbulence is assumed to be isotropic and then the second equality holds.

Baldyga and Bourne (1999, p. 593) propose that another time constant for turbulent dispersion can be estimated as a complement

$$\tau_{TD} = \frac{d^2}{4D_t} \tag{10}$$

However, a clear reasoning is unfortunately not given. The general format agrees with Eq. 7, but the scale of segregation is rather described by the feed pipe diameter.

The origin of the inertial-convective disintegration mechanism can be traced back to the early works by Corrsin (1964) and Rosensweig (1964). The dissipation of concentration unmixedness in a turbulent field is assumed to proceed through a spectral cascade process, from large concentration eddies through progressively smaller eddy sizes, for final dissipation at the smallest scales (Corrsin, 1951). This resembles very much the transfer of turbulent energy from large eddies to progressively smaller eddies and dissipation into heat. In terms of a time constant the result of Rosensweig (1964) can be expressed as (Baldyga and Bourne, 1986)

$$\tau_{IC} = \frac{\sqrt{\pi}\Gamma(1/3) C_\gamma}{2\Gamma(5/6)} \left(\frac{k_0^{-2}}{\varepsilon}\right)^{1/3} = A\left(\frac{\Lambda_\gamma^2}{\varepsilon}\right)^{1/3} \tag{11}$$

where k_o is a characteristic wave number of big concentration eddies, and Λ_γ is the integral scale of concentration fluctuations. k_0 is almost equal to the inverse of the integral scale of concentration fluctuations: $k_0 = 0.75/\Lambda_\gamma$ and C_γ is assumed to have a universal value which is proposed to be approximately 1/3 (Rosensweig, 1964). Hence, the constant A becomes 0.85. In a system with feed pipes it is assumed that Λ_γ is proportional to the hydraulic diameter of the inlet (Rosensweig, 1964), and it is explicitly suggested that smaller feed pipes favor higher mixing rates. Hence, in the following we assume that Λ_γ equals the inner diameter of the feed pipe, if nothing else is stated.

A so-called homogenization time constant was proposed by Corrsin (1964)

$$\tau_{Corr} = 2\left(\frac{\Lambda^2}{\varepsilon}\right)^{1/3} + \frac{\ln Sc}{2}\sqrt{\frac{\nu}{\varepsilon}} \tag{12}$$

for Schmidt numbers much larger than unity. The first term on the righthand side involves mixing in the inertial-convective subrange, and the second term involves mixing in the viscous-convective subrange. Hence, this time constant includes both mesomixing and micromixing and A in Eq. 11 has the value of 2, which is commonly used. In this work if nothing else is stated the time constant for inertial-convective mesomixing is calculated as

$$\tau_{IC} = 2\left(\frac{d^2}{\varepsilon}\right)^{1/3} \tag{13}$$

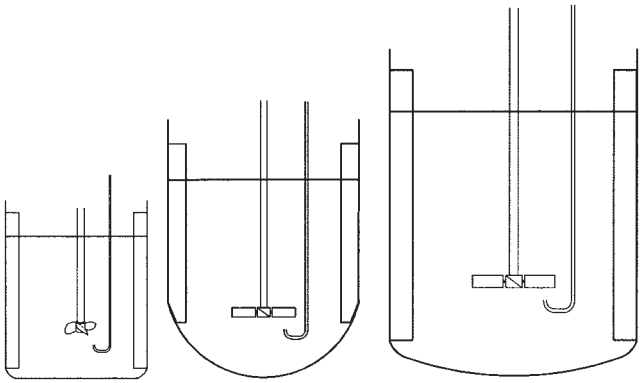

Figure 1. Stirred-tank reactors (2.5 L, 10 L and 200 L).

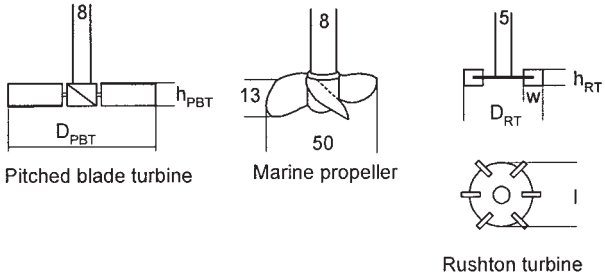

Figure 2. Agitators.

Torbacke and Rasmuson (2001) studied semi-batch reaction crystallization of benzoic acid in a loop reactor, and found that the results could be fairly well correlated by a dimensionless number *TR* being the ratio of the total feeding time to a mixing time constant

$$TR = \frac{t_f}{\tau_{mix}} \tag{14}$$

It was found that the mixing time constant, τ_{mix} could be successfully described by a time constant aimed at characterizing primarily the mesomixing time

$$\tau_{meso} = K \frac{d}{u_{bulk}} \tag{15}$$

and results were correlated against

$$TR = \frac{u_{bulk} t_f}{d} \tag{16}$$

TR was interpreted as a dimensionless number describing the need for mixing relative to the rate of mixing provided.

Equation 16 can be analyzed within the framework of the inertial-convective mixing theory described by Eq. 11. The local turbulent energy dissipation rate in an agitated tank is often approximated by (Pettersson and Rasmuson, 1997)

$$\varepsilon = C_T \frac{u_{rms}^3}{\Lambda_T} \tag{17}$$

under the assumption that the turbulence is isotropic. Λ_T is the macroscale of turbulence. The root-mean-square of the fluctuating velocity u_{rms} may be related to the mean flow rate by the intensity of turbulence T_U

$$T_U = \frac{u_{rms}}{u_{bulk}} \tag{18}$$

Insertion of Eqs. 17 and 18 into Eq. 11 leads to

$$\tau_{IC} = A T_U^{-1} \left(\frac{\Lambda_T}{d}\right)^{1/3} \frac{d}{u_{bulk}} = B \frac{d}{u_{bulk}} \tag{19}$$

by which a basis for Eq. 15 is established within the inertial-convective disintegration theory. *K* can obviously be regarded as a parameter that depends on the local macroscale and intensity of turbulence.

Experimental Work

Apparatuses

Semibatch reaction crystallization of benzoic acid has been carried out in three sizes of agitated tanks: 2.5 L, 10 L, and 200 L. Reactors are illustrated in Figure 1 and agitators in Figure 2, and data are given in Table 1. Geometric similarity has not been preserved, and this is addressed in the discussion. However, all reactors have four stainless steel baffles having a width equal to 1/10 of the tank diameter (in the 10 L tank they are only 20 mm). They all have an agitator with a diameter being 1/3 of the tank diameter and placed at a clearance height from the bottom equal to 1/3 of the tank diameter. They are all initially filled with liquid to a height equal to the tank diameter. The feed pipes were positioned half the length of the impeller blade horizontally away from the rotation axis.

The jacketed 2.5 L glass reactor has 150 mm internal dia. and a flat bottom, and was agitated by a 50 mm marine propeller. A two-piston pump (Desaga KP 2000) was used for feeding. The glass feed pipe had an inner diameter of 1.5 mm, and the feed point was positioned approximately 10 mm below the propeller.

The jacketed 10 L glass reactor has 210 mm internal diameter and a rounded bottom, and was agitated by a 70 mm pitched blade turbine. The same two-piston pump as earlier was used. The feed pipes of acid-resistant steel had inner diameters of 1.0 mm, 1.8 mm, 2.7 mm, and 3.8 mm. The feed point was positioned 15–20 mm below the propeller with the opening pointing at the impeller outflow. In one experiment with a 1.0 mm feed pipe diameter the feed pipe was pointing at the vertical center-line of the stirred- tank reactor, instead of upward at the impeller outflow.

The 200 L glass reactor has a 0.6 m internal diameter and a

Table 1. Impellers

Impeller	*D*	*h*	*w*	*l*
PBT 70 mm	70 mm	10 mm		
PBT 200 mm	200 mm	30 mm		
RT 25 mm	25 mm	5 mm	6 mm	18 mm

Table 2. Experimental Conditions

Stirred Tank Volume	Initial Volume	Added Volume	Total Feed Time (min)	Stirring Rate (rpm)
2.5 L	1930 ml	483 ml	90	500
10 L	7300 ml	1825 ml	60, 90, 120, 180	300–900
200 L	150 L	37.3 L	42, 195, 274	170–350

weakly rounded bottom, and was agitated by a 0.2 m pitched blade turbine. Feed flow was obtained by gravity. The hydrochloric acid was stored in a can placed on a balance two stories above the stirred-tank reactor. The flow of acid was controlled by a valve, and was recorded by the weight change with time. The accuracy of the feeding procedure is ± 10 g/min. The feed pipe was made of acid-resistant steel, and had an inner diameter of 15 mm. A reduced inner diameter (to either 5 or 10 mm) was obtained by Teflon plugs inserted into the feed pipe. The feeding point was fixed at approximately 40 mm below the propeller.

Procedures

In all experiments, 1.4 M hydrochloric acid is added to a 0.35 M aqueous solution of sodium benzoate. At the end of the experiment, the amount of added hydrochloric acid is stoichiometric to the sodium benzoate originally in the solution (Table 2). The hydrochloric acid is prepared from concentrated hydrochloric acid (p.a quality except for the 200 L scale where technical quality is used), and filtered distilled water in all experiments. The sodium benzoate solution is prepared from solid sodium benzoate (purum quality except for the 200 L scale where technical quality is used) and distilled water in all experiments. In the 2.5 L experiment the sodium benzoate solution is initially saturated by benzoic acid, but this is not the case in the 10 L and the 200 L scales. The 2.5 L and the 10 L experiments were performed at 30 °C, while the 200 L experiments were performed at ambient temperature (that is, 19.7–22.1 °C). In the 2.5 L experiments, the sodium benzoate solution was thermostated at 30 °C and, thereafter, filtered. In the 10 L experiments, the sodium benzoate solution is thermostated at 30 °C without further filtering of the solution. The total feeding time is varied from 42 to 274 min. The impeller stirring rate is varied from 170 to 900 rpm (Table 2).

The product size distribution is determined by an electrosensing zone instrument (ELZONE® 180 XY). One sample of 20 mL is withdrawn with a syringe, with a hose connected to it, from the suspension at the end of each experiment. The sample is taken in the bulk (closer to the wall than to the stirring axis) where the liquid moves upward. In order to disintegrate aggregated crystals, three droplets of dispersing agent are added to the sample, which is then treated three times for 5 s in an ultrasonic bath. A weighed amount of the disintegrated sample is withdrawn with a blunted Pasteur pipette, and added to a weighed amount of electrolyte solution in a beaker thermostated at 30.0 °C. The electrolyte is a distilled water solution of sodium chloride saturated with benzoic acid at the temperature of interest by contact with an excess amount for at least 24 h. The electrolyte solution is filtered through a 0.2 μm membrane filter before each measurement. An orifice tube of 300 μm is used for the measurements. The volume measured each time is 5.00 mL.

Results

One experiment was performed in the 2.5 L scale, 21 experiments in the 10 L scale, and 3 experiments in 200 L scale. The feed pipe diameter, the total feeding time, and the stirring rate have been varied in the experiments. The result presentation focuses on the product weight mean size, which is estimated as previously (Torbacke and Rasmuson, 2001).

The results from the 10 L experiments are given in Table 3. Almost half of the experiments have been repeated once, and the difference in product mean size between experiments at equal conditions is usually less than 1 μm. In three cases the difference is larger than this, which is believed to relate to difficulties in exactly reproducing the feed point location. In the experiments that have been repeated, an average is calculated which is used in the presentation of the results. The results show that the weight mean size increases with increased stirring rate (Figure 3) for all but one condition (1.0 mm, 90 min). At 1.0 mm feed pipe diameter, 500 rpm and 90 min total

Table 3. 10 L Stirred Tank Reactor Experiments

d (mm)	t_f (min)	N (rpm)	τ_{TD} (s)	τ_{IC} (s)	τ_{micro} (s)	τ_{IC}/τ_{micro}	L_{43} (μm)
1.0	60	500	0.0008	0.021	0.014	1.5	36.8
1.0	90	500	0.0006	0.021	0.014	1.5	42.7
1.0	90	700	0.0003	0.015	0.008	1.9	42.2
1.0	120	500	0.0004	0.021	0.014	1.5	49.6
1.0	180	500	0.0003	0.021	0.014	1.5	51.1
1.0	180	900	0.00009	0.012	0.006	2	60.1
1.8	60	500	0.0008	0.032	0.014	2.3	32.7
1.8	90	500	0.0006	0.032	0.014	2.3	39.3
1.8	180	500	0.0003	0.032	0.014	2.3	49.7
2.7	90	300	0.002	0.069	0.030	2.3	36.8
2.7	90	400	0.0009	0.052	0.020	2.6	36.6
2.7	60	500	0.0008	0.042	0.014	3	37.9
2.7	90	500	0.0006	0.042	0.014	3	41.1
2.7	120	500	0.0004	0.042	0.014	3	43.4
2.7	180	500	0.0003	0.042	0.014	3	45.8
2.7	90	600	0.0004	0.035	0.011	3.2	39.0
2.7	90	900	0.0002	0.023	0.006	3.8	43.1
3.8	55	300	0.003	0.087	0.030	2.9	33.0
3.8	60	500	0.0008	0.052	0.014	3.7	39.9
3.8	90	500	0.0006	0.052	0.014	3.7	38.7

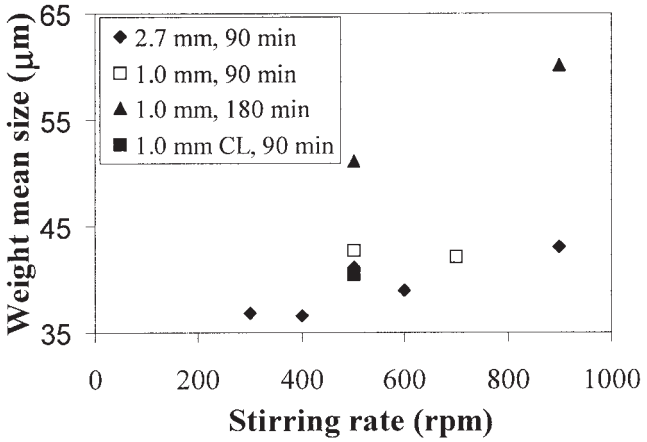

Figure 3. Influence of the stirring rate in the 10 L stirred-tank reactor.

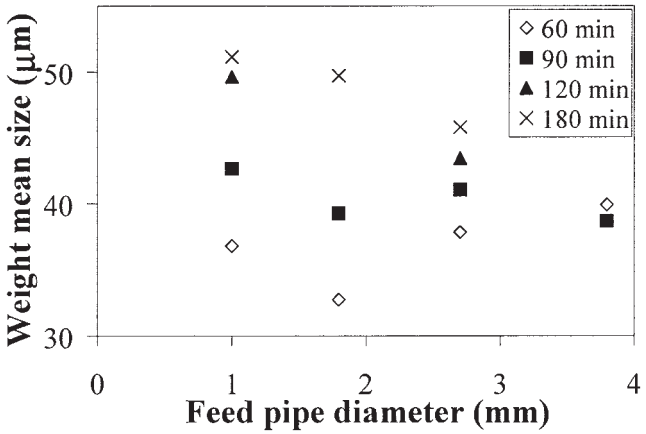

Figure 5. Influence of the feed pipe diameter in the 10 L experiments (N=500 rpm).

feeding time, the weight mean size is not significantly affected by whether the feed pipe points toward the outflow of the agitator (42,7 μm), or in a direction perpendicular to this outflow (40,5 μm). This agrees with the findings in the loop reactor experiments (Torbacke and Rasmuson, 2001). The weight mean size increases with increasing total feeding time for all feed pipe diameters apart from the 3.8 mm feed pipe diameter (Figure 4). The weight mean size decreases with increasing feed pipe diameter (Figure 5) at lower feed rates, but the behavior is more complex at higher feed rates. Each of the 60 and 90 min experiments using the 1.8 mm feed pipe diameter have been repeated once, and the mean size is reproduced within 0.6 and 1.5 μm, respectively. Also, the 90 min experiment with a 1.0 mm feed pipe diameter has been repeated, and the mean size differs by 3.4 μm - a difference that does not influence the trends in Figures 4 and 5.

In the 200 L stirred-tank reactor experiments, the stirring rate, the feed pipe diameter, and the total feeding time were all changed simultaneously. The strategy was to study the influence of the TR number (Torbacke and Rasmuson, 2001) on the weight mean size. Experiments were performed at low, at intermediate, and at high TR number (Table 4). Reproducibility

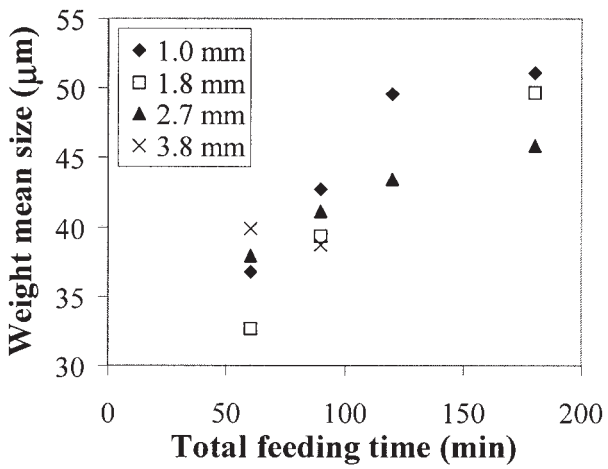

Figure 4. Influence of the total feeding time in the 10 L experiments (N=500 rpm).

experiments have not been performed, but from each experiment three samples have been taken, and the standard deviation is given in the table. The results show that the weight mean size increases with increasing *TR* number.

The 2.5 L stirred-tank reactor experiment was performed to verify that the results previously obtained in the loop reactor could be included in a scale-up evaluation. The weight mean size obtained agrees fairly well with the experimental results obtained in the loop reactor at similar mixing conditions (Torbacke, 1998).

The results show that the volume of the reactor does not exhibit a dominating influence on the weight mean size (Figure 6). The influence of the different processing conditions in each tank is at least as strong as the influence of the tank volume even at equal total feeding time.

The coefficient of variation (CV), that is, the standard deviation of the size distribution divided by the weight mean size is approximately 0.4 in these experiments. Åslund and Rasmuson (1992) obtained CV-values of about 0.3 for the same system in the 1 L experiments.

In this work, the product crystals of each experiment were examined in a microscope, but no quantification was made with respect to shape or other features. Product crystals are shaped like platelets and are similar to those of the previous loop-experiments (Torbacke and Rasmuson, 2001). We have not noticed a specific trend in the shape vs. tank reactor size. In short experiments the rhombic shape is less well developed, and the crystals tend to be more rounded. In longer experiments and at higher agitation rates crystal corners are damaged. However, since the weight mean size increases monotonously with increased agitation, the importance of attrition and breakage should not be that significant.

Analysis and Discussion

The 10 L results show that the stirring rate, the total feeding time, and the feed pipe diameter all influence the product weight mean size regardless of vessel volume. Increased agitation and reduced feeding rate lead to increased product weight mean size, which is in agreement with our previous results in the loop reactor (Torbacke and Rasmuson, 2001), and in a smaller agitated tank (Åslund and Rasmuson, 1992). There

Table 4. 200 L Stirred Tank Reactor Experiments

d (mm)	t_f (min)	N (rpm)	u_{res} (m/s)	τ_{TD} (s)	τ_{IC} (s)	τ_{micro} (s)	τ_{IC}/τ_{micro}	TR	L_{43} (μm)
15	42	170	0,81	0.01	0.18	0.02	9	1.36 E5	20.5 ± 0.4
10	274	250	1,29	0.0007	0.094	0.01	9.4	2.11 E6	40.6 ± 1.1
5	195	335	1,86	0.0005	0.042	0.008	5.3	4.34 E6	51.8 ± 3.9

is a significant influence of the feed pipe diameter, and this was also found in the loop reactor experiments. In the loop reactor, the volume is approximately 2.5 L. Åslund and Rasmuson (1992) used an active volume of about 1 L. Both studies will be included in the analysis of the influence of crystallizer size, and for this reason data for estimation of the time constants for the conditions of these experiments are also given together with the data needed for the experiments of this study.

Introductory analysis

There is clear evidence, especially in the results from the loop reactor (Torbacke and Rasmuson, 2001), that increased mixing does lead to larger particles for the system where benzoic acid is crystallized by mixing hydrochloric acid into an aqueous solution of sodium benzoate. The chemical reaction is a simple acid-base reaction, and is believed to be infinitely fast compared to the rate by which crystallization and mixing proceed. The mean macroscale circulation time is estimated by Eq. 1. N_q is equal to 0.5 for a marine propeller and 0.8 for a Rushton turbine (Oldshue, 1983), and is equal to 0.8 for a pitched blade turbine (Weetman and Oldshue, 1988). The mean circulation times are 4 s (2.5 L), 2.2–6 s (10 L) and 5–10 s (200 L), that is, much shorter than the total feeding times, and, hence, the process can be regarded as completely macro-mixed (Baldyga et al., 1997).

At least two observations support that mesomixing is of importance. The product crystal mean size increases with increasing feeding time, but this increase levels off at long total feeding times. The same was found in our previous studies (Åslund and Rasmuson, 1992; Torbacke and Rasmuson, 2001), and this is a characteristic of a process influenced by mesomixing (Baldyga et al., 1997). For long feeding times, the weight mean size is expected to depend on micromixing only. When the feed flow increases, the weight mean size decreases because the early mixing of the feed, the mesomixing, is insufficient. The second observation is that the weight mean size is significantly influenced by the feed pipe diameter (Figure 5). If micromixing only is governing the process there should be no such influence.

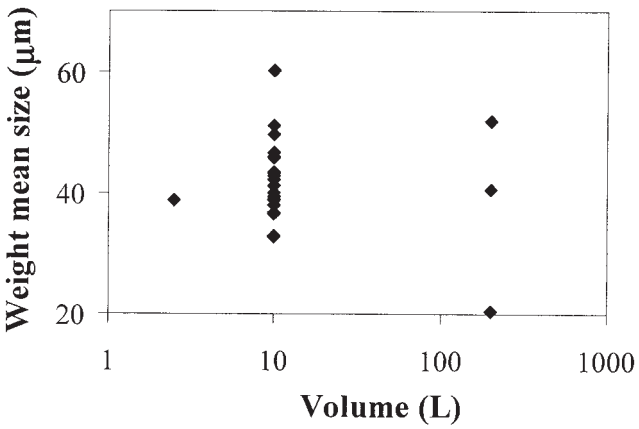

Figure 6. Influence of the reactor volume.

The turbulent-dispersive mesomixing length scale d_D is estimated by Eq. 3. The velocity, u_{bulk} is calculated as the resultant velocity u_{res} of the axial, radial and tangential contributions of the bulk flow at the feed point

$$u_{res} = \sqrt{u^2 + v^2 + w^2} = \sqrt{(C_a N\pi D)^2 + (C_r N\pi D)^2 + (C_t N\pi D)^2}. \quad (20)$$

The constants C_a, C_r and C_t, are given in Table 5. The mean tangential velocity is not as well documented as the radial and axial mean velocities. For both the Rushton turbine and the marine propeller, both for bulk and impeller feeding, the constant C_t has been given estimated values (Wu and Patterson, 1989; Ng et al., 1998; Lee and Yianneskis, 1998; Geisler, 1991). For these experiments, d_D becomes: 0,5 mm (2.5 L), 0.34–1.31 mm (10 L), and 1.42–4.4 mm (200 L). There is a tendency for the length scale to increase with increasing crystallizer size. The values of d_D are smaller than the feed pipe diameter in all but two cases of the 10 L stirred-tank reactor using the 1.0 mm feed pipe, and, hence, the model behind Eq. 4 should be used to describe the dispersion of the feed.

The laminar feed flow is added to a turbulently mixed bulk. The impeller Reynolds number is $2.1 \cdot 10^4$ in the 2.5 L experiment, at least $2.4 \cdot 10^4$ in the 10 L experiments, and at least $1.1 \cdot 10^5$ in the 200 L experiments. With only few exceptions, the linear feed flow rate is much lower than the linear bulk flow rate at the feeding point. The kinetic energy of the laminar feed flow can be compared with the turbulent kinetic energy of the bulk (in terms of J/m^3)

$$E^V_{feed} = \frac{\rho u^2_{feed}}{2} \quad (21)$$

$$E^V_{bulk} = \rho k \quad (22)$$

The ratio E^V_{feed}/E^V_{bulk} is always lower than unity and most often much lower than unity. Values above 0.10 are only found in the 10 L tank and for the 1.0 mm feed pipe, the highest value being 0.9. This suggests that the influence of the feed flow on the mixing intensity at the feed point is negligible in most experiments $U \ll 1$ and hence backmixing can occur. However, as is discussed below, we believe that backmixing is only of importance for the largest feed pipe diameter.

Examination of the governing mixing mechanism

If we carefully examine the 10 L results at constant agitation rate equal to 500 rpm, the following observations can be made as is summarized in Table 6:

Table 5. Constants Used for Calculating the Resultant Bulk Velocity

	RT Impeller	RT Bulk	MP Impeller	MP Bulk	PBT Impeller	PBT Bulk
C_a	0.03	0.05	0.23	0.04	0.38	0.05
C_r	0.28	0.08	0.05	0.11	0.05	0.08
C_t	0.09	0.03	0.05	0.04	0.2	0.05

RT = Rushton turbine (Geisler, 1991).
MP = marine propeller (Geisler, 1991).
PBT = pitched blade turbine (Pettersson, 1996).

(1) If Q_{feed} increases at constant d or at constant u_{feed}, the mean size always decreases strongly, Figures 4 and 7, except for the largest feed pipe (d=3.8 mm) (where we believe that backmixing is influencing).

(2) If d increases at constant u_{feed}, the mean size always decreases strongly (Figure 7).

(3) If d increases at constant Q_{feed}, the mean size decreases at lower feeding rates (Figure 5). At the highest feeding rate, the dependence is the same for the small feed pipes, but becomes essentially reversed for larger feed pipe diameters. At the feeding rate corresponding to a total time of 90 min, the influence of the feed pipe diameter is fairly weak.

(4) If u_{feed} increases at constant d, the mean size decreases strongly (Figure 7) except for the largest feed pipe (d=3.8 mm) (where we believe that backmixing is influencing).

(5) If u_{feed} increases at constant Q_{feed}, the mean size increases at low feed rates (Figure 7). This influence is the same for high feed rates and small feed pipes, but changes into the opposite for the larger feed pipes.

In the turbulent dispersion theory, the characteristic mesomixing length d_D and the time constant increases when Q_{feed} increases. The theory could then possibly explain observation 1, but cannot explain observation 3. However, at very short times, Eq. 5 is applicable, in which the mixing rate depends on the feed pipe diameter even at constant volumetric feed flow. However, then the theory predicts that the mixing rate increases at increasing feed pipe diameter, which is opposite to our observations 2 and 3. As shown in Figure 5, the overall tendency is that the mean size decreases and, hence, the mixing rate decreases at increasing diameter. In Eq. 5, the linear feeding rate is the governing parameter, not the volumetric feeding rate nor the feed pipe diameter. This agrees with observation 4, but opposes much of observation 5, and definitely observation 1. As shown in Figure 8, the results cannot be well correlated by the parameter U. The same was found in the loop reactor study (Torbacke and Rasmuson, 2001). At very long times, the influence of the feed pipe diameter vanishes, as shown in Eq. 6.

Table 6. Influence of Process Variables on Product Weight Mean Size

Mean Size	Q_{feed}	u_{feed}	d	Shown in Figure
↓[1]	↑	C		4 and 7
↓[1]	↑		C	4 and 7
↓		C	↑	7
↓[2]	C		↑	5
↓[1]		↑	C	7
↑[2]	C	↑		7

↑ = increases, ↓ = decreases, C = constant.
1. Except for the largest feed pipe (d = 3.8 mm) where we believe that backmixing is influencing.
2. Except for at high feeding rate and larger feed pipes where it is opposite.

The time constant of the inertial-convective mesomixing, Eq. 13, decreases with decreasing feed pipe diameter. This theory could then possibly explain much of observations 2 and 3. However, in the inertial-convective mesomixing there is no dependence on the volumetric feed flow rate nor of the linear feed rate. The three parameters Q_{feed}, u_{feed} and d are, of course, not independent. It is our belief that the governing parameters in this work are the volumetric feed rate and the feed pipe diameter. As shown earlier, the feed flow should have no influence on the mixing itself in this study. The volumetric feed rate instead represents the need for mixing: at increased feeding rate, the mixing rate has to increase in order to keep the conditions unchanged. The feed pipe diameter determines the initial scale of segregation of the feed, such that the mixing process overall takes longer time for a larger feeding pipe because the mixing has to start from a larger scale of segregation. Qualitatively, the experimental results of this work are better explained by the inertial-convective mixing mechanism than by the turbulent dispersion mechanism.

Time constants have been estimated and are presented in Tables 3, 4, and 7. The time constant for turbulent dispersion is estimated by Eqs. 7–9. It is assumed that, u_{rms}/u_{tip}, v_{rms}/u_{tip} and w_{rms}/u_{tip} are 0.07, 0.12, and 0.08, respectively, for the pitched blade turbine in the bulk region, and 0.13, 0.12, and 0.12, respectively, in the impeller region (Pettersson, 1996). For the Rushton turbine, the turbulence is assumed to be isotropic and in the impeller region u_{rms}/u_{tip} is 0.256 (Lee and Yianneskis, 1998). The local energy dissipation rate is estimated by adopting a power number of 0.5 for the marine propeller, 1.3 for the pitched blade turbine, and 5 for the Rushton turbine. In the impeller region we assume that the local dissipation rate is 5, 6, and 15 times the mean value for the marine propeller (Geisler, 1991), the pitched blade turbine (Jaworski and Fort, 1991), and the Rushton turbine (Baldyga et

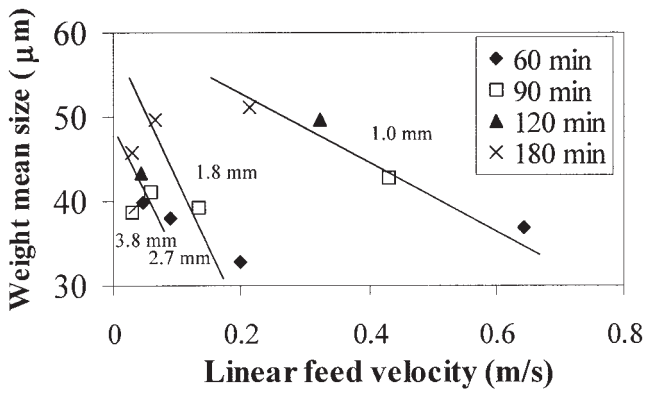

Figure 7. Influence of the linear feed velocity in the 10 L experiments (N=500 rpm).

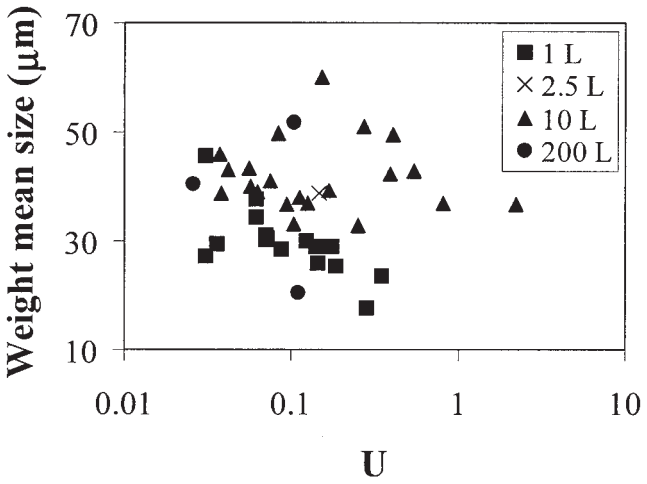

Figure 8. Influence of the ratio U.

al., 1995), respectively. In the bulk region we assume that the values are correspondingly 0.5, 0.4, and 0.25. The time constant of inertial convection is estimated by Eq. 13, and for micromixing by Eq. 2 using C=12.7. The time constant values should be regarded as rough estimates since the local values of the turbulent diffusivity, the energy dissipation rate, and the resultant bulk flow velocity are only estimates based on data from the literature.

The estimations show that the mesomixing time constant for the inertial-convective mechanism is always significantly longer than that of the turbulent dispersion mechanism, and is usually much longer than the time constant of micromixing. It seems reasonable to assume that to some extent turbulent-dispersion operates ahead of inertial-convective disintegration, and that micromixing acts essentially in series with mesomixing. Hence, we conclude that the inertial-convective mechanism should be governing the mesomixing step, and that the process is usually more governed by mesomixing than by micromixing. However, micromixing cannot be entirely neglected and can even in some cases be of equal importance as mesomixing. The results also suggest that the process becomes increasingly mesomixing controlled with increasing feed pipe diameter. The process also tends to become more mesomixing controlled at increasing crystallizer volume, and this is in accordance with Baldyga and Bourne (1999). It is noteworthy that the turbulent dispersion time constant, even for the 200 L experiments, is shorter than the micromixing time constant, and that the corresponding time constant ratio of mesomixing to micromixing decreases with increasing vessel size.

At the highest feeding rate, the influence of the feed pipe diameter is somewhat complex, as shown by Figure 5. From the smallest feed pipe, the mean size initially decreases at increasing feed pipe diameter, passes through a minimum at d=1.8 mm, and then increases again. Reproducibility experiments confirm that this is not an artefact of experimental uncertainty. There is a tendency for a similar minimum also in the corresponding curve for the second highest feeding rate. An attractive explanation would be that there is a shift in the determining mesomixing mechanism. At low feed rates, mesomixing would be governed by inertial-convection and the rate of mixing increases with decreasing feed pipe diameter. At high feed rates, the turbulent dispersion becomes limiting, and the rate of mixing increases with increasing feed pipe diameter as described by Eq. 5. However, in the transition range, we would expect turbulent dispersion to be governing at small feed pipe diameters and inertial-convection at larger feed pipe diameters at constant feeding rate. This should then lead to a maximum in the curve over the mean size vs. the feed pipe diameter rather than to a minimum. We may also add that the estimated time constant for turbulent dispersion is always much shorter than that of inertial-convection and that of micromixing. The minimum can neither be explained if backmixing is governing. Backmixing should be of greatest importance at low feeding rates and large feed pipe diameters, and should lead to that the mean size increases at decreasing feed pipe diameter or at increasing feed rate. Backmixing is discussed further below.

The experimental results of this work show that the rate of mixing increases with decreasing feed pipe diameter, and the analysis suggests that the rate of mixing is primarily governed by inertial-convective mesomixing. Baldyga et al. (1993) and Baldyga and Bourne (1999) found experimentally the mixing rate to decrease with decreasing feed pipe diameter at short feeding times. The result was explained in terms of the turbulent-dispersive mechanism and Eq. 4, because the time constants of micromixing and inertial-convective disintegration were shorter. Bourne et al. (1981) and Zauner and Jones (2000) found the rate of feed plume mixing to increase with decreasing feed pipe diameter. If mixing time constants are estimated, the inertial-convective disintegration time constant is always the highest for the conditions of these two studies. The difference in the governing mechanism and in the influence of feed pipe diameter may relate to that U-values actually are higher in the experiments of Baldyga and coworkers. For competing chemical reactions, Tipnis et al. (1994) found in 20 L experiments both an increase and a decrease in mixing rate with decreasing feed pipe diameter. In 200 L experiments the mixing rate increased with a decreasing feed pipe diameter while the opposite behavior is observed in the 600 L experiments. If mixing time constants are estimated, we find that the time constant of the inertial-convective disintegration mesomixing is always higher than that of the turbulent-dispersive mesomixing, and that of the micromixing. The shortest time constants are always obtained by the turbulent dispersion mechanism. No clear influence of the *U*-ratio is found.

Importance of backmixing

Backmixing denotes that the bulk flow is flushed into the feed pipe, and may be of importance since *U* is low. In case of backmixing, reactants reach contact and crystallization may commence, already in the feed pipe at poor mixing conditions, and this may lead to smaller product crystals than expected. Fasano et al. (1992) found for a Rushton turbine and feeding in

Table 7. Mesomixing Time Time Constants

Vol.	Turbulent Dispersion (s)	Inertial Convective Disintegration (s)	d/u_{bulk} (s)
1 L	0.0007–0.003	0.006–0.17	0.001–0.029
2.5 L, loop	0.007–0.51	0.004–0.13	0.002–0.013
2.5 L	–	0.04	0.004
10 L	0.00009–0.002	0.01–0.07	0.001–0.008
200 L	0.0005–0.01	0.04–0.2	0.003–0.02

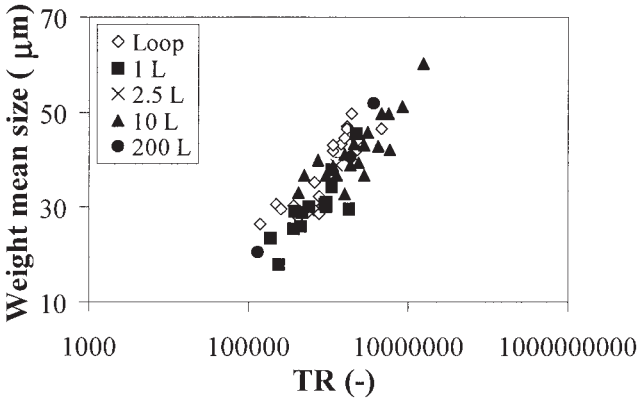

Figure 9. Correlation to TR.

the impeller discharge region that backmixing occurs when the ratio of the linear velocity of the feed flow to the tip speed is lower than 0.18. For a three-bladed pitched blade turbine, backmixing occurred when the ratio is lower than 0.02–0.05, but the exact location of the feed point is not clear. In this study, the ratio of the linear feed velocity to the tip speed is 0.016–0.35 (10 L) and 0.01–0.047 (200 L), but since the geometrical conditions are not fully described by Fasano et al. (1992), a proper comparison is not possible. However, based on the work of Fasano et al. (1992), we believe that backmixing is potentially of importance only in experiments at 500 rpm stirring rate, and where the linear feed velocity is below 0.03 m/s (that is, 2.7 mm feed pipe diameter and 180 min feeding time; 3.8 mm feed pipe diameter, and 90 min). Furthermore, there are several observations that are inconsistent with that backmixing has a dominating influence on the results. Backmixing should increase and the weight mean size should correspondingly decrease when (1) the agitation rate increases, (2) the feed flow rate decreases, and (3) the feed pipe diameter increases. As seen in Figure 3, the mean size never decreases with increasing agitation rate. As shown in Figure 4, the mean size increases at decreasing feed flow rate except for the largest feed pipe. As shown in Figure 5, the mean size does decrease at increasing feed pipe diameter at lower feed rates. At higher feed rate, this effect vanishes and actually reverses. In Figure 7, it is suggested that except for the largest feed pipe, the mean size increases with decreasing feed pipe diameter, at constant linear feed velocity. However, an increasing linear feed velocity leads to a reduced mean size for all feed pipe diameters, but the largest one.

Correlation of results and scaling up

As shown in Figure 6, we do not find a clear influence of the crystallizer size on the product weight mean size. The spread in the product weight mean size depending on the conditions of operation in each crystallizer is more pronounced than the influence of the crystallizer size. This picture remains the same even if we only include data at constant total feeding time. It may be argued that this to some extent is due to lack of geometric similarity. However, retaining geometric similarity does not capture any mixing similarity (Oldshue, 1985). In this work we try to correlate the results for different process conditions as well as different crystallizer sizes.

As shown in Figure 9, all data can be quite well correlated by the dimensionless number TR, Eq. 16, regardless of reactor type and geometry, reactor volume, agitator type, feed pipe diameter, and total feeding time. The coefficient of correlation is 0.86 for the loop reactor data, 0.82 for the 1 L data, 0.84 for the 10 L data, and 0.99 for the 200 L data. The coefficient of correlation is 0.9 for all data taken together as one experimental set. As shown in Figure 10, the experimental results are correlated fairly well also if the mixing time constant is taken as the Corrsin (1964) homogenization time, Eq. 12. However, the experimental results from the loop reactor and the 1 L stirred tank reactor do not correlate so well with the results of this study. If the mixing time constant in Eq. 14 is calculated according to Eq. 13, the correlation is slightly worse since in this case the influence of micromixing is completely neglected. If the mixing time constant is calculated by Eq. 7, the stirred-tank reactor data are correlated well, but the loop reactor data are not correlated at all with the stirred-tank reactor experiments.

In order to estimate the value of the proportionality constant B, by Eq. 19, we need to estimate the intensity and the macroscale of turbulence. The intensity of turbulence in an agitated tank varies significantly. Pettersson and Rasmuson (1997) report values from 50% in the impeller region up to almost 400% out in the bulk region where low mean velocities prevail. The macroscale of turbulence in an agitated tank is difficult to determine and few results are presented in the literature. Today, it is common to assume that the macroscale is related to the diameter of the trailing vortex and is, hence, determined by the height of the impeller blade. In this case C_T receives the value of unity. Pettersson and Rasmuson (1997) used laser-Doppler anemometry to determine the macroscales in a baffled tank, agitated by a pitched blade turbine. If the total value for the three-dimensional (3-D) flow is estimated as a geometric mean of the values for each coordinate direction

$$\Lambda_{3D} = \sqrt[3]{\Lambda_u \Lambda_v \Lambda_w} \tag{23}$$

it becomes about 1/3 of the impeller diameter or about twice the impeller blade height. Insertion of the intensity and the macroscale of turbulence values for the impeller region lead to B-values ranging from 4 to 6 for the 10 L stirred-tank reactor,

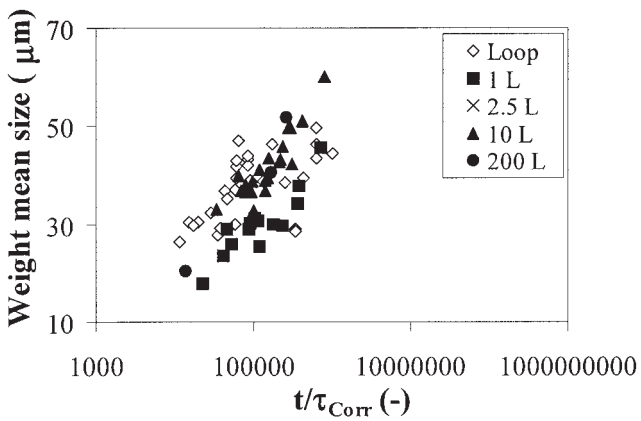

Figure 10. Correlation to the feeding time divided by the homogenisation time constant.

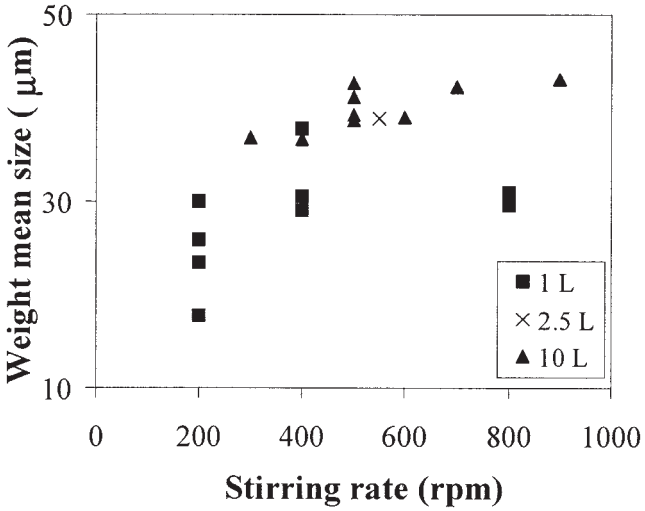

Figure 11. Correlation to the stirring rate.

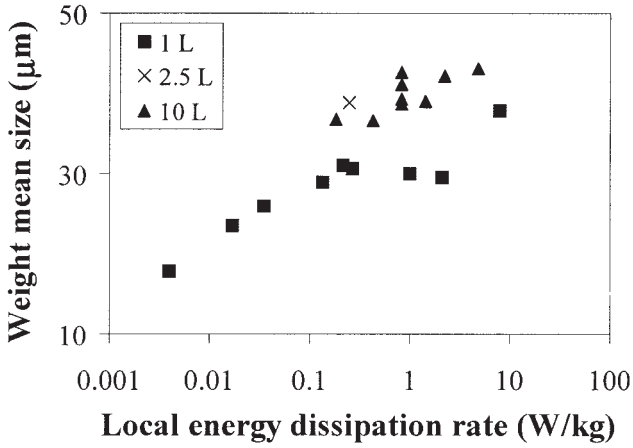

Figure 13. Correlation to the local mixing intensity.

and from 3.6 to 5 for the 200 L stirred-tank reactor. In Table 7 we show the range of mesomixing time constants for the different experiments, and these values are compared with the corresponding range of values for the ratio d/u, in which u is estimated as u_{res} according to Eq. 20. The Table shows that the value of τ_{IC} is about 10 times larger than the value of d/u, which according to Eq. 19 then suggests that the value of B should be approximately 10. However, if A in Eq. 11 is taken as 0.85, instead of 2, the ratio of to d/u in Table 7 reduces to about 4 to 5 which then agrees quite nicely with the B values calculated earlier according to Eq. 19.

The experimental results are plotted against the stirring rate (Figure 11), the impeller tip speed (Figure 12), and the local energy dissipation rate (Figure 13). In these figures only results at equal total feeding time, that is, 90 min, are included and, hence, unfortunately, there are no 200 L experiments in the diagrams. However, the results of Åslund and Rasmuson (1992) and of Torbacke and Rasmuson (2001) are included. Åslund and Rasmuson (1992) observed at high-mixing intensity a decrease in weight mean size with increasing local energy dissipation rate. This is neither observed in this work, nor in the loop reactor experiments (Torbacke and Rasmuson, 2001). Since the mechanism for this decay in weight mean size is unclear, these 1 L results are not included in this work and neither are the experiments where the feed was onto the surface. Figures 11–13 reveal that the three common scale-up rules cannot satisfactorily correlate the results, even though some correlation can be observed. Each of these scale-up rules do only consider a part of the total mixing process, and none is focused on mesomixing.

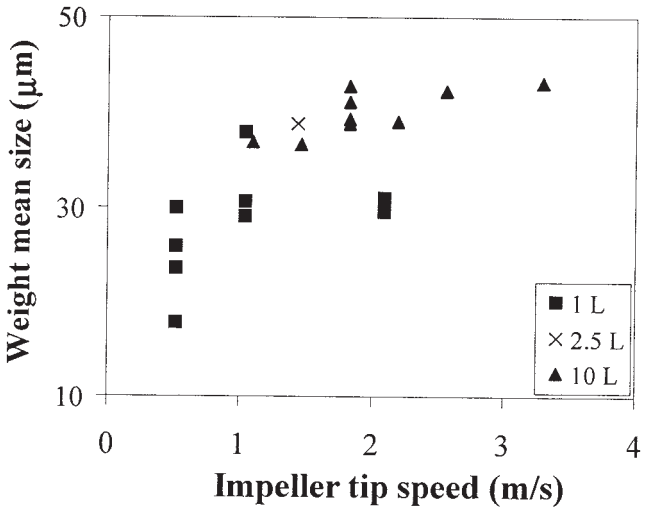

Figure 12. Correlation to the impeller tip speed.

Two-Impeller Experiments

As a small extension of the work, a few experiments in 10 L scale were performed to explore whether a small additional agitator at the feed pipe outlet can provide the required feed point mixing intensity, and replace an increased agitation and mixing in the whole tank. The 70 mm pitched blade turbine was placed as described earlier. A smaller 25 mm Rushton turbine (Figure 2) was placed in the bulk 60 mm above the pitched blade turbine and approximately 60 mm horizontally away from the pitched blade turbine axis (Figure 14). The feed pipe, having a diameter of 1.0 mm, was positioned with the feed point approximately 2 mm above and 5 mm horizontally away from the Rushton turbine tip. The stirring rate of the

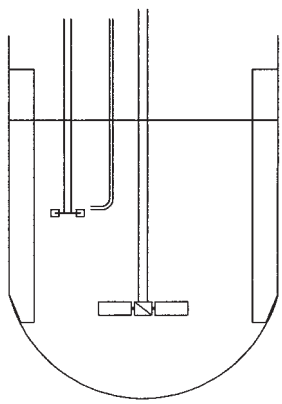

Figure 14. Stirred tank reactor configuration used for the two-impeller-experiments.

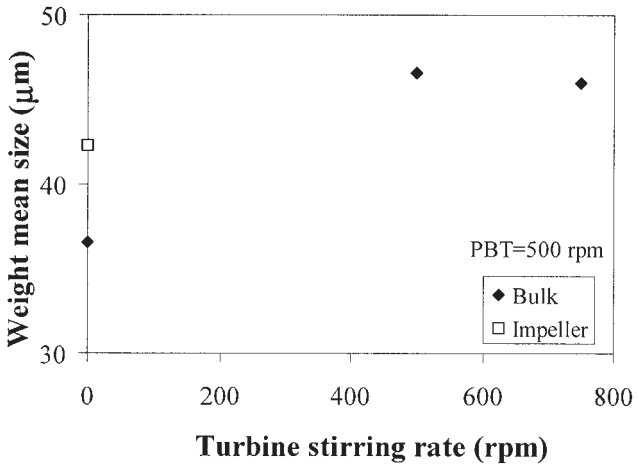

Figure 15. Influence of the local mixing intensity in the two-impeller-experiments.

pitched blade turbine was kept constant at 500 rpm, and the stirring rate of the Rushton turbine was either 0 rpm, 500 rpm or 750 rpm. The total feeding time was 90 min. Results are shown in Figure 15 and are compared with the corresponding single agitator experiment where the feed is close to the pitched blade turbine. The weight mean size decreases significantly when the feed point is moved into the bulk from the impeller region in a one-impeller-experiment. This agrees with the results of Åslund and Rasmuson (1992). However, by applying separate mixing directly at the feed point in the bulk, much larger crystals are produced, and in fact larger than those obtained by feeding close to the main agitator. This size increase is achieved, essentially without increasing the overall power input to the crystallizer. Hence, undesirable side effects of increased overall agitation are avoided such as increased abrasion and breakage, increased energy consumption, and so on. In addition, large crystals can be produced without very long pipes feeding close to the main agitator, and thus mechanical stress on the pipe and risk of damaging the main agitator system are reduced.

Conclusions

The influence of mixing and reactor size on the product weight mean size in semi-batch reaction crystallization of benzoic acid have been investigated. Three different stirred-tank reactors of volumes 2.5 L, 10 L, and 200 L were used, and previous data from experiments in 1 L and 2.5 L scale are included in the scale-up evaluation. The experimental results show that the weight mean size increases with increasing mixing intensity and with decreasing feeding rate. At low feeding rates, the weight mean size increases with decreasing feed pipe diameter. At high feeding rates, the influence of the feed pipe diameter is more complex. Compared to other processing parameters, the weight mean size is not strongly influenced by the reactor size. Backmixing occurs in a few experiments, but is not of major importance in these experimental results.

The analysis shows that the process is mainly governed by mesomixing. The importance of micromixing tends to decrease when the reactor size increases, but is in a few cases as important as the mesomixing. The inertial-convective disintegration mesomixing mechanism can correlate the experimental product weight mean sizes, and explain the influence of different process parameters fairly well. The turbulent dispersion mechanism is possibly of some relevance at the highest feed rates, but the importance of this mechanism otherwise seems to be low in this work. The results are best correlated by a dimensionless parameter being equal to the ratio of the total feeding time to a time constant of mixing. The mixing time constant is defined as being proportional to the feed pipe diameter divided by the resultant linear bulk flow velocity passing the feed pipe. This definition can be related to the inertial-convective mechanism, and the proportionality constant can be calculated from turbulence data over the bulk flow at the feed point. This mixing time constant definition provides a better correlation than the Corrsin homogenization time constant that includes mesomixing and micromixing, which in turn is somewhat better than a correlation, based on the traditional inertial-convective disintegration time constant only.

Acknowledgment

The financial support of the Swedish Research Council for Engineering Sciences (TFR) and the Swedish Industrial Association for Crystallization Research and Development (IKF) is gratefully acknowledged.

Notation

A = constant in Eq. 11
B = constant in Eq. 19
C = constant in Eq. 2
C_a = ratio of axial velocity and impeller tip speed
C_r = ratio of radial velocity and impeller tip speed
C_t = ratio of tangential velocity and impeller tip speed
C_T = universal constant in Eq. 17
C_γ = constant in Eq. 11
d = feed pipe diameter, m
d_D = characteristic length scale of mesomixing, m
D = diameter of impeller, m
D_t = turbulent diffusivity, m^2/s
E^V_{bulk} = kinetic energy of the bulk per unit volume, J/m^3
E^V_{feed} = kinetic energy of the feed per unit volume, J/m^3
h = height of impeller blade, m
k = turbulent kinetic energy, m^2/s^2
k_0 = characteristic wave number of big eddies, m^{-1}
K = proportionality constant in Eq 15
l = Rushton turbine disc dia., m
L_{43} = weight mean size, m
N = stirring rate s^{-1}
N_q = flow number
Q_{bulk} = volumetric bulk flow rate generated by the agitator, m^3/s
Q_{feed} = volumetric feed flow rate, m^3/s
Sc = Schmidt number
t = time, s
t_f = total feeding time, s
T_U = intensity of turbulence
TR = dimensionless number
u = axial mean velocity, m/s
u_{bulk} = mean velocity of the bulk flow passing the feed point, m/s
u_{feed} = linear velocity in the feed pipe, m/s
u_{res} = resultant mean velocity passing the feed point, m/s
u_{rms} = root-mean-square of fluctuating velocity in axial direction, m/s
U = ratio of linear feed flow velocity to linear bulk flow velocity
v = radial mean velocity, m/s
v_{rms} = root mean square of fluctuating velocity in radial direction, m/s
V = tank volume, m^3
w = tangential mean velocity, m/s
w = in Table 1 and Figure 2 only: Rushton turbine blade width, m

w_{rms} = root mean square of fluctuating velocity in tangential direction, m/s
X^0_{feed} = volume fraction of feed fluid at center of expanding plume
ε = local energy dissipation rate, W/kg
Λ_γ = integral scale of random concentration fluctuations, m
Λ_u = macroscale length, axial component, m
Λ_v = macroscale length, radial component, m
Λ_w = macroscale length, tangential component, m
Λ_T = macroscale of turbulence, m
Λ_{3D} = 3-D macroscale length, m
ν = kinematic viscosity, m^2/s
ρ = density, kg/m^3
τ_{circ} = circulation time, s
τ_{Corr} = homogenisation time constant of Corrsin (1964), s
τ_{IC} = inertial-convective mesomixing time constant, s
τ_{meso} = characteristic mesomixing time, s
τ_{micro} = micromixing time constant, s
τ_{mix} = characteristic mixing time, s
τ_{TD} = turbulent dispersion mesomixing time constant, s

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