SOLID-LIQUID AND LIQUID-LIQUID MIXING LABORATORY

For Chemical Engineering Undergraduates

Sanaz Barar Pour, Gregory Benoit Norca, Louis Fradette, Robert Legros, Philippe A. Tanguy
Ecole Polytechnique • Stn. Centre-ville, Montreal

In the chemical industries, fluid mixing in stirred tanks is involved in a wide variety of operations such as homogenization of miscible liquids (blending), gas dispersion, mixing of immiscible liquids (emulsification or dispersion), and suspension of solid particles. Emulsification is a process consisting of dispersing droplets of an oil phase into a water phase (O/W) or a water phase in an oil phase (W/O). A surfactant is normally added to stabilize the dispersion.

This operation is the basis for many manufacturing processes in the food, cosmetics, and pharmaceutical industries, to name a few. The suspension of solid particles in agitated vessels is required in many industrial reactors dealing with catalytic reactions, crystallization, leaching, polymerization, dissolution, ion exchange, and adsorption. The typical objectives of solid-liquid suspension are to produce a homogenous slurry (concentration and particle size) and to promote the rate of mass transfer between the solid and liquid phases. In most applications, determining the minimum impeller speed for off-bottom suspension or to disperse a liquid phase in mechanically agitated liquids in stirred vessels has considerable importance.[1]

In order to provide chemical engineering students with practical experience on suspension and emulsification processes, laboratory solid-liquid and liquid-liquid mixing experiments have been developed. The objective is to highlight the main

Sanaz Barar Pour *received her B.S. from the University of Tehran in chemical engineering in 2004. She is currently a master's student at Ecole Polytechnique of Montreal. Her research activity is related to solid-liquid dispersion in highly viscous media.*

Gregory Benoit Norca *received his B.S. from Ecole Polytechnique of Montreal in mechanical engineering. Currently he is following an engineering program in ENSTA, France. His field of interest is mechanical structure design and analysis in automobile, aeronautics, and aerospace.*

Louis Fradette *completed his B.S. and M.S. degrees at Laval University (Quebec City). He holds a Ph.D. from Ecole Polytechnique of Montreal and Institute National Polytechnique de Lorraine, France. He has more than 10 years of process engineering experience in various fields such as petroleum refining, petrochemicals, and software development. Since 2004, he has been working as a research associate at Ecole Polytechnique of Montreal with research activities related to the development of efficient continuous and batch processes for the production of emulsions.*

Robert Legros *is a professor of chemical engineering at Ecole Polytechnique of Montreal. Currently he is the chair of the department. He received his B.S. from Ecole Polytechnique in 1983 and his Ph.D. from the University of Surrey in 1987. His academic research involves solids thermal treatments in fluid beds, modeling of combustion reactor, heat and mass transfer, and hydrodynamics of spouted beds. Some of his current research interests are related to pharmaceutical engineering, namely in powder technology and downstream processes.*

Philippe A. Tanguy *is a professor of chemical engineering at Ecole Polytechnique of Montreal. He received his B.S. in 1976, his doctorate de specialite in 1979 from Universite de Paris, and his Ph.D. in 1982 from Laval University. His research interests are in non-Newtonian fluid mechanics, CFD and process engineering involving complex fluids, in particular coating processes, and agitation and mixing operations. He is currently Director of the Research Unit in Industrial Flow Processes (URPEI).*

aspects governing process efficiency, namely the influence of the operating parameters and the effect of the impeller type. The mixing laboratory is a part of a senior-year unit operations course and a graduate mixing course, both offered by the Department of Chemical Engineering at Ecole Polytechnique of Montreal.[2] The experiments are carried out as a group exercise. Each group consists of a maximum of three students, and must perform the required laboratory work in less than four hours. After finishing the experiments, each group should hand over a full report or prepare an oral presentation the following week. Both report and oral presentation consist of the following sections: description of the experiment objectives, theoretical basis, engineering method used, experimental set-up and operating conditions, experimental data, analysis of the results, and comments or recommendations for improvements.

EXPERIMENTAL SET-UP

The mixing system used in the experiments is a modified Turbotest (VMI Rayneri) laboratory mixer. It consists of a transparent polycarbonate vessel of 165 mm inner diameter and 230 mm height, with an open top fixed to a rigid table for safe operation. Two impellers are tested for liquid-liquid dispersions: a radial-flow impeller (6-blade Rushton turbine) and an axial-flow impeller (marine propeller). In the solid-liquid suspension work, the 6-blade Rushton turbine is compared to a pitched blade turbine (four 45° blades). The impellers are mounted on a rigid shaft driven by a DC motor, in which speed is carefully regulated in a range from 10 to 1,800 rpm by means of a DC controller. The motor is mounted on a rigid structure that can be moved to adjust the vertical position of the impeller. A standard mixing configuration is used as a starting point, with the impeller placed on the vessel centerline at 1/3 of the liquid height in liquid-liquid dispersion and 1/6 of the liquid height in solid-liquid suspension. The agitation torque is measured by a noncontact type torque meter (range between 0.1 and 1.42 N.m) fitted between the motor and the agitation shaft.

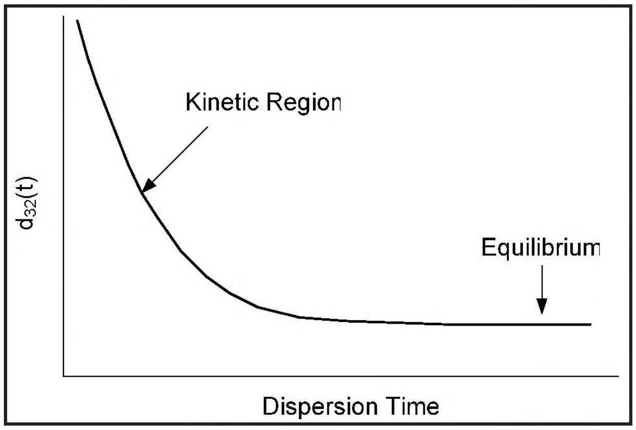

Figure 1. *Droplet size as a function of time.[4]*

LIQUID-LIQUID EMULSION

Theory

The term immiscible liquid-liquid system designates two or more thermodynamically incompatible liquids present as separate phases. Emulsion is defined as a liquid-liquid dispersion stabilized by means of one or more surfactants that allow long-term stability (from days to months depending on the formulation). The term liquid-liquid dispersion is kept for nonstabilized (unstable) systems where the dispersed state is maintained by continuous agitation. Agitators that provide high shear and good pumping capacity are common choices for liquid-liquid dispersion and emulsification.

When the densities between the dispersed phase and the continuous phase are different, and the agitator does not generate a sufficient circulation throughout the vessel, settling and coalescence occur. Consequently, it is important to determine the minimum speed for dispersion of the droplets.

Skelland and Seksaria[3] proposed a dimensionless correlation to predict the minimum speed for liquid-liquid dispersion in two liquids with different densities:

$$\frac{N_{min}D^{0.5}}{g^{0.5}} = C_{20}\left(\frac{T}{D}\right)^{\alpha_5}\left(\frac{\mu_c}{\mu_d}\right)^{1/9}\left(\frac{\Delta\rho}{\rho_c}\right)^{0.25}\left(\frac{\sigma}{D^2\rho_c g}\right)^{0.3} \qquad (1)$$

where D is the diameter of the impeller, g the gravitational acceleration, T the tank diameter, σ the interfacial tension, μ_c and μ_d the viscosity of the continuous phase and the dispersed phase, respectively, and ρ_c and ρ_d the density of the continuous phase and dispersed phase, respectively. In Eq. (1) $\Delta\rho = |\rho_d - \rho_c| \cdot C_{20}$ is a model constant scaling the ease of forming a suspension. Table 12-5 in the *Handbook of Industrial Mixing*[4] gives more information as to the value of these constants for different types of impellers.

Flow regimes are characterized by the value of the Reynolds number, Re, which is the ratio of inertial forces over the viscous forces. The Reynolds number of an agitator in a liquid-liquid system is:

$$Re = \frac{D^2N\bar{\rho}}{\bar{\mu}} \qquad (2)$$

where $\bar{\rho}$ and $\bar{\mu}$ are the density and viscosity of the mixed volume phases. For diluted dispersions ($\varphi < 0.01$), these values are equal to density and viscosity of the continuous phase. The laminar regime corresponds to $Re \le 10$, the transition regime to $10 \le Re \le 10^4$ and the turbulent regime to $Re > 10^4$.

The emulsification time is the time necessary to reach a stable droplet size and droplet size distribution. This time depends on the frequency of droplet passage in the agitator's region or, alternatively, to the circulation time t_c:

$$t_c = \frac{V}{N_q N D^3} \qquad (3)$$

where V is the tank volume, N_q the circulation number, N the rotational speed of the agitator, and D the diameter of the agitator. Circulation number or pumping number, N_q, varies with impeller type and hydrodynamic regime. The typical value of circulation number for Rushton turbine and marine propeller is 0.67 and 0.53, respectively. Figure 1 shows a typical curve of a droplet-size evolution in an emulsification tank.

The droplet mean diameter can be defined by the following equation:

$$d_{mn} = \left[\frac{\sum_i n_i d_i^m}{\sum_i n_i d_i^n} \right]^{1/m-n} \qquad (4)$$

where m= 1,2,3; n= 0,1,2; and m > n.

A common choice for the mean diameter is the Sauter mean diameter (d_{32}), since it is directly related to the interfacial area per unit volume, a_v, which determines the transfer rate of energy, mass, and/or the chemical reaction in the dispersion:

$$a_v = \frac{6\varphi}{d_{32}} \qquad (5)$$

where φ is the volume fraction of dispersed phase.

EXPERIMENT 1

Objective

The objective of this experiment is to determine the influence of the type of agitator and its position on the speed required for complete dispersion in multiphase systems with separate phases. The corresponding power consumption will be obtained. In liquid-liquid dispersion, knowing the minimum agitator speed for complete dispersion, N_{min}, enables us to design the mixer efficiently.

Procedure

In order to reach the objectives the detailed procedure is as follows:

1) Use the configuration shown in Figure 2 with the Rushton turbine.
2) Add 3 L (85%v) water and 0.5 L (15%v) of sunflower oil up to H=165 mm level.
3) Set the rotational speed equal to zero rpm and verify that the torque screen displays zero.

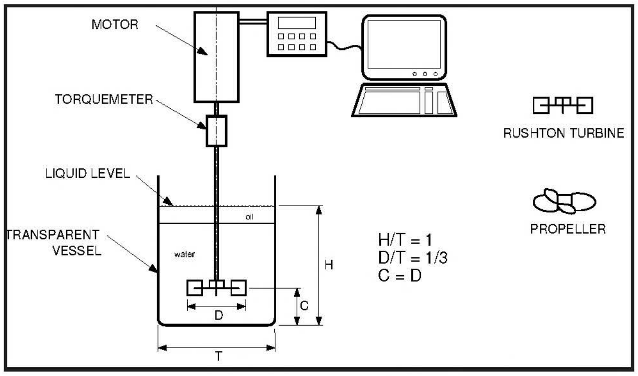

Figure 2. Experimental setup.

4) Gradually increase the rotational speed of the agitator up to the point where no continuous layer of the dispersed phase remains in the vessel. Write down the torque and the rotational speed and then stop the agitator. Calculate the power consumption by using the following expression:

$$P = 2 \cdot \pi \cdot N \cdot M_c \qquad (6)$$

where P is the power consumption, N is the rotational speed of the agitator, and M_c is the corrected torque.

The shaft guiding system induces a residual torque due to friction; hence, the torque value must be corrected by subtracting the residual torque from the measured torque for both impellers:

$$M_c = M_m - M_r \qquad (7)$$

where M_m and M_r are measured and residual torques, respectively.

5) Wait 3–4 minutes to allow the phases to separate. Place the Rushton turbine at position C/H = 0.5 and repeat from step 4.
6) Repeat the experiment with the marine propeller.

In each case determine the flow regime. Use the following values for viscosity and density: $\overline{\mu} = 0.008$ Pa.s and $\overline{\rho} = 985$ kg/m^3. What is the influence of the vertical position of the agitator on the minimum speed for dispersion and power consumption? What is the best position for the agitator in this setup? Which agitator would you use for liquid-liquid dispersion? Why?

The complete dispersion speed, N_{jd}, is determined in each case. The Rushton turbine is more efficient than the helical marine impeller in terms of power consumption. For the Rushton turbine, dispersion was not obtained in laminar regime in both cases. For the helical marine impeller, dispersion was obtained in turbulent regime where C/H=1/3 and in transitional regime where C/H=1/2. Hence, by placing the agitator at C/H=1/2, the complete dispersion speed and power consumption are decreased by more than 50% in both cases. The best position of the agitator would be the interface of the two phases. For this application of liquid-liquid dispersion, the Rushton turbine is preferred.

EXPERIMENT 2

Objective

The objective of this experiment is to determine the effect of mixing duration and rotational speed of agitator on the particle size and particle size distribution. Particle size distribution is one of the most important characteristics of liquid-liquid dispersion. A laser granulometer will be used to measure the particle size of the droplets.

Procedure

1) Place the Rushton turbine on the shaft as in Experiment 1.
2) Add water up to H=165 mm level.

3) Add 7 mL of surfactant in order to stabilize the emulsion.

4) Start the agitator in order to dissolve the surfactant completely in the water. Set the agitator at 225 rpm, add 35 mL of sunflower oil, and start the chronometer immediately.

5) Take samples at time = 1, 4, 7, 10, 13, 16, and 20 minutes or beyond and measure the particle size of the droplets by means of granulometer until the diameter, d_{32}, becomes constant. The samples can be taken by a syringe from the same location in the vessel. Typical experimental results are shown in Table 1.

6) Repeat the same procedure for 300 and 375 rpm.

7) Save the files in the Excel format (.xls) according to the procedure written in the user manual of the granulometer. These files will be used in order to investigate the droplet size distribution.

Plot the droplet size distribution in terms of volume frequency (vol% vs. droplet size distribution) at 1, 4, 7, and 13 minutes for the first speed (225 RPM). What do you observe?

An example of typical plots is presented on Figure 3.

Plot the Sauter diameter, d_{32}, as a function of time for each speed. How does the droplet size change with time? How does it change with the rotational speed? How does the equilibrium time change with the rotational speed?

TABLE 1

Time (min)	d32 (μm)
1	30.09
4	24.50
7	22.71
10	14.98
13	13.05
16	11.04
20	11.04

An example of typical curves is shown in Figure 4. This figure shows that the droplet size is decreasing with time. Increasing the rotational speed, however, will reduce droplet size and equilibrium time.

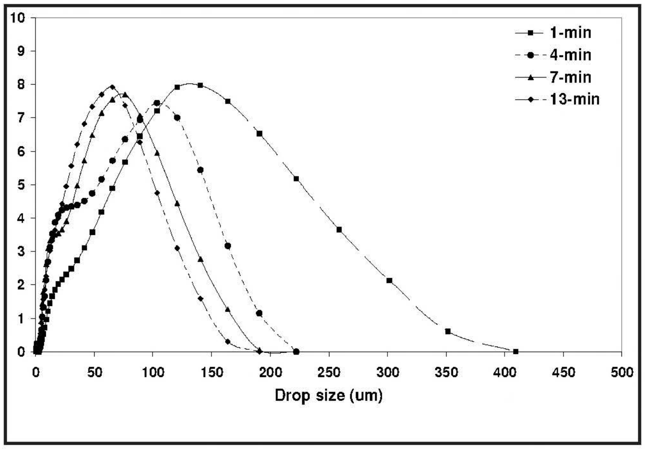

Figure 3. Particle size distribution obtained at 1, 4, 7, and 13 minutes.

SOLID-LIQUID SUSPENSION

Theory

A suspension is a dispersion of particulate solids in a continuous liquid phase that is sufficiently fluid to be circulated by a mixing device.[5] The systems resulting from incorporating powders and fine particles throughout the liquid medium shall be considered as dispersion or colloidal suspensions.[6] In agitated vessels, the degree of solid suspension can be classified as follows:[4]

- *On-bottom motion*
- *Complete off-bottom suspension*
- *Uniform suspension*

The required level of suspension depends on the desired process result and the unit operation involved. For example, a high degree of suspension is required for crystallization or slurrying, whereas a lower degree of suspension is usually sufficient for the dissolution of a highly soluble solid.

The state of suspension known as complete off-bottom suspension is characterized by complete motion, with no particle remaining at rest on the vessel bottom for more than one or two seconds. This criterion is known as the Zwietering[7] criterion. Operation at the minimum impeller speed for a just-complete suspension condition is adequate for many reaction or mass transfer processes, and much of the study on the solid-liquid mixing is concerned with the measurement and correlations of N_{js} values.

According to Zwietering,[7] the following correlation can be used to estimate the just suspended impeller speed, N_{js}:

$$N_{js} = S\upsilon^{0.1}\left[\frac{g_c(\rho_s - \rho_l)}{\rho_l}\right]^{0.45} X^{0.13} d_p^{0.2} D^{-0.85} \qquad (8)$$

where

$$S = Re_{imp}^{\;0.1}\, Fr^{0.45} \left(\frac{D}{d_p}\right)^{0.2} X^{0.13} \qquad (9)$$

$Re_{imp} = \frac{N_{js}D^2}{\upsilon}$ is the impeller Reynolds number, the Froude number is $Fr = \frac{\rho_l N_{js}^2 D}{(\rho_s - \rho_l)g}$, D is the impeller diameter, d_p is the mass-mean particle diameter, X the mass ratio of suspended solids to liquid, υ is the kinematic viscosity of the liquid, g is the gravitational acceleration, ρ_s and ρ_l are the density of particle and the density of liquid, respectively.

Objective

The objective of this experiment is to determine the just-suspension impeller speed, N_{js}, and the value of S for a radial and an axial impeller, namely a Rushton turbine and a pitched-blade turbine. In most of the chemical process industries it is essential to provide complete off-bottom suspension.

Below this speed the total solid-liquid interfacial area is not completely and efficiently used for mass transfer. Above this speed the mass transfer rate increases slowly and the power dissipated increases considerably. Therefore, it is important to determine the just-suspension impeller speed, N_{js}.[8]

Experiment

1) Place the Rushton turbine on the shaft as before.
2) Add water into the tank to H level with H=T.
3) Set the impeller at 1/6 of the liquid height C/T=1/6. It is necessary to keep this bottom clearance constant in all the experiments.
4) According to the given concentration of 1,000 μm glass beads, weigh the exact amount of glass beads by means of a balance and then add them to the tank.
5) Set the rotational speed equal to zero rpm and verify that the torque screen displays zero.
6) Increase the rotational speed of the impeller until you reach the Zwietering visual criterion for just suspension. Note the just suspended speed.
7) With this concentration of solids, proceed with the change of impellers and repeat step 6.
8) Increase the amount of glass beads to the next desired concentration and repeat steps 5-7 for the new concentration. Note N_{js} for both impellers at the new concentration.
9) Repeat step 5-8 for two different diameters of glass beads.
10) Prepare a PEG/water fluid mixture with 0.02 Pa.s of viscosity. Use Figure 5 to estimate the amount of PEG (Polyethylene Glycol, grade 35000) to be mixed with water until you reach H = T. After dissolving the PEG, wait two minutes until a clear fluid is obtained. (PEG increases the viscosity of the solution without having considerable effect on the density).
11) Repeat steps 5-8 with this fluid for both impellers and glass beads with 3000 μm of diameter.

With the experimental measurements and Figure 5, fill in Table 2 and answer the following questions:

a) Determine the value of S for both impellers from the first experiment when the fluid was water. What is the value of S for each impeller?

b) For the second experiment with higher viscosity fluid, replace the value of S that you have obtained in the Zwietering correlation to compute N_{js}. Compare just sus-

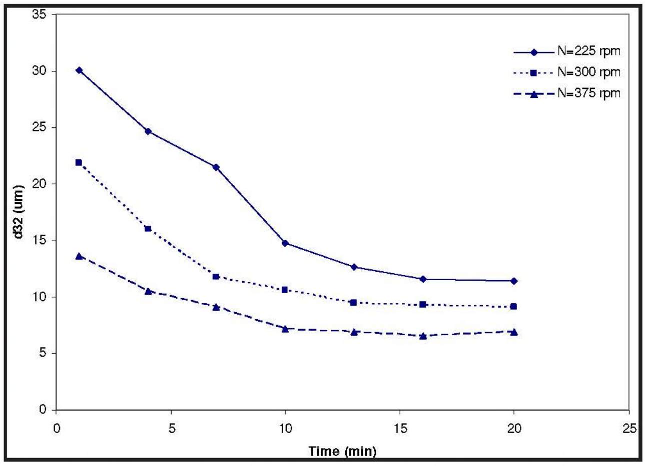

***Figure 4**. Sauter diameter and equilibrium time as a function of rotational speed for Rushton turbine.*

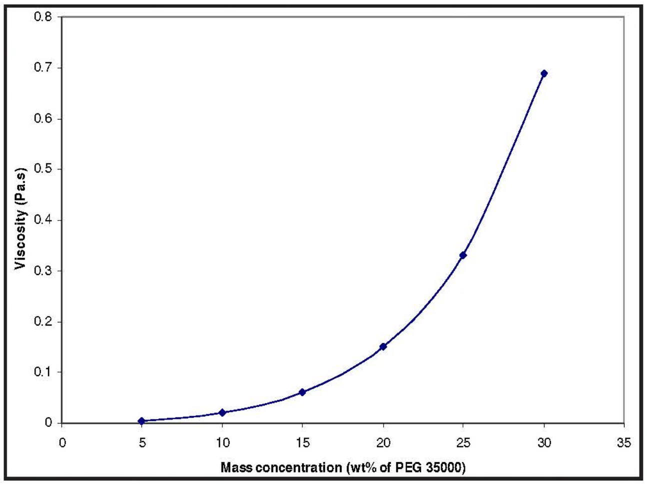

***Figure 5**. Viscosity vs. mass concentration of PEG (grade 35000).*

TABLE 2

Continuous phase	d_p (μm)	X	N_{js}(Hz)	N_{js} (rps)	S
Water $\mu = 10^{-3}$ Pa.s	1000	0.5	22.9	10.81	4.75
	1000	1.5	25.9	12.32	4.66
	1000	5	32.3	15.25	4.97
	d_p (μm)	X	N_{js}(Hz)	N_{js} (rps)	S
	3000	0.5	26.6	12.56	4.43
	3000	1.5	33.1	15.63	4.47
	3000	5	37.3	16.61	4.61
Water + PEG $\mu = 0.02$ Pa.s	d_p (μm)	X	N_{js} (Hz)	Experimental N_{js} (rps)	Calculated N_{js} (rps)
	3000	0.5	34.4	16.24	16.57
	3000	1.5	39.2	18.51	19.11
	3000	5	43.9	20.73	21.35

pension speed from the correlation with what you got from the experimental value.

c) Draw $N_{js} / (\rho_s - \rho_l)^{0.45}$ vs. d_p (μm) for two different impellers and both fluids. In each curve, how can you estimate the just-suspension impeller speed for the glass beads with diameter 2000 μm?

Which of these impellers would you use for suspension? Why? See Figure 6

The pitched-blade turbine is preferred in solid-liquid suspension application. Generally, axial flow impellers are more efficient than radial flow impellers to suspend the solids.

CONCLUSION

As mentioned, two-phase mixing is involved in numerous industrial processes, hence, a general knowledge of these operations is essential for the chemical engineer. These experiments are designed to allow students to become acquainted with basic understanding of important parameters that affect liquid-liquid and solid-liquid mixing. Analyzing their results in a critical manner, and answering specific questions, will help the students to investigate the behavior of different impellers in different mixing situations. All this should provide students with criteria to choose the best mixing system for a given application.

NOMENCLATURE

a_v	interfacial area per unit volume [m^{-1}]
C_{20}	coefficient of variation [dimensionless]
d_{32}	Sauter mean drop diameter, general use [m]
d_i	nominal diameter of drops in size class i [m]
d_{mn}	droplet mean diameter [m]
d_p	particle size or diameter [m]
D	impeller diameter [m]
Fr	Froude number [–]
g	gravitational acceleration [m/s^2]
m	number of size classes representing drop size distribution
M_c	corrected torque [N.m]
M_m	measured torque [N.m]
M_r	residual torque [N.m]
n_i	number of drops in size class i
N	impeller speed [rps]
N_{js}	minimum impeller speed to just suspended solid particles in vessel [rps]
N_{min}	minimum impeller speed to suspend liquid drops in vessel [rps]
N_q	flow number
P	power [W]
Re	Reynolds number
Re_{imp}	impeller Reynolds number
S	Zwietering constant
t_c	contact time between two colliding drops [s]
T	tank diameter [m]
V	volume of tank [m^3]
X	mass ratio of suspended solids to liquid [kg solid/100 kg liquid]

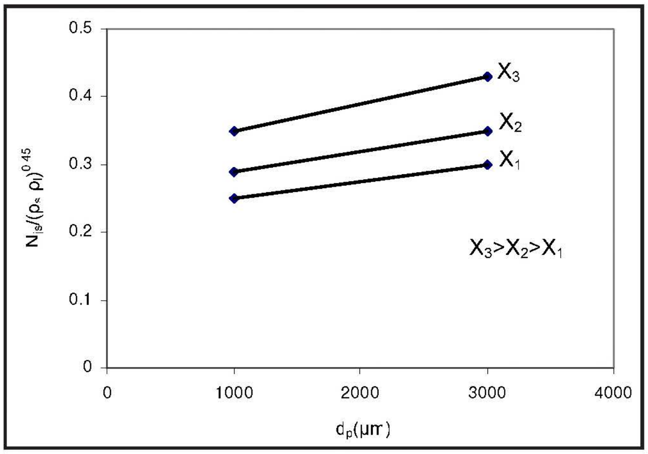

***Figure 6**. $N_{js} / (\rho_s - \rho_l)^{0.45}$ vs. d_p as a function of concentration of solid particles.*

Greek symbols

μ_c	viscosity of continuous phase [Pa.s]
μ_d	viscosity of dispersed phase [Pa.s]
$\bar{\mu}$	bulk viscosity of liquid-liquid mixture [Pa.s]
υ	kinematic viscosity of the liquid [m^2/s]
ρ_c	density of continuous phase [kg/m^3]
ρ_l	density of liquid [kg/m^3]
ρ_s	density of solid or particle [kg/m^3]
$\bar{\rho}$	bulk density of liquid-liquid mixture [kg/m^3]
$\Delta\rho = \lvert\rho_d - \rho_c\rvert$	density difference between phases [kg/m^3]
σ	interfacial tension [N/m]
φ	volume fraction of dispersed phase

REFERENCES

1. Armenante, P.M., Y.T. Huang, and A.T. Li, "Determination of the Minimum Agitation Speed to Attain the Just Dispersed State in Solid-Liquid and Liquid-Liquid Reactors Provided With Multiple Impellers," *Chem. Eng. Sci.*, **47**(9-11), 2865 (1992)
2. Ascanio, G., R. Legros, and P.A. Tanguy, "A Fluid Mixing Laboratory for Chemical Engineering Undergraduates," *Chem. Eng. Ed.*, **37**(4), 296 (2003)
3. Skelland, A.H., and R. Seksaria, "Minimum Impeller Speed for Liquid-Liquid Dispersion in Baffled Vessels," *Ind. Eng. Chem. Process Des. Dev.*, **17**, 56 (1978)
4. Paul, E.L., V.A. Atiemo-Obeng, and A.M. Kresta, *Handbook of Industrial Mixing*, John Wiley & Sons, Inc. (2004)
5. Uhl, V.W., and J.B. Gray, *Mixing Theory and Practice, Vol. 1 and 2*, Academic Press, London (1966)
6. Harnby, N., M.F. Edwards, and A.W. Nienow, *Mixing in the Process Industries*, Antony Rowe, Great Britain (1985)
7. Zwietering, T.N., "Suspending of Solid Particles in Liquid by Agitators," *Chem. Eng. Sci.*, **8**(3-4), 244 (1958)
8. Baldi, G., R. Conti, and E. Alaria, "Complete Suspension for Particles in Mechanically Agitated Vessels," *Chem. Eng. Sci.*, **33**(1), 21 (1978) ❐