Turbulent Dispersion of Aqueous Drops in Organic Liquids

Drops sizes for turbulent dispersion of aqueous solutions in organic liquids were determined and correlated for three types of dispersion devices: small-diameter tubes, motionless mixer units, and a rotating annular disperser with Couette flow. Empirical correlation based on dimensional analyses were useful for application of these devices to gel-sphere processes. The results support the Kolmorogoff theory of turbulence as a dispersion mechanism for a range of 10^4 for the rate of energy dissipation.

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Introduction

Three different devices using controlled velocities of organic liquids were applied to disperse aqueous solutions as drops. One consisted of simple tubes of small diameters. A second contained motionless mixer units inside larger tubes. The third employed Couette flow of the organic liquid between a cylindrical rotor and a stationary cylinder. These processes were developed to prepare nuclear fuels as part of the consolidated fuel reprocessing program at Oak Ridge National Laboratory (ORNL).

These devices were applied to gel-sphere processes in which the liquid drops are converted into solid gel spheres of hydrated metal oxides. The gel-sphere products are strong spheres and allow accurate measurement of the sphere and the drop size distributions. The drop diameters must be controlled and predictable to allow preparation of product spheres of the desired sizes. Empirical correlations were determined for application to the gel-sphere processes.

The theory of turbulent dispersion based on eddy velocities has been developed by Kolmorogoff (1949), Hinze (1955), and others. Davies (1985) reviewed this theory and the agreement of theory with four types of dispersion devices for energy dissipation rates of 6 to 400,000 W/g. The gel-sphere results for drop size distributions are for energy dissipation rates of 10^{-3} to 1.5 W/g. These combined results support the theory of turbulence as the dispersion mechanism over a range of 10^9 for the rate of energy dissipation.

Gel-Sphere Processes and Role of Drop Formation

A limited description of gel-sphere processes is given here to explain the drop formation conditions and results. Some detailed descriptions of chemical flowsheet conditions and equipment and procedures have been published. Depending on the mechanism of gelation, the drops must be dispersed into trichloroethylene (TCE), 2-ethyl-1-hexanol (2EH), silicone oil, NH_3 gas or solution, or other fluids. These gel-sphere processes have been described by Haas et al. (1967, 1980), Haas and Clinton (1966), Bischoff et al. (1974), and Zimmer et al. (1978).

The preparation of gel spheres by processes developed at ORNL and elsewhere is based on the conversion of liquid drops into solid gel spheres of hydrated metal oxides. Each liquid or broth drop must contain the amount of metal needed in one product sphere. (Depending on the gel-sphere process, the feed liquid may contain colloidal oxides, metal salts, organic polymers, metals in complexed or hydrolyzed form, or other chemicals. This feed to the gelation step may be termed a "broth.") The diameters of the drops determine the diameters of the product spheres. The solidification of the drop may result from mass transfer of NH_3, NH_4OH, HNO_3, H_2O, or from chemical reactions inside the drop. Mass transfer from the drop also occurs during washing to remove solutes, drying to remove H_2O and other volatile constituents, and thermal treatments. The product spheres are generally much smaller than the liquid drops. There is no significant loss of metal during these process operations, and the metal concentrations in the liquid drop and the product sphere are accurately measured; therefore, the diameter ratio of liquid drop to product sphere is accurately known.

The gel-sphere products are usually excellent, strong spheres and allow a convenient and an accurate measurement of the liquid drop sizes. The complete batch of product can be used as a sample, or procedures that ensure representative sampling can be used. In this way, many of the uncertainties involved in measuring liquid drop diameters in a second liquid are avoided. The possibility of coalescence or of an additional secondary dispersion before solidification occurs must be considered.

Some of the results reported in this paper were obtained from many hours of operation to prepare test batches of nuclear reactor fuels. These results demonstrated good reproducibility of the drop formation procedures. All the turbulent disperser results were for dispersion of aqueous drops into organic liquids; the gelation procedures using aqueous NH_4OH solutions were not practical for small gel spheres. Controlled and uniform product diameters are important to many applications of gel-sphere processes. Procedures that gave excellent size uniformity for drops of 500 to 5,000 μm (Haas and Fowler, 1984) were not practical for smaller drops. Simple mixing using agitators at controlled speeds gave control of the average diameters, but the uniformity of drop size was poor. The turbulence produced by an agitator is obviously both nonuniform and nonisotropic. The results in this paper are for three dispersers, which were expected to provide a more uniform turbulence than that from agitators.

Theory of Turbulent Dispersion Mechanism

The dispersion of one liquid into another as a result of turbulence has been discussed by a large number of authors. A theory of homogeneous, isotropic turbulence was developed by Kolmogorov (1949). The dispersion results from the turbulent fluctuations or eddies, and the fluctuation velocities are determined by the rate of energy dissipation. The pressure from the fluctuation velocities is opposed by the surface tension holding the drop together. The maximum drop size that is stable with respect to further dispersion is given by

$$d_{\max} = C_2 \left(\frac{\gamma}{\rho}\right)^{0.6} P^{-0.4} \qquad (1)$$

where the constant, C_2, is dimensionless and has a value near one. The viscosity of the liquid drop was neglected for the theory used to derive Eq. 1 and the viscous forces inside the drop can also contribute to the resistance to breakup. Davies (1985) proposed adding a viscous force term to the interfacial tension as follows:

$$d_{\max} = C_2 \left(\gamma + \frac{\mu_d v}{4}\right)^{0.6} \rho^{-0.6} P^{-0.4} \qquad (2)$$

Substitution of equations for P in Eq. 1 can give many of the relationships in the literature for $d_{\max}$ vs. Weber number (We), Reynolds number (Re), and velocities or RPM. The interfacial tension would not normally appear in P, and an almost universal result is

$$d_{\max} \propto (We)^{-0.6} \qquad (3)$$

Davies (1972) has given a more complete discussion of isotropic turbulence and a comparison of the theory of turbulent dispersion with experimental data.

Three Turbulent Dispersion Devices

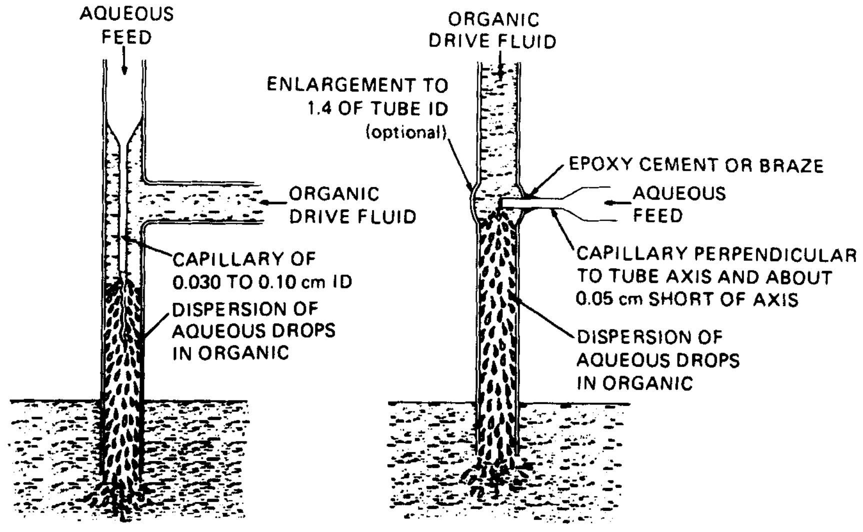
The theory of turbulent dispersion mechanism and the gel-sphere process requirements indicated a need for a controlled and uniform turbulence. Also, the flow through the turbulent zone should be positive and the holdup time limited so that the dispersed drops can solidify or gel without continued exposure to the turbulence. Dispersion devices with these attributes should give improved uniformity of drop size distribution as compared to previous results with dispersion by an agitator. The dispersion by an agitator also gave some dumbbell or clumped spheres, which were believed to result from recycle of partly solidified spheres through the agitator. The three dispersion devices tested in this study were selected from these considerations.

An open tube provides a well-defined flow regime and is the simplest of the three types of dispersers studied. The tube is characterized by a single dimension, the diameter, D. The system Weber number and the Reynolds number are unambiguous. The aqueous phase to be formed into drops was introduced through capillaries at velocities that were usually within a factor of 4 of the organic velocity, and the diameter of this capillary was not a significant variable. The first tests were done with glass units, but later tests were more commonly performed with hypodermic needles brazed into stainless steel tubing. Typical units of both types are diagramed in Figure 1.

The motionless or static mixers, the second kind of disperser used in these studies, were 0.335 to 0.80 cm ID tubes containing 21 mixing elements (Kenics Corporation, North Andover, MA, model series 37). These Kenics mixers have 180° helical elements of alternating right- and lefthand twists, with each element length equal to 1.5 dia., Figure 2. The elements divide the tube into two semicircular flow channels. The alternating twist elements are joined 90° out of phase so that each flow from one element is split between the two channels of the next element.

To make empirical correlations, these motionless mixers were characterized by the ID, and the average velocities were calculated using these ID's. The empirical constants should not be used for other static mixers with mixing elements of different proportions. The true velocities would be much higher than the average velocities calculated using the ID because the mixing elements both reduce the cross section for flow and increase the path length. The hydraulic diameters of the flow channels would be smaller than the ID. The void volumes inside the tube were measured by water displacement and were from two-thirds to five-sixths of the volume calculated using the nominal ID. Calculations from the weights of the dry units also indicated that the mixing element occupied one-third to one-sixth of the tube volume. This high-volume fraction of mixing elements agrees with the visual appearance of these small units and would probably not apply to larger units.

The third dispersion device may be called a Couette flow or annular disperser. A shear field can be generated in the annulus between two cylinders rotating at different speeds. The flow patterns for such a thin annulus are termed Couette flow. This type of disperser was tested as a possible means of preparing UO_3 spheres via internal gelation by using a motor-driven rotor mounted inside a stationary cylinder, Figure 3. The bottom of the rotor was provided with an enlarged diameter to pump fluid by centrifugal action. The first test unit had conical surfaces for the entire rotor and stator at a 1.5° angle. Thus, the annulus thickness could be varied by simply moving the stator up or down with respect to the rotor. However, this arrangement gave an excessive pumping rate of the continuous phase as well as a variable shear field along the annulus. Experimental data from the conical unit were used to evaluate the effect of annulus thickness, but an annulus of uniform diameter (no taper) appeared to provide better control of drop diameter. The hydraulic diameter of the flow channel, D, would be twice the



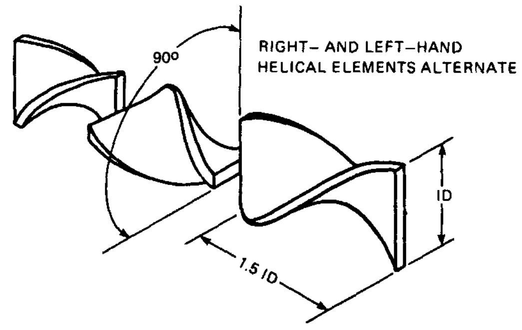
a. CONCENTRIC AQUEOUS FEED CAPILLARY (material: glass)

b. RIGHT ANGLE AQUEOUS FEED CAPILLARY (preferred for turbulent operation)

Figure 1. Two types of two-fluid nozzles.

annulus thickness for the Couette disperser. The other dimension, ID, was taken as the rotor diameter. In addition to the D/ID term, the rotor diameter is used to calculate the velocity, $V = \pi(ID)$ (RPM/60).

For all the gel-sphere tests of turbulent disperser devices, the fraction of dispersed phase and the size of the aqueous feed inlet did not have any significant effects on the drop diameters. The ratios of continuous or organic phase flow rates to the aqueous flow rates were usually from 40:1 to 100:1, with a few tests with ratios of 12:1 to 40:1. These high ratios were necessary to provide the water extraction capacity for external gelation or to provide heat capacity for internal gelation. The aqueous velocities in the feed inlets were commonly from 0.25 to 1 times the organic velocities for the simple tube and static mixer dispersers, and were much lower for the aqueous feed inlet to the Couette disperser. The observed drop sizes were always smaller (usually much smaller) than the ID of the aqueous feed inlet and were therefore not determined by this diameter.



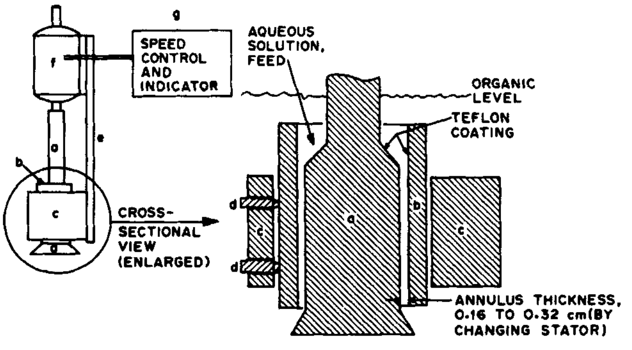
MODEL NO.	ID,cm	L=31.51D cm	NUMBER OF ELEMENTS	VOID VOLUME OPEN TUBE VOLUME
37-03-062	0.335	10.6	21	2/3
37-04-065	0.493	15.5	21	...
37-06-109	0.80	25.5	21	5/6

Figure 2. Kenics static mixers for this study.

Determination and Representation of Drop Size Distributions

Three different areas of size information and calculation are important to these gel-sphere results. These are the size measurements for the product spheres, the calculation of drop sizes from the product sizes, and the representation of size data for correlation.

The principle size measurements for the dry product spheres were by standard test sieves and procedures (U.S. Sieve Series). Large batches of spheres for nuclear fuel samples were blended and sampled by riffling procedures. As compared to materials of irregular shape, the spheres were free-flowing and allowed good size separations. For some compositions, the smallest spheres showed some tendency to be present as small clusters or to be



a. ROTOR, STAINLESS STEEL WITH TEFLON COATING, 3.18-cm OD
b. STATOR, STAINLESS STEEL WITH TEFLON COATING ON ID
c. SUPPORT BLOCK (BOLTED TO MOTOR BRACKET)
d. SET SCREWS TO CENTER STATOR; THREE SETS OF TWO
e. MOTOR BRACKET
f. SERVOMOTOR
g. MOTOR SPEED CONTROL

COUETTE DISPERSER APPARATUS

Figure 3. Couette disperser apparatus.

weakly attached to a larger sphere. The significance of such fines on a weight basis is small.

The known concentrations of metal in the liquid before gelation and in the product sphere allow a simple and accurate calculation of diameter ratios or shrinkage factor, *SF*.

$$SF = \frac{\text{diameter of drop}}{\text{diameter of product}}$$

$$= \left(\frac{\text{concentration of product}}{\text{concentration of liquid feed}}\right)^{1/3} \quad (4)$$

This equation is a simple material balance for thorium, uranium, or other metals that remain in the sphere without significant losses. The products were usually oxides (ThO_2, UO_3, or UO_2) of high purity so that the product concentration was determined by measuring the sphere density. For a single-size fraction of U.S. Sieve Series, the spheres poured into a graduate slowly and tapped lightly would pack to 38% void volume, and the sphere density would be: (bulk density)/0.62. The ThO_2 gel spheres can be sintered to the theoretical density (10 g/cm^3) of ThO_2. An example of a shrinkage calculation for dense ThO_2 is

Feed concentration: 2.50 *M* Th

$$\text{Product concentration: } \frac{(10)(1{,}000)}{264} = 37.88\ M\,\text{Th}$$

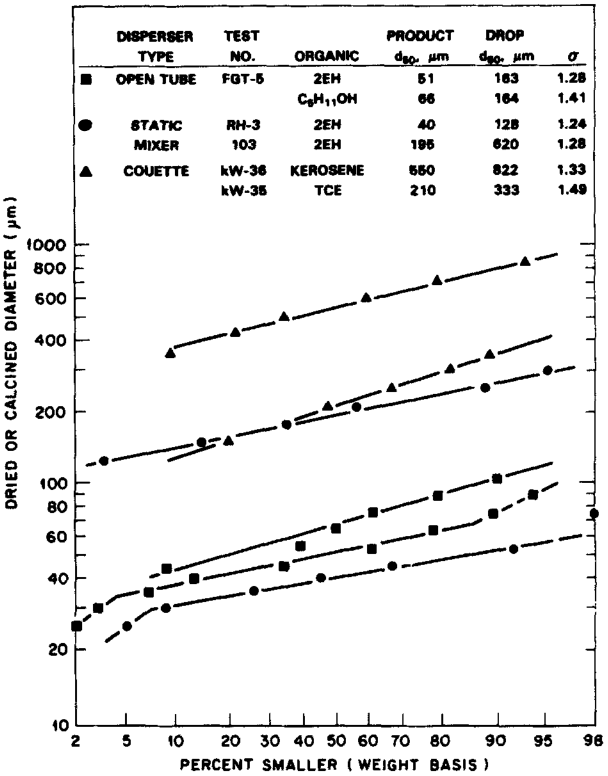
$$SF = \frac{\text{diameter of drop}}{\text{diameter of product}} = \left(\frac{37.88}{2.50}\right)^{1/3} = 2.47$$

The microstructure and density are important for nuclear fuels and were found to be uniform and independent of sphere size.

For gel-sphere products, weight or mass average diameter and the yield within a range of diameters are pertinent. The exact distribution of very large or fine sizes is not important. A log normal distribution is mathematically convenient to use. The use of log probability paper provides a simple test of the suitability of the log normal fit and permits a simple determination of the mean and standard deviation. The mean of the drop sizes on a weight or mass basis is the d from the log probability plot times the shrinkage factor *SF*. The standard deviations, $\sigma = d_{84}/d$, for log normal distributions are the same for the product spheres and the drops.

Typical examples are shown in Figure 4 of sieve analyses of product spheres for two tests each of the three types of dispersers. The d and σ for the product are determined from these curves. The average drop diameters are calculated using the shrinkage factor, *SF*, from concentrations as previously described.

Size distributions for the three types of dispersers were compared, as were some literature size distributions using the σ's for log normal distributions. Nearly all of the σ's were from 1.21 to 1.50, with 1.28 to 1.35 as the most typical values. While clear quantitative conclusions are uncertain, the following qualitative conclusions were indicated. It appears that the smallest σ values or best uniformity for this type of dispersion (an equilibrium drop size distribution in a uniform turbulence) are $\sigma = 1.28$; lower values are not reproducible. Conditions that do not achieve an equilibrium size distribution or a uniformity of turbulence give larger σ values. The static mixers provide the best uniformity (lowest σ values), Figure 4. Equally good results can be obtained with the Couette disperser, but conditions that include short exposure times (i.e., high continuous-phase pumping rates) yield larger σ values. The open-tube data show less uniformity of size, with σ values ranging from 1.3 to 1.4, Figure 4.



	DISPERSER TYPE	TEST NO.	ORGANIC	PRODUCT d_{50}, µm	DROP d_{50}, µm	σ
■	OPEN TUBE	FGT-5	2EH	51	163	1.28
			$C_6H_{11}OH$	66	164	1.41
●	STATIC	RH-3	2EH	40	128	1.24
	MIXER	103	2EH	195	620	1.28
▲	COUETTE	kW-36	KEROSENE	550	822	1.33
		kW-35	TCE	210	333	1.49

Figure 4. Size distributions for oxide spheres with drop formation by several dispersers.

Empirical Correlation of Results

Correlations of drop-formation data were needed for convenient operation of gel-sphere processes. The empirical correlations were based on a dimensional analysis with an exponential relationship. Dimensionless ratios of two densities and of a density to a density difference appeared in the dimensional analyses results, but either these ratios were not significant or the data were not able to show their effect. With these density ratios omitted, the following three arrangements of the results were used:

$$\frac{d}{D} = C_1\left(\frac{D\rho V^2}{\gamma}\right)^a\left(\frac{D\rho V}{\mu_c}\right)^b\left(\frac{\mu_d}{\mu_c}\right)^c\left(\frac{D}{ID}\right)^g \quad (5)$$

$$\frac{d}{ID} = C_1\left(\frac{ID\rho V^2}{\gamma}\right)^a\left(\frac{ID\rho V}{\mu_c}\right)^b\left(\frac{\mu_d}{\mu_c}\right)^c\left(\frac{D}{ID}\right)^{g+1+a+b} \quad (6)$$

$$d = C_1\left(V^{a+b}\gamma^{-a}D^{1+a+b+g}\,ID^{-g}\mu_c^{-b-c}\mu_d^c\rho^{a+b}\right) \quad (7)$$

The dimensionless numbers with the exponents a and b are, respectively, the system Weber numbers and the Reynolds numbers. The first arrangement, Eq. 5, was applied to the simple tube and to the Couette disperser. The second arrangement, Eq.

6, was used for the static mixer to indicate that the correlation uses the tube ID to calculate average velocities and as the diameters in the Weber and Reynolds numbers. The third arrangement, Eq. 7, indicates a logical procedure for examining the fit and determining exponents. Table 1 shows the range of experimental parameters and results.

The first test of the experimental data is to plot log d vs. log V for data with d and V as the only variables (one disperser and a single pair of fluids, Figure 5). All of the well-defined lines were well represented by straight lines with slopes of -1.4 to -1.6. These included several dispersers and several fluid pairs for both the open tube and the couette disperser. Values that were outside the range -1.4 to -1.6 were either for a narrow range of velocities or were very dependent on a single extreme data point. Allowing for these limitations, all the data could be adequately represented by slopes of $2a + b = -1.5$. Using this value for $2a + b$, log $dV^{1.5}$ was plotted vs. log D or log ID with other variables constant (i.e., data for the same pair of fluids in dispersers of different diameters). For the simple tube with d as the only diameter variable, the slopes were $2.5 - a = 3.15$ or $a = -0.65$. This gives $b = -0.2$. The above procedure has determined a coefficient for the Weber number, $a = -0.65$, without use of the surface tension or viscosity data. Most of the differences between fluids are accounted for by the effects of surface tension, but high viscosities also have significant effects. The largest viscosity differences indicate values of $c = 0.4$ or 0.5, while the smaller differences are indeterminant. The dependence of d on ID for the Couette disperser was used to estimate the exponent, g. For the static mixer, the D/ID term was combined with C_1 as an empirical coefficient. The density of the continuous phase appears in both Weber and Reynolds numbers. The effects of the discontinuous phase density in terms of density ratios or a density difference ratio were not detectable in our data, and these terms were dropped from Eqs. 5, 6, and 7.

Table 1. Range of Experimental Parameters and Results

Parameter	Units	Range of Values for Simple Tube	Kenics Mixer	Couette Disperser
C_2d/d_{max}	—	0.452–1.383	0.401–0.860	0.325–1.456
d	μm	25–636	40–640	174–1,455
D	cm	0.15–0.50	—	0.316–0.676
$F + G$	cm^3/min	220–1,320	130–986	—
F/G	—	0.006–0.08	0.02–0.08	—
ID	cm	—	0.335–0.794	2.7–3.2
L	cm	7–20	15–28	—
P	10^6cm^2/s^3	0.0559–15.0	0.0042–4.441	0.0226–3.52
RPM	min^{-1}	—	—	1,200–3,750
Re	—	442–25,600	184–8,090*	58–46,000
V	cm/s	106–500	—	
We	—	156–7,350	5.0–236*	367–10,700
γ	dyne/cm	2–21	9–10	9–34
ρ	g/cm^3	0.79–1.38	0.79–1.38	0.79–1.38
μ_c	cp(mPa · s)	0.37–5.7	0.45–2.0	0.45–70
μ_d	cp(mPa · s)	4–10	6	6
No. of data points		51	24	39

**Re* and *We* for Kenics mixers are calculated using *ID* and are not compareable (much lower) than true values if values of *D* were known and used.

The final empirical correlations are as follows:

1. The simple tubes or turbulent two-fluid nozzles with TCE, 2EH, and isoamyl alcohol (iAA) gave

$$\frac{d}{D} = 15(We)^{-0.65}(Re)^{-0.2}\left(\frac{\mu_d}{\mu_c}\right)^{0.5}. \tag{8}$$

2. The static mixers with TCE and 2EH gave

$$\frac{d}{ID} = 1.2\,(We)^{-0.65}(Re)^{-0.2}\left(\frac{\mu_d}{\mu_c}\right)^{0.5}. \tag{9}$$

3. The Couette disperser with a single-rotor diameter and four organic liquids (TCE, 2EH, Varsol [hydrocarbon], and a silicone oil) gave

$$\frac{d}{D} = 150(We)^{-0.65}(Re)^{-0.2}\left(\frac{\mu_d}{\mu_c}\right)^{0.5}\left(\frac{D}{ID}\right)^{0.5}. \tag{10}$$

These empirical correlations in the form of Eq. 7 were used for the preparation of nuclear fuel spheres. The effects on d of changes in disperser diameters, flow rates, or Couette disperser rotational speed were easily and dependably predicted by the correlations.

For operation with a new pair of process fluids, it was usually more convenient to determine the effect of the fluid properties from a brief period of test operation instead of waiting for laboratory measurements. The drop diameter of the test batch was measured, and the disperser flow rate or RPM was changed according to the empirical correlation to adjust the drop diameter as desired.

Several comparisons can be made concerning the consistency of the empirical correlations. The calculated values of d show good agreement with the experimental values (see supplementary material). The relationship of d proportional to $V^{-1.5}$ can be checked easily for many different sets of literature data. While correlations of d with $We^{-0.6}$ are commonly suggested (based on the theory of turbulent dispersion), many of the data show better agreement with d proportional to $V^{-1.5}$. The data of Middleman (1974) for benzene in water cover a wide range of We as shown in Figure 5. The data of Karabelas (1978) for tubes show better agreement with d proportional to $V^{-1.5}$ than with $V^{-1.2}$ For another comparison, look at the constants C_1 for the three types of dispersers. In the static mixers, the hydraulic diameter is perhaps 0.3 ID, and the velocity is doubled as a result of the reduced cross sections and the longer flow path. In the Couette disperser, the true average fluid velocity is less than half the rotor velocity, and the D/ID term also is significant. In both cases, a large part of the difference in constants is accounted for. However, the static mixer gives smaller drops and the Couette disperser gives larger drops than the tube at comparable We, Re, and D values.

Correlation of Results by Turbulent Dispersion Theory

The basic theory of turbulent dispersion indicates that the drop sizes should follow the relationship given as Eq. 1. The gel-sphere results for turbulent dispersion were primarily concerned with control of the average diameter, d, by control of the flow rate or the Couette disperser RPM. A typical set of experimen-

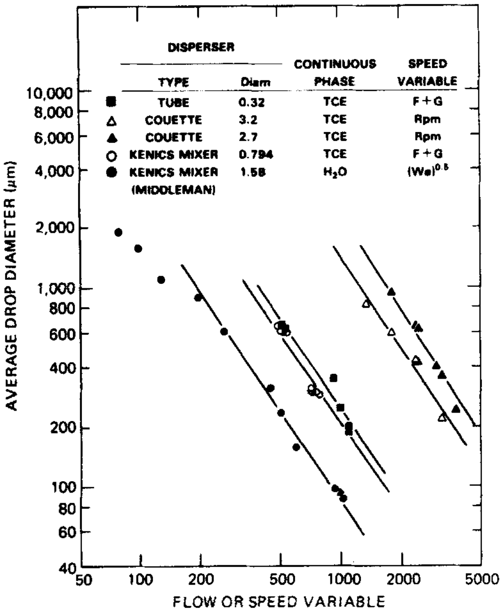


Figure 5. Drop diameter vs. velocity for turbulent dispersion.

tal data includes the dimensions of the disperser, the flow rate or the Couette disperser RPM, the average diameter of the product, the shrinkage factor *SF*, and periodic measurements of fluid properties for samples of the process fluids. With some reasonable calculations, the gel-sphere results can be shown to agree with turbulent dispersion theory.

The discussions of and selection of values of d_{max} from size distribution measurements are somewhat ambiguous and uncertain. Many authors suggest use of d_{95} as d_{max}, while others attempt to identify the largest drop of a sample. The gel-sphere data will first be examined by rearranging Eq. 1 as follows:

$$C_2\left(\frac{d}{d_{max}}\right) = P^{0.4} d \left(\frac{\gamma}{\rho}\right)^{-0.6}. \tag{11}$$

The measured size distribution of gel-sphere products from turbulent dispersion showed $d/d_{95} = 0.52$ to 0.67, and $d/d_{98} = 0.43$ to 0.60. Therefore $d_{max}/d = 1.8$ is used to allow comparisons of the gel-sphere data with theory.

The concentrations and densities of the aqueous feeds were determined for every test, as these values were necessary for calculation of shrinkage ratios. The densities and viscosities of the organic liquids were handbook values with corrections for the measured temperatures. The interfacial tensions and the viscosities of the aqueous feeds were measured periodically on samples taken from the process but were not measured for every gel-sphere test. These measured interfacial tensions were static or equilibrium values for the process fluids using a ring tensiometer. For the internal type of gelation, the interfacial tensions were measured for the organic samples vs. the two stable aqueous solutions measured separately, as the mixed solutions thicken too rapidly at room temperature for convenient measurement. The two stable solutions gave approximately the same values of interfacial tensions. The mixed feed at 0°C has a viscosity of 6 cp (mPa · s) (Saybolt and Brookfield viscosity measurement methods).

Even though the interfacial tensions were measured for samples of the process fluids, the results have the following serious uncertainties:

1. The process organics all contained a sorbitan mono-oleate surfactant (usually 0.1 to 1 vol. % for drop formation by turbulent mechanisms). The new surface formed during turbulent dispersion requires a finite time to approach the equilibrium surface tensions measured in the laboratory.
2. For the gelation-by-water extraction, the water concentrations near the interface would be different for the lab measurements and the turbulent dispersion.
3. For the internal type of gelation, the laboratory measurements are for two separate concentrated solutions at temperature equilibrium, while the turbulent dispersion during process operation involves the mixed aqueous solution starting at 0°C and the organic liquid at 60 to 90°C.

Because of these uncertainties, the exponents for Weber and Reynolds numbers in the empirical correlations were evaluated for sets of data with the disperser dimensions and the flow rates as variables and the fluid properties constant. The fluid properties were then used to evaluate the remaining constants in the empirical correlations. The results indicate that any bias in the measurement of interfacial tensions is uniform or consistent, as the empirical constants did not differ significantly for the different organic liquids.

All power inputs were calculated from the disperser dimensions and flow rates or RPM. The basic form of equation for calculation of frictional pressure drops by use of a Fanning friction factor (Perry and Chilton, 1973) is:

$$\frac{\Delta P}{L} = 2\frac{fV^2}{D} = \frac{32fQ^2}{\pi^2 D^5}$$

$$\begin{aligned} f &= 16/Re \quad \text{for} \quad Re < 2{,}000 \\ f &= 0.08/Re^{0.25} \quad \text{for} \quad Re > 4{,}000 \\ f &= 0.10 \quad \text{for} \quad Re\ 2{,}000 \ \text{to} \ 4{,}000 \end{aligned} \tag{12}$$

Multiplication by the velocity gives a power per unit mass as follows:

$$P = \frac{\Delta PV}{L} = 2\frac{fV^3}{D} = \frac{128fQ^3}{\pi^3 D^7}. \tag{13}$$

Substitution of $f = 16/Re$ for laminar flow conditions gives:

$$P = \frac{\Delta PV}{L} = \frac{32\mu V^2}{D^2\rho} = \frac{512\,\mu Q^2}{\pi^2\rho D^6}. \tag{14}$$

Substitution of $f = 0.08/Re^{0.25}$ for turbulent flow conditions gives

$$P = \frac{\Delta PV}{L} = \frac{0.16\mu^{0.25}V^{2.75}}{D^{1.25}\rho^{0.25}} = \frac{2.56\mu^{0.25}Q^{2.75}}{\pi^{2.75}\rho^{0.25}D^{6.75}}. \tag{15}$$

For the Couette disperser, the basic definition of viscosity gives the following expressions for laminar flow (annulus thickness = $D/2$):

$$\text{Force} = \frac{2\mu VA}{D}. \tag{16}$$

$$P = \left(\frac{2\mu A V}{D}\right)(V)\left(\frac{2}{DA\rho}\right) = \frac{4\mu}{D^2\rho}V^2. \qquad (17)$$

Note that the fluid velocity will only average half of the rotor velocity so that

$$Re = \frac{D\rho V}{2\mu}. \qquad (18)$$

For laminar flow with $f = 16/Re$ and by comparison of Eq. 17 with Eqs. 13 and 14,

$$P = \frac{4\mu V^2}{D^2\rho} = \frac{1}{8}\frac{fV^3}{D}. \qquad (19)$$

For turbulent flow, use $f = 0.08/Re^{0.25}$ to give

$$P = 0.01189\frac{\mu^{0.25}V^{2.75}}{\rho^{0.25}D^{1.25}}. \qquad (20)$$

The manufacturer's literature for the Kenic static mixers gives pressure-drop equations as

$$\text{Pressure drop (psi)} = \text{Flow rate (gpm)} \times \text{viscosity (cps)} \times A'. \qquad (21)$$

The values of A' are 2.1, 0.55, and 0.13 for the three static mixers used in this study. This equation has the form of a laminar flow equation. By setting Eq. 21 equal to the laminar flow equation for pressure drop, values of D can be calculated and are 0.62 to 0.64 *ID*. Use of the experimentally measured void volumes and some assumptions for velocities and hydraulic diameters also indicates the $D \simeq 0.6\ ID$. Therefore, 0.62 *ID* was used as the value of D to calculate values of P for Kenic mixers using Eq. 14 or 15.

The values of energy dissipation per unit mass of liquid were calculated using these equations and assumptions, Table 1. The average of 48 values of $C_2(d/d_{max})$ from Eq. 11 with $Re > 2{,}000$ is 0.769. For $d/d_{max} = 1.8$, this gives an average value of 1.38 for C_2. During turbulent dispersion with formation of new interfacial area, the true values of the interfacial tensions are probably higher than the measured equilibrium values used for calculations. Use of higher values of γ in Eq. 11 would give smaller values of C_2. The maximum reasonable values of γ would be those for pure liquids, and the high sorbitan mono-oleate concentrations in the organic liquid probably result in a rapid approach to equilibrium interfacial tensions. The values of C_2 for $Re < 2{,}000$ average only 13% higher than the values for $Re > 2{,}000$.

The agreement of the gel-sphere data with turbulent dispersion theory is shown by plotting P vs. $1.8\ d\ (\gamma/\rho)^{-0.6}$, Figure 6. For the Couette disperser and the static mixer, only the data for $Re > 2{,}000$ are shown. For the tube disperser, data for Re of 1,100 to 2,000 are shown with a separate symbol for comparison. Figure 6 also shows the values found by Davies (1985) for four emulsifying machines. A line is shown for Eq. 1 with $C_2 = 1.2$. This figure shows that Eq. 1 with a constant in agreement with theory can be applied for a wide range (10^9) of energy dissipation rates P, for $Re > 2{,}000$. For $Re < 1{,}000$, the drop sizes were larger but were within a factor of two of the values predicted by Eq. 1. As mentioned previously, if the true interfacial tensions were between those from equilibrium measurements and those for pure liquids, the values of C_2 would be smaller and closer to the theoretical value for turbulent dispersion. Values of C_2 from 1.1 to 1.38 could be reasonable from consideration of the effects discussed above.

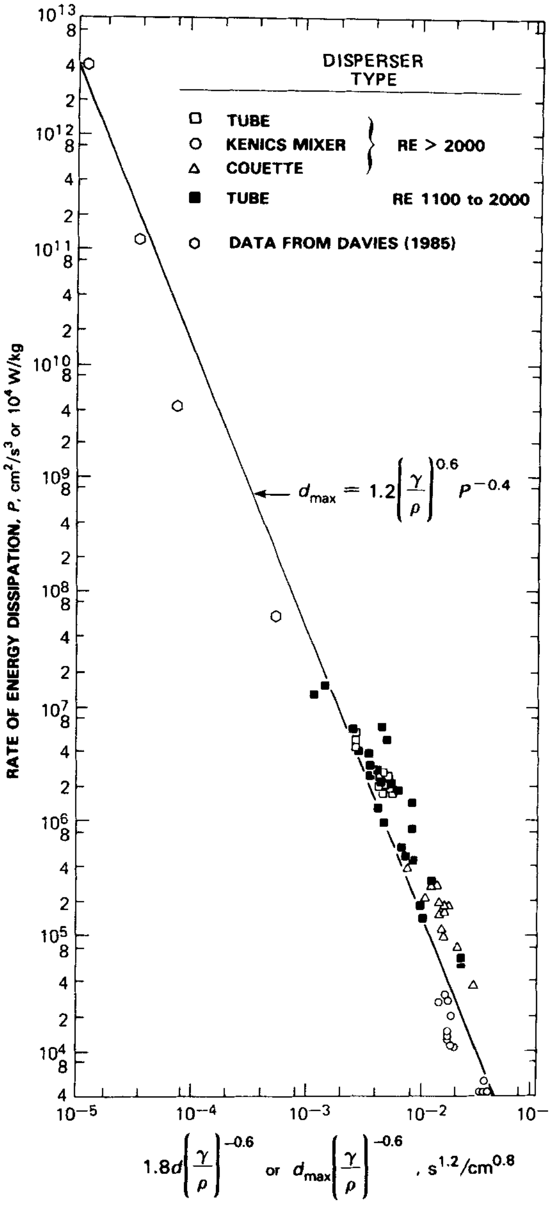


Figure 6. Experimental data for turbulent dispersion.

Since Eqs. 15 and 20 give P proportional to $V^{2.75}$, then d_{max} from Eq. 1 should be proportional to $V^{-1.10}$. But the data and literature data for single dispersers and fluid pairs show d proportional to $V^{-1.5}$. Some, but not all, of the dependencies on $V^{-1.5}$ are explained by the indeterminate friction factors in the transition region for $Re < 4{,}000$.

Comparisons of Correlations with Literature Data

Some of the turbulent dispersion data in the literature are of interest for examining the effects of individual variables. Such selected comparisons are discussed here. The data of Karabelas (1978) is for dispersion of water into two hydrocarbons with viscosities that differ by a factor of 10. The empirical form of correlation using $(\mu_d/\mu_c)^{0.5}$ accounts very well for the viscosity effect. But the empirical constant is 50, as compared to the gel-sphere constant of 15 for tubes of much smaller diameter. The plot in Figure 7 of 1.8 d $(\gamma/\rho)^{-0.6}$ vs. calculated values of P shows only a fair agreement with Eq. 1 for kerosene as the continuous phase, but the agreement is distinctly poorer for viscous transformer oil as the continuous phase. The viscous force term suggested by Davies, Eq. 2, is for the dispersed phase and does not account for the viscosity effect.

The data of Middleman (1974) describe Kenics mixers using water as the continuous phase. The data for dispersion of benzene are for a wide range of Weber numbers. The average drop diameter vs. $We^{0.5}$ has a slope of -1.5 for most of this range, Figure 5, and this agrees with the d vs. velocity or speed dependence for the gel-sphere data. Middleman gives a friction factor of 100 times the factor for a tube. For a void volume of two-thirds of the open tube volume (this may be low for the larger Kenics mixers), the values of P would be 150 times those from Eq. 13. These values of P give an excellent agreement with the theoretical correlation for turbulent dispersion, Figure 7. However, the values of P are higher than would be expected from scale-up of the pressure drop equations for the smaller Kenics mixers, and Middleman determined Sauter diameters as the average diameters.

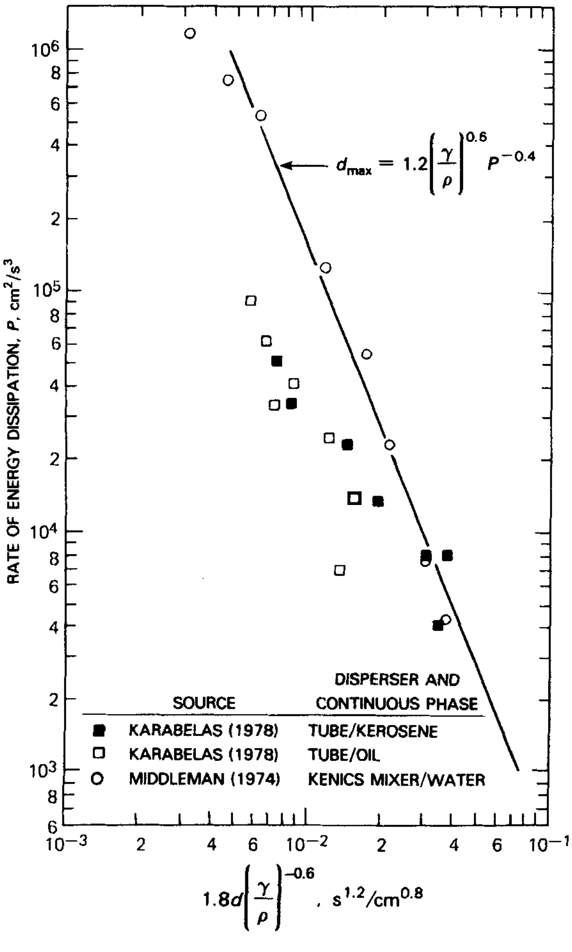


Figure 7. Correlation of selected literature data by turbulent dispersion theory.

Summary and Conclusions

Drop sizes for turbulent dispersion of aqueous solutions in organic liquids were determined for three types of dispersion devices, which were selected to give nearly homogeneous and isotropic turbulence. The gel-sphere processes used for preparation of nuclear fuels provided a convenient and reproducible measurement of the size distribution of product spheres and of the aqueous solution drops in the organic liquids. Uncertainties in the appropriate values of the interfacial tensions for gel-sphere process conditions are a major problem for correlation of results.

Empirical correlations of average drop diameters were determined using the results of a dimensional analysis and assuming an exponential form. The exponents for Weber and Reynolds numbers in the empirical correlations were evaluated without the use of interfacial tension values. Laboratory equilibrium values for interfacial tensions were measured using samples of process fluids, and these values were used to determine the remaining empirical constants. These empirical correlations are convenient for selection of gel-sphere preparation conditions but should not be extrapolated.

A theory of turbulent dispersion involving eddy velocities and the rate of energy dissipation provides a theoretical correlation for maximum drop diameters. The theory of homogeneous, isotropic turbulence was originated by Kolmorogoff (1949). Davies (1985) compares this theory with four types of dispersion devices with high rates of energy dissipation. The gel-sphere data were compared in the same way using calculated values of the rate of energy dissipation, P, and using $d_{max} = 1.8\ d$, where d is the weight average diameter and d_{max} is the maximum diameter of the turbulent dispersion theory. The gel-sphere results show good agreement with

$$d = \frac{2}{3}\left(\frac{\gamma}{\rho}\right)^{0.6} P^{-0.4} \tag{22}$$

where γ is the interfacial tension measured for process samples and ρ is the continuous phase density. The range of values for P was 10^3 to 10^7 cm^2/s^3 or 10^{-1} to 10^3 W/kg. Using $d_{max} = 1.8\ d$, the values of Davies (1985) and the gel-sphere results together show agreement with the turbulent dispersion theory for P of 10^3 to $4 \cdot 10^{12}$ cm^2/s^3. The relationship is:

$$d_{max} = 1.2\left(\frac{\gamma}{\rho}\right)^{0.6} P^{-0.4}. \tag{23}$$

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Notation

A = area of Couette flow
a, b, c, g = exponents for a dimensional analysis correlation
C_1, C_2 = dimensionless constants in correlations of drop diameters
d = average drop diameter on a volume or weight basis
d_{84}, d_{95} = drop diameter with 84 or 95 vol. % or wt. % of the total smaller than d_{84} or d_{95}
d_{max} = maximum stable drop diameter
D = hydraulic diameter of dispersion device
$F + G$ = total flow rate of the two phases

f = Fanning friction factor, Perry and Chilton (1973)
g = gravitational constant
ID = diameter other than D or d: inside diameter for static mixer, rotor diameter for Couette disperser
L = length of flow channel
P = rate of energy dissipation per unit mass of liquid
ΔP = pressure drop due to friction
Q = volumetric rate of flow
RPM = Revolutions per minute for couette disperser rotor
Re = Reynolds number = $DV\rho/\mu$, or $DV\rho/2\mu$ for Couette disperser
SF = shrinkage factor or ratio of drop diameter to gel-sphere product diameter
V = average velocity
v = turbulent (eddy) velocity
We = Weber number = $DV^2\rho/\gamma$

Greek letters

γ = interfacial tension
ρ = fluid density of continuous phase
$\Delta\rho$ = fluid density difference
σ = standard deviation for drop size distribution, log normal basis
μ_c = fluid viscosity of continuous phase
μ_d = fluid viscosity of dispersed phase

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