27
1-21-82
②
242 HTIS
ornl

②

I 937

ORNL/MIT-337

MASTER

OAK
RIDGE
NATIONAL
LABORATORY

UNION
CARBIDE

**A Study of Droplet Formation
for Preparation of Spheres by
Internal Gelation**

Chihang R. Chu
John C. Card

ORNL/MIT--337

DE82 007500

OPERATED BY
UNION CARBIDE CORPORATION
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Contract No. W-7405-eng-26

A STUDY OF DROPLET FORMATION FOR PREPARATION OF SPHERES BY INTERNAL GELATION

Consultants
P.A. Haas, A.D. Ryon, and W.D. Bond

Oak Ridge Station
School of Chemical Engineering Practice
Massachusetts Institute of Technology
C.H. Byers, Director

Oak Ridge National Laboratory
Oak Ridge, Tennessee 37830
Operated by
Union Carbide Corporation
for the
Department of Energy

DISCLAIMER

It is hereby acknowledged that the following information was obtained from the National Security Agency (NSA) and is being provided to you for your information only. The NSA is not responsible for the accuracy, reliability, or completeness of the information provided. The NSA is not responsible for the use of the information provided. The NSA is not responsible for the consequences of the use of the information provided. The NSA is not responsible for the consequences of the use of the information provided.

DISTRIBUTION OF "C" CATEGORY IN QUANTITIES

Contents

	<u>Page</u>
1. Summary	1
2. Introduction	2
2.1 Treatment of Nuclear Wastes	2
2.2 Literature Review	2
2.3 Objectives	4
3. Apparatus and Procedures	6
3.1 Apparatus	6
3.1.1 Droplet-Formation Apparatus	6
3.1.2 Catcher-Fluid Apparatus	9
3.2 Procedure	9
3.2.1 Droplet Formation	9
3.2.2 Catcher-Fluid Stream	12
4. Results	12
4.1 Experimental Observations	12
4.2 Dimensional Analysis	18
4.3 Computer Models	22
4.4 Catcher-Fluid Stream	22
5. Discussions	24
5.1 Limitations of the Correlations	24
5.2 Comparison with the Literature	30
6. Conclusions	30
7. Recommendations	30
8. Acknowledgments	30
9. Appendix	31
9.1 Newtonian-Jet Breakup	31
9.2 Accurate Calibration Curves	34
9.3 Dimensional Analysis	36
9.4 SAS Program Listing and Data Base	38
9.5 Sample Calculations	45
9.6 Location of Original Data	46
9.7 Nomenclature	46
9.8 Literature References	48

1. SUMMARY

Internal gelation has been suggested as a means of permanently immobilizing highly radioactive nuclear wastes from the Savannah River Plant of the DOE. These wastes would be incorporated into a gel structure in the form of small, hardened ceramic spheres, and as such, could be easily transported and safely stored. To form these spheres by internal gelation, a chilled feed broth containing the wastes, matrix material, and other components is fed to a vibrating nozzle, forming a jet which breaks up into a stream of droplets in the air. These droplets then fall into a hot forming liquid, where the release of ammonia triggers a gelation reaction. The spheres are then coated for additional protection against leaching. To ensure a uniform coating, the spheres must be of uniform size.

The critical step in forming spheres of uniform size is good droplet formation at the vibrating nozzle. An experimental study was conducted to investigate the factors affecting droplet formation. Three different fluids were investigated, whose physical properties cover the range which will be encountered in the feed broth. Three stainless-steel nozzles and one glass unit in the diameter range 325-586 μm were used with jet velocities varying from 1 to 10 m/s, which allowed the generation of an extensive data base. The important dimensionless groups, such as Reynolds (Re), Weber (We), Froude (Fr), and Power (Po) numbers, and a dimensionless droplet spacing (λ/D_n) were identified by a dimensional analysis. The data were then correlated as a power function of these groups. Two correlations resulted. The upper bound of good droplet formation, which corresponded to the maximum allowable flow rate at a fixed frequency, was given by:

$$\frac{\lambda}{D_n} = 1.66 \text{ Re}^{-0.00043} \text{ We}^{-0.43} \text{ Fr}^{0.38} \text{ Po}^{-0.037}$$

The lower bound, representing the minimum flow rate when the frequency was fixed, was given by:

$$\frac{\lambda}{D_n} = 7.32 \text{ Re}^{0.016} \text{ We}^{-0.066} \text{ Fr}^{-0.049} \text{ Po}^{-0.020}$$

A further obstacle to good gelled-sphere formation is the coalescence of droplets at the surface of the hot forming liquid at high vibrator frequencies and flow rates. The use of a catcher stream, which traps the droplets in the air and transports them below the surface of the forming liquid, was investigated. The catcher fluid, trichloroethylene (TCE), was found to remain as a liquid sheet when it was directed onto a glass plate. It was recommended that this catcher-stream design and the two correlations be applied at the pilot plant at ORNL.

2. INTRODUCTION

2.1 Treatment of Nuclear Wastes

Over the past twenty-five years, highly radioactive nuclear wastes have accumulated at the Savannah River Plant (SRP) of the Department of Energy. These wastes are derived from the chemical reprocessing of nuclear reactor fuel and target elements from production reactors. They are alkaline slurries of primarily hydrous oxides of iron, aluminum, and manganese that have been precipitated with excess NaOH. Presently these wastes are stored in high-integrity, double-walled tanks at SRP. Several methods for the permanent immobilization of these high-level wastes are presently being developed as part of the Department of Energy's plan for long-term management of all federally owned reprocessing waste (3, 15).

A proposed immobilization method is incorporation of the SRP sludge solids into a gel structure in the form of small spheres (approximately 1-mm diameter). Once they are washed, calcined, and coated, these hardened, ceramic spheres offer excellent resistance to leaching and are easily transported and stored.

Internal gelation [using a feed brach containing the waste, matrix components, urea, and hexamethylenetetramine (HMTA) that is chilled, mixed, and fed to a vibrating nozzle] is the proposed sphere-production method. The jet, which leaves the nozzle, breaks up into a stream of droplets in the air. Subsequently, the droplets enter a hot organic liquid, such as trichloroethylene (TCE) or silicone oil, where the HMTA decomposes, releasing ammonia, which causes gelation of the droplets. The gelled spheres are then washed, dried, and calcined. For additional protection against leaching, a coating of pyrolytic carbon is applied to the spheres. Finally, for increased mechanical strength and protection against oxidation, the spheres are coated with $\alpha\text{-Al}_2\text{O}_3$.

To ensure that a uniform coating is applied to the spheres in the coating process, spheres of a uniform size are desired. Since the spheres are a direct result of the droplets formed by the vibrating nozzle, the critical step in the uniform-sphere size is the formation of uniform droplets by the vibrating nozzle. The following section discusses a method for obtaining uniform droplet.

2.2 Literature Review

According to the classical mechanism postulated by Lord Rayleigh, a free column of liquid is unstable if its length is greater than its circumference (14). The process by which drops are produced from a liquid stream depends on the nature of the flow such as the way in which energy is imparted to the liquid, the properties of the liquid (viscosity, density, and surface tension), and the properties of the ambient atmosphere (2).

One practical way to produce droplets is through the use of a pulsed nozzle. If a jet is excited by a small-amplitude axial vibration of appropriate frequency, the jet breaks up into uniformly spaced drops. The size of the drops can be controlled by the liquid flow rate and by the frequency of vibration imposed on the jet. It is assumed that one drop is formed per cycle of vibration per orifice (16). Therefore, a simple mass balance gives:

$$Q = \left(\frac{\pi D^3}{6}\right) f N \quad (1)$$

where:

Q = volumetric flow rate of liquid

f = frequency

N = number of orifices

For a single nozzle, Eq. (1) reduces to the following expression for droplet diameter:

$$D = \left(\frac{6}{\pi} \frac{Q}{f}\right)^{1/3} \quad (2)$$

For the jet to be stable, however, the droplet diameter can only fall within a certain range. Merrington and Richardson (12) found that with nozzles up to 1000 μm , the drop size obtained at low velocities was roughly twice the nozzle diameter. The ratio of drop diameter to nozzle diameter ranged from 1.8 to 2.3.

Although a pulsed nozzle is good for producing uniform drops, it also has a tendency to form relatively smaller droplets interspersed among the main drops (satellite drops), which is undesirable in the internal gelation process. Several satellite studies focused on the region of operating conditions, which would suppress the formation of satellites.

In a paper by Pimbley and Lee (13), a second-order spatial-instability analysis was used to simulate the formation of satellites from the main drops. According to Pimbley and Lee, the two most important parameters which control satellite formation are the amplitude of the perturbation and the wavelength-to-diameter ratio. A satellite drop can separate from a larger drop on the fore side of the satellite first, on the aft side first, or on both ends simultaneously. The time interval between the two separations is defined as the satellite interaction time. Within this time period momentum transfer occurs, which results in the slowing down or speeding up of the satellite relative to the main stream. With very small perturbation amplitude, the first separation occurs on the fore

side of the satellite, producing a slightly lower satellite velocity. At a later time, the satellites will merge with the main drops, causing non-uniform droplet sizes. Visually, the jet thus contains rear-merging satellites. Figure 1A schematically illustrates such a situation.

When the perturbation amplitude is increased, a point is reached where the two ends of the satellite break off simultaneously. No momentum transfer takes place, and the satellites move with the same velocity as the main stream. This is the infinite satellite condition shown in Fig. 1B. When the amplitude is increased beyond this point, the jet breaks up at the aft side of the satellites first, producing forward-merging satellites as shown in Fig. 1C.

In summary, a pulsed nozzle can produce uniform spherical droplets, although several parameters seem to affect the uniformity of the droplets. A major difficulty is the formation of satellite droplets. At Oak Ridge National Laboratory (ORNL), previous work applied this technique to the internal gelation process (7, 8, 9, 18). However, the operating conditions for good droplet formation were mostly determined by trial-and-error. A study was needed to define the limits of operating range.

Once formed in the air, the droplets must enter the heated organic fluid without coalescence. The researchers at ORNL found that at high frequency of droplet formation and high velocities, the droplets tended to coalesce as they struck the surface of the heated organic medium, rather than passing into it and hardening into uniform-size spheres. To transport the droplet into this forming liquid and to avoid this coalescence at the surface, a catcher stream can be used. Droplets formed at the nozzle fall into a passing stream, which conveys them below the surface of the forming liquid. While some catcher-stream concepts have been suggested, an extension of these ideas is necessary before they will be usable, particularly with a TCE medium. Later a study of the hydrodynamics of such a configuration will be necessary.

2.3 Objectives

The objectives of this study were:

1. To study the effect of viscosity, density, and surface tension of the fluid, oscillation frequency and power input to the nozzle, orifice diameter, and fluid flow rate upon the range of good droplet formation.
2. To develop a correlation to describe the operable range of all such variables.
3. To investigate the use of a catcher-fluid stream, which traps and carries the droplets into the organic medium, especially at high frequencies.

A) Rear-Merging Satellites

B) Infinite Satellite Condition

C) Forward-Merging Satellites

MASSACHUSETTS INSTITUTE OF TECHNOLOGY
SCHOOL OF CHEMICAL ENGINEERING PRACTICE
AT
OAK RIDGE NATIONAL LABORATORY

THREE TYPES OF SATELLITE FORMATION

DATE	DRAWN BY	FILE NO.	FIG.
9-18-81	JCC	CEPS-X-337	1

3. APPARATUS AND PROCEDURE

3.1 Apparatus

3.1.1 Droplet-Formation Apparatus

The central feature of the experimental apparatus (see Fig. 2) was a steel or glass nozzle with a circular orifice, which was attached to the diaphragm of the electromechanical vibrator (Alpha-M Corp. Model AV-6), so that it produced axial vibration of the fluid stream. The shaft of the nozzle was mounted at a 45° angle below horizontal. Four stainless steel nozzles and one glass nozzle were used in the experiments (see Fig. 3). The 180- μ m orifice, however, was apparently too narrow, and it was not possible to issue a liquid jet from any of the test liquids.

The orifice of each nozzle was examined under a microscope (50X magnification); the orifices of the stainless-steel nozzles were circular at least to the extent that their shape was visible, while the orifice of the glass nozzle was somewhat elliptical (maximum diameter/minimum diameter = 1.2). The edges of all orifices appeared very rough. Other nozzle characteristics are listed in Table 1.

Table 1. Nozzle Characteristics

Orifice Diameter (μ m)	Material of Construction	Weight (g)	Straight Tubing Section Before Orifice (cm)
180	stainless steel	13	1.3
325	stainless steel	13.78	1.3
350	stainless steel	13.92	1.3
501	stainless steel	12.62	1.3
586	glass	10.60	3.8

The vibrator was driven by a 0-to-25-watt power amplifier (1) (Alpha-M Corp. Model OC-25) with a frequency range of 20 to 20,000 Hz. An AC ammeter on the front panel of the amplifier and a Simpson Model 313 voltmeter permitted the vibration power to be calculated. The frequency of the vibrator-drive signal was accurately measured (± 0.1 Hz) with a digital frequency counter (HP Model 5308A), while a stroboscope (General Radio Model 1538A Strobotac) allowed the droplets to be observed. The stroboscope frequency was set at a fractional harmonic (usually within

A) Stainless-Steel Nozzle

B) Glass Nozzle

MASSACHUSETTS INSTITUTE OF TECHNOLOGY
SCHOOL OF CHEMICAL ENGINEERING PRACTICE
AT
OAK RIDGE NATIONAL LABORATORY

TWO TYPES OF NOZZLES USED
IN DROPLET FORMATION STUDY

DATE
9-18-81

DRAWN BY
JCC

FILE NO.
CEPS-X-337

FIG.
3

the range of $1/10$ to $1/50$) of the vibrator frequency. This made the droplet stream appear stationary and facilitated measurements of droplet spacing.

The test liquid flowed to the nozzle from a pressurized feed tank through flexible tubing to avoid the introduction of extraneous signals to the nozzle. Fluid flow rate was controlled by regulating the air pressure above the liquid (0-50 cm-Hg); this pressure was measured on a mercury manometer. The liquid level in the feed tank was measured with a ruler, so that its static head contribution to the total pressure upstream of the nozzle could be calculated. The static head ranged from 0.7 to 38.3% of the total pressure. However, its contribution to the total pressure changed at most by 1.5% during any single run.

3.1.2 Catcher-Fluid Apparatus

Figure 4A is a schematic diagram of the catcher-fluid stream apparatus. A close-up side view of the catcher-stream nozzle is shown in Fig. 4B. The nozzle was constructed from a 10-in. section of $\frac{1}{2}$ -in.-OD stainless-steel tubing, which was sealed at one end. A slot 2.54 cm long and 500 μ m wide was cut into the wall of the tubing, 2.5 cm from the end. A needle valve was attached to the other end to control the catcher-fluid flow rate out of the slot.

The catcher fluid trichloroethylene (TCE) flowed from a pressurized feed tank similar to the one used for the test liquid. The catcher fluid issued from the slot as a flat sheet 2.5-cm wide and approximately 0.5-mm thick. The thickness of the jet was very non-uniform and many ripples were present, making the thickness vary from about 0.3 to 0.7 mm along the width of the jet. At approximately 2.5 cm from the nozzle, the jet collapsed into a single thread.

To provide a catcher stream with a more uniform surface, the catcher fluid was directed onto a 15-cm square sheet of Pyrex glass plate. The nozzle was mounted approximately 1 cm above the surface of the glass. The catcher fluid then flowed out of the slot and onto the glass surface, expanding to an 8-cm-wide film with 0.2 mm thickness as it flowed down the plate. The surface of the catcher-fluid film on the glass was much more uniform than that of the free jet. The droplet stream was directed into this moving film.

3.2 Procedure

3.2.1 Droplet Formation

Three fluids were used in this study. Their physical properties of interest are listed in Table 2.

A) Catcher-Stream Apparatus

B) Catcher-Stream Nozzle

MASSACHUSETTS INSTITUTE OF TECHNOLOGY
SCHOOL OF CHEMICAL ENGINEERING PRACTICE
AT
OAK RIDGE NATIONAL LABORATORY

CATCHER-FLUID-STREAM DESIGN

DATE	DRAWN BY	FILE NO.	FIG.
9-18-81	JCC	CEPS-X-337	4

Table 2. Physical Properties of Model Fluids at 20°C

Fluid	Viscosity (Pa-s)	Density (kg-m ⁻³)	Surface Tension (N-m ⁻¹)
Ethylene glycol	1.99×10^{-2}	1113	4.77×10^{-2}
20% glycerol (aqueous)	1.769×10^{-3}	1046.9	$7.2' \times 10^{-2}$
Distilled water	1.005×10^{-3}	1000	7.275×10^{-2}

Experiments consisted of systematically setting flow rates and frequencies and observing the resulting droplet stream under the stroboscope to determine if a condition of stable, uniform droplet formation existed. For each combination of orifice size and fluid tested, frequency and flow rate were adjusted until the entire range of obtainable frequencies and flow rates for which stable droplet formation was obtained. The minimum operable frequency, a lower pinch-point, was the starting point of all experiments on a particular combination of orifice and fluid. In the ethylene glycol runs, flow rate was increased stepwise and the maximum and minimum frequencies for good droplet formation were found at each step. Step size was chosen to give 8 to 16 data points (minimum and maximum) before the pressure limitations of the equipment (50 cm Hg) were reached. On the other hand, in the water and 20%-aqueous glycerol runs, frequency was increased stepwise and the minimum and maximum flow rates for good droplet formation were found at each step.

At the upper and lower bounds of each step, measurements of manometer pressure, feed-tank level, amplifier voltage, current and frequency, droplet spacing, and length and position of good droplet region within the jet were recorded. Droplet spacing was determined by positioning a ruler behind the droplet stream and counting the number of droplets in one inch of jet length. In the experiments using ethylene glycol, the flow rate was measured directly for each run by diverting the droplet stream into a beaker for a measured time and weighing its contents. Calibration plots of flow rate versus pressure upstream of the nozzle were developed by the same flow measurement technique for water and 20%-aqueous glycerol for all the nozzles used (see Appendix 9.3). Flow rates in the water and 20%-aqueous glycerol experiments were read directly from these plots.

In the 67 experiments with water, all four nozzles were investigated; while in the 25 glycol and 25 20%-aqueous glycerol experiments, time constraints permitted investigation with only the three smaller nozzles (325-500 μ m). These resulted in 117 data points to be used in the correlations.

3.2.2 Catcher-Fluid Stream

Several factors can affect the performance of the catcher streams: catcher-stream velocity, choice of catcher fluid, ratio of catcher-stream velocity to droplet-stream velocity, angle between catcher stream and droplet stream, and shape of catcher-stream nozzle. In this study a single catcher fluid (TCE) and a single catcher-stream nozzle (see Fig. 3B) were investigated. At velocities up to the maximum obtainable on the equipment (80 cm/s), no uniform catcher stream could be formed; ripples always appeared in the surface of the stream.

To form a more uniform catcher-stream surface, the stream was directed onto a 15-cm square Pyrex glass plate. For the two flow rates investigated (7.8 and 10.1 cm³/s), the angle of incidence of the catcher stream upon the glass was varied from 0 to 180°, and the uniformity of the catcher stream surface was observed.

4. RESULTS

4.1 Experimental Observations

In the experiments, the behavior of about 22 cm of the jet was observed. Figures 5 through 7 are a series of photographs taken for a water jet issuing from a 350- μ m nozzle. Figure 5 shows a natural jet which breaks up into droplets of various diameters. The coherent portion of the jet is 4 cm. Droplet spacing is nonuniform, and coalescence of droplets appears throughout the jet. In Figs. 6 and 7, a frequency of 800 Hz was imposed on the nozzle. Within a certain power range, a steady jet breaks into uniform droplets. At steady state the first break in the liquid stream occurs at 1.5 cm from the orifice. The region of good drops extends to 13 cm, where doublets begin to appear. When excessive power is applied, causing a large amplitude of vibration, the jet disintegrates into several streams (atomization), which can be seen in Fig. 7.

Droplet diameters are correspondingly larger, and more details of the drops are observed when a larger nozzle is used. Figure 8 shows a water jet issuing from a 501- μ m nozzle with an imposed frequency of 1000 Hz. The photograph shows the oscillation and deformation of droplets caused by the waveform of the vibrator. The photographs in Cline and Anthony (4) show similar droplet behavior.

Both forward-merging and rear-merging satellites were observed for almost all runs and for different values of oscillator power. However, forward-merging satellites were more common than the rear-merging variety. Figure 9 is a photograph of a water jet, which shows the appearance of forward-merging satellites. It can also be noticed that the satellite stream is slightly off-center.

Fig. 5. Natural Water Jet (No Imposed Oscillation)

$D_n = 350 \text{ } \mu\text{m}$
 $Q = 0.228 \text{ cm}^3/\text{s}$

Fig. 6. Water Jet at Steady State

$D_n = 350 \text{ } \mu\text{m}$
 $Q = 0.220 \text{ cm}^3/\text{s}$
 $f = 800 \text{ Hz}$
 $P = 0.04 \text{ W}$

Fig. 9. Breakup of a Water Jet with Satellite Formation

$D_n = 350 \mu m$
 $Q = 0.473 \text{ cm}^3/\text{s}$
 $f = 1280 \text{ Hz}$
 $P = 1.5 \text{ W}$

Fig. 7. Atomization of Water Jet Due to Excessive Power

$D_n = 350 \text{ } \mu\text{m}$
 $Q = 0.060 \text{ cm}^3/\text{s}$
 $f = 800 \text{ Hz}$
 $P = 9.3 \text{ W}$

Sixty runs were made to determine the minimum fluid flow rate allowable at a given frequency. For each fluid and a particular nozzle, these minimum flow rates, or equivalently, minimum velocities, were then related to the frequencies. Figure 10 is a plot generated in this manner. Each point in Fig. 10 represents an experiment where a stable operation and uniform droplet formation existed. Similarly, other combinations of nozzles and fluids were examined. Furthermore, another 54 runs were made to determine the maximum flow rate for a given frequency. Figure 11 shows all data points for the minimum case, and Figure 12 summarizes runs at maximum flow rates.

The data obtained indicated that two correlations are necessary to fully describe the upper and lower bounds of good droplet formation. The upper bound is the maximum flow rate at a fixed frequency, which produces good drop formation. Equivalently, the upper bound is the minimum frequency at a fixed flow rate, with all else being constant, which will form good drops. Physically, excessive flow rate and low frequency causes more fluid to be available than is required for single-drop formation. Hence, the drop disintegrates and forms side streams. Similarly, the lower bound for uniform droplet formation is the minimum flow rate at a fixed frequency attainable if the flow rate is fixed. The droplet diameter as well as the void spacing between the drops are excessively small, causing coalescence to occur.

4.2 Dimensional Analysis

The problem of selecting parameters to enter into the correlations can be solved by a dimensional analysis, because this will combine parameters into meaningful dimensionless groups. In this analysis, the operable frequency is assumed to be a function of viscosity, surface tension, density, nozzle diameter, gravitational force, droplet spacing, and power output of the amplifier. The actual amplitude of the displacement of the vibrator diaphragm may be an important variable, but it is assumed here that the diaphragm displacement is linearly proportional to the power supplied to the vibrator. The dimensional analysis has an infinite number of possible solutions (see Appendix 9.4). However, for all cases examined, five important dimensionless groups emerged: Reynolds, Weber, and Froude numbers, a Power number P_0 , and a dimensionless droplet spacing. The droplet spacing λ is defined as the center-to-center distance of the main droplets. The result of dimensional analysis is given as:

$$\left(\frac{\lambda}{D_n}\right) = F'(Re)^A (We)^B (Fr)^C (P_0)^D \quad (3)$$

where

$$F' = \text{constant}$$

MASSACHUSETTS INSTITUTE OF TECHNOLOGY
SCHOOL OF CHEMICAL ENGINEERING PRACTICE
AT
OAK RIDGE NATIONAL LABORATORY

LIMITING STABLE OPERATING CONDITIONS
FOR WATER JET USING A 325- μ m NOZZLE

DATE 9-19-81	DRAWN BY CRC	FILE NO. CEPS-X-337	FIG. 10
-----------------	-----------------	------------------------	------------

MASSACHUSETTS INSTITUTE OF TECHNOLOGY
SCHOOL OF CHEMICAL ENGINEERING PRACTICE
AT
OAK RIDGE NATIONAL LABORATORY

STABLE OPERATING CONDITIONS
FOR ALL FLUIDS AT MINIMUM
VELOCITIES FOR VARIOUS FREQUENCIES

DATE 9-19-81	DRAWN BY JCC	FILE NO. CEPS-X-337	FIG. 11
-----------------	-----------------	------------------------	------------

MASSACHUSETTS INSTITUTE OF TECHNOLOGY
SCHOOL OF CHEMICAL ENGINEERING PRACTICE
AT
OAK RIDGE NATIONAL LABORATORY

STABLE OPERATING CONDITIONS
FOR ALL FLUIDS AT MAXIMUM
VELOCITIES FOR VARIOUS FREQUENCIES

DATE	DRAWN BY	FILE NO.	FIG.
9-18-81	JCC	CEPS-X-337	12

$$\frac{\lambda}{D_n} = \text{dimensionless droplet spacing}$$

$$Re = \frac{D_n V \rho}{\mu} = \text{Reynolds Number}$$

$$We = \frac{C_n V^2 \rho}{\sigma} = \text{Weber Number}$$

$$Fr = \frac{V^2}{g D_n} = \text{Froude Number}$$

$$P_o = \frac{P}{\rho f^3 \lambda^2 D_n^3} = \text{Power Number}$$

4.3 Computer Models

With the aid of SAS (Statistical Analysis System) programs (15), a correlation was developed for the upper-bound case, and a similar relationship was developed for the lower-bound case. These are shown in Table 3.

The model for the upper bound case indicates that dependence on Reynolds number and Power number is negligible, while Weber and Froude numbers are more important. The model for the lower bound, however, shows that all dimensionless groups are about equally insignificant. The model practically gives a constant value of λ/D_n . The table has also listed the values of R^2 and the coefficient of variation. R^2 measures how much variation in the dependent variable can be accounted for by the model. It can range from 0 (no correlation) to 1 (perfect correlation). Coefficient of variation describes the amount of variation in the population. Appendix 9.5 contains a listing of the program, which was written to access the SAS routines and to perform the data regression.

4.4 Catcher-Fluid Stream

When trichloroethylene (TCE) was introduced through the 500- μm slot, the liquid sheet contained nonuniformities in the form of ripples. The liquid sheet collapsed into a single thread within the first inch. When the liquid sheet was directed onto a 15 x 15-cm Pyrex glass plate, the liquid sheet stayed coherent, although it spread and became thinner. The surface of this moving liquid film was more uniform than before.

Table 3. Models for the Upper and Lower Bounds
of Stable and Uniform Droplet Formation

Model for the Upper Bound

$$\left(\frac{\lambda}{D_n}\right) = 1.66 \text{ Re}^{-0.00043} \text{ We}^{-0.43} \text{ Fr}^{0.38} \text{ Po}^{-0.037}$$

$$R^2 = 0.52$$

$$\text{C.V.} = 9.6\%$$

$$\left(\frac{\lambda}{D_n}\right)_{\text{mean}} = 7.54$$

Model for the Lower Bound

$$\left(\frac{\lambda}{D_n}\right) = 7.32 \text{ Re}^{0.016} \text{ We}^{-0.066} \text{ Fr}^{-0.049} \text{ Po}^{-0.020}$$

$$R^2 = 0.15$$

$$\text{C.V.} = 17\%$$

$$\left(\frac{\lambda}{D_n}\right)_{\text{mean}} = 4.83$$

5. DISCUSSION

5.1 Limitations of the Correlations

Figures 13 and 14 are the predicted $\ln(\lambda/D_n)$ vs actual $\ln(\lambda/D_n)$ for the upper- and lower-bound cases, respectively. The 45° straight line represents a perfect fit. The scattering of the data points is random indicating that no systematic bias exists in the models. These models contain five adjustable constants, and the models are empirical in nature. Judging from the values of R^2 and the coefficients of variations (C.V.) for both models as shown in Table 3, the model for the upper bound is more successful than the model for the lower bound in terms of fitting the data. The latter predicts an approximately constant value of λ/D_n , regardless of the values of all four dimensionless groups. Both models should only be applied within the range of operating conditions investigated in this study. Extrapolation is not recommended.

Theoretically, the models can be used to predict the operable range of a parameter once everything else has been specified. In practice, however, the application of the model is not straightforward because several variables, such as the nozzle diameter and the velocity, appear more than once in the correlations. Typically, one may want to know the limiting velocities for given D_n , f , and fluid properties. By definitions, velocity is included in three dimensionless groups, Re , We , and Fr , and can therefore be solved by trial-and-error only.

To overcome this difficulty, it is suggested that Figs. 11 and 12 should be used directly for a first estimation of the limiting velocities. Within the range of fluid properties tested, the minimum and maximum velocities can be obtained if the frequency is known. Furthermore, linear fits for the maximum and minimum velocities vs frequency are given in Table 4. These linear relationships are entirely empirical, based on recorded values of f and calculated values of V . Appropriate units must be used in both equations as shown in Table 4. It is interesting to notice that these best line fits are quite satisfactory. The values of R^2 are greater than 0.7, which are better than the fits obtained for both the upper-bound and lower-bound models. When these best line fits are drawn, as shown in Fig. 15, a "pocket" of allowable operation is thus defined to be the region between the two lines. It is again emphasized that the linear relations should not be extrapolated beyond the ranges of parameters investigated. Table 5 summarizes the ranges of several variables.

It is also assumed that one drop is formed per cycle of vibration per orifice (16), and therefore $V = f\lambda$. This is an approximation at best. The acceleration of gravity changes V , and the drop spacing changes with distance from the nozzle. However, only the first 22 cm of the jet was examined in any run. Gravity was relatively unimportant in this region. Furthermore, the calculated droplet spacing λ confirmed with the observed λ to within 10%; so the assumption of $V = f\lambda$ was acceptable.

Line shown for Predicted $\ln(\frac{\lambda}{D_c}) = \text{Actual } \ln(\frac{\lambda}{D_n})$

MASSACHUSETTS INSTITUTE OF TECHNOLOGY
SCHOOL OF CHEMICAL ENGINEERING PRACTICE
AT
OAK RIDGE NATIONAL LABORATORY

COMPARISON OF UPPER-BOUND
CORRELATION WITH ACTUAL DATA

DATE 10-4-81	DRAWN BY JCC	FILE NO. CEPS-X-377	FIG. 13
-----------------	-----------------	------------------------	------------

MASSACHUSETTS INSTITUTE OF TECHNOLOGY
SCHOOL OF CHEMICAL ENGINEERING PRACTICE
AT
OAK RIDGE NATIONAL LABORATORY

COMPARISON OF LOWER-BOUND
CORRELATION WITH ACTUAL DATA

DATE	DRAWN BY	FILE NO.	FIG.
10-4-81	JCC	CEPS-X-33	14

**Table 4. Linear-Regression Result for
Maximum and Minimum Velocities**

$$V_{\max} = 0.14 f + 267$$

$$R^2 = 0.75$$

$$\text{C.V.} = 20.8\%$$

$$V_{\min} = 0.094 f + 169$$

$$R^2 = 0.72$$

$$\text{C.V.} = 25.6\%$$

$$\text{Units: } V = \text{cm}^3/\text{s}$$

$$f = \text{Hz}$$

Darkened symbols for upper-bound data.
Open symbols for lower-bound data.

MASSACHUSETTS INSTITUTE OF TECHNOLOGY
SCHOOL OF CHEMICAL ENGINEERING PRACTICE
AT
OAK RIDGE NATIONAL LABORATORY

LINEAR REGRESSION OF VELOCITY
WITH IMPOSED FREQUENCY

DATE 9-22-81	DRAWN BY JCC	FILE NO. CEPS-X-337	FIG. 15
-----------------	-----------------	------------------------	------------

Table 5. Ranges of Parameters Investigated

	<u>Minimum</u>	<u>Maximum</u>	<u>Possible Error</u>
<u>Fluid Properties (at 20°C)</u>			
Density, ρ (kg/m ³)	1000	1113	
Viscosity, μ (Pa-s)	1.005×10^{-3}	1.99×10^{-2}	
Surface Tension, σ (N/m)	4.77×10^{-2}	7.275×10^{-2}	
<u>Measured Quantities</u>			
Frequency, f (Hz)	460	7050	± 0.1
Nozzle Diameter, D_n (μ m)	325	586	± 14
Flow Rate, Q (cm ³ /s)	0.000	1.63	± 0.03
Droplet Spacing, λ_{obs} (mm)	1.016	5.080	± 1 drop/2.5 cm
Amplifier Current, I (amp)	0	1.5	± 0.05
Amplifier Voltage, E (V)	0	6.2	± 0.1
<u>Derived Quantities</u>			
Velocity, V (cm/s)	179	1133	
Droplet Spacing, λ_{calc} (mm)	1.024	5.067	
Droplet Diameter, D (μ m)	545	1199	
Amplifier Power, P (W)	0	9.3	
<u>Dimensionless Groups</u>			
Reynolds No., Re	77	3664	
Weber No., We	15	804	
Froude No., Fr	702	40,300	
Power No., Po	3.96×10^{-7}	2.03×10^{-2}	
Dimensionless Droplet Spacing, λ/D_n	2.9	12	
Diameter Ratio, D/D_n	1.56	2.62	

5.2 Comparison with the Literature

Table 5 shows that the droplet diameter-to-nozzle diameter ratio D/D_n was within the range of 1.56 to 2.62. This agreed with Merrington and Richardson results (12).

The dimensionless droplet spacing for the maximum-flow-rate case was found to have a mean value of 7.54; for the minimum flow rate, the mean value for λ/D_n was 4.83. Pimbley and Lee (13) concluded that it was least likely to have satellite formation if λ/D_n fell between 5 and 7. The data obtained confirmed their findings.

6. CONCLUSIONS

1. A set of data was obtained which define the region of good droplet formation over a wide range of fluid properties and operating conditions.
2. Correlations of these data in the form of upper and lower bounds of good droplet formation have been developed.
3. A unique catcher-stream design which can be used with trichloroethylene (TCE) was devised. The TCE remained as a moving liquid film without collapsing into a single thread.
4. Experimental observations of dimensionless droplet spacing and ratio of droplet-to-nozzle diameter were found to agree well with previous studies.

7. RECOMMENDATIONS

1. The computer-derived correlations should be tested with data obtained from the internal-gelation-process pilot plant in Bldg. 3541, ORNL.
2. The catcher-stream design should be further tested and modified.
3. Develop a physical model for the droplet formation process.

8. ACKNOWLEDGMENTS

The authors thank Paul Haas, Al Ryon, and Walt Bond for their guidance. Special thanks are given to Vic Fowler, Rex Moody, and Wiley Johnston for their technical assistance.

9. APPENDIX

9.1 Newtonian-Jet Breakup

Theories have been established (6) to predict the breakup length of low-speed laminar jets formed by a Newtonian fluid in stagnant air. The breakup length L , which is the coherent portion of the jet, is often expressed as a function of jet velocity. The average velocity in the nozzle V is used in almost all correlations because it can be readily obtained. The velocity of the jet surface is zero immediately on emerging from the nozzle. The velocity profile relaxes, and the jet diameter may slightly expand or contract, depending on the fluid properties. For these reasons, the velocity of jet surface is more difficult to define and use in correlations.

The general shape of the breakup curve is shown in Fig. 16. At very low jet velocity, no jet is formed, and the liquid drips out of the nozzle. This accounts for the curved segment ABC. The linear portion of the curve (points ACD) was first described by Weber's analysis and later confirmed by others (6). The jet becomes turbulent beyond point F in Fig. 16. For our investigation, however, only the linear portion ACD will be examined further.

Weber derived the following result:

$$\frac{L}{D} = C_1 We^{\frac{1}{2}} (1 + 3Z) \quad (4)$$

where $Z = We^{\frac{1}{2}}/Re$, called the Ohnesorge number, was found to be a useful parameter in measuring the relative importance of viscosity and surface tension in controlling stability. It was found that jets are more stable at large Ohnesorge numbers (i.e., high viscosity and low surface tension). The constant C_1 has an experimental value of 12 (6). Equation (4) is only valid for the linear segment (ACD) on the breakup curve and does not account for the effect of ambient pressure. A modification of Weber's theory, which predicts the entire breakup curve, is available (6). In the same study, two other conclusions are worth mentioning. Over the linear portion of the L - V curve: (1) the first break in the jet, or the breakup length, is always well-defined; (2) the velocity profile of the jet has no effect on the propagation of the disturbance.

There are two minimum physical requirements for droplets to form. First, a certain pressure is necessary before a stream can be emitted from the nozzle. In this case, liquid drips out from the nozzle under gravity force. (The minimum pressure to form a liquid stream is about 1 cm Hg for our experimental system. This minimum pressure requirement is always exceeded for all practical purposes.)

MASSACHUSETTS INSTITUTE OF TECHNOLOGY
SCHOOL OF CHEMICAL ENGINEERING PRACTICE
AT
OAK RIDGE NATIONAL LABORATORY

THE GENERAL SHAPE OF THE BREAKUP CURVE
FOR A NATURAL JET

DATE
9-22-81

DRAWN BY
JCC

FILE NO.
SEPS-X-337

FIG.
16

Secondly, there must be enough room in the stream for drops to form. Consider a segment of the stream which would eventually become a single drop. If the frequency is too high or the droplet spacing (which is defined as the center-to-center distance between two drops) is too short, it can be shown that the diameter of the drop that contains this volume of liquid would be larger than the spacing. Droplets will overlap one another, making it impossible to have steady-state drop formation. The wavelength for which the drop diameter just equals the wavelength is given by:

$$\frac{\pi D_j^2 \lambda}{4} = \frac{\pi}{6} \lambda^3 \quad (5)$$

Therefore,

$$\frac{\lambda}{D_j} = \sqrt{1.5} = 1.2247 \quad (6)$$

Drop formation is impossible for spacings less than 1.2247 diameters of the stream.

When the liquid jet breaks into droplets, the fluid spheres oscillate about their equilibrium shape with a characteristic natural frequency. For two low viscosity fluids bounded by a free interface, the natural frequency is given by (10, 11):

$$\beta_I = \beta^* - \frac{(2n+1)^2 \sqrt{\beta^* \mu_i \mu_o \rho_i \rho_o}}{2\sqrt{2} R \Gamma [\sqrt{\mu_i \rho_i} + \sqrt{\mu_o \rho_o}]^2} \quad (7)$$

where

$$\beta^* = \sqrt{\frac{\sigma n(n+1)(n-1)(n+2)}{R^3 \Gamma}} \quad (8)$$

and

$$\Gamma = \rho_o n + \rho_i (n+1) \quad (9)$$

Fluid properties, as well as the droplet size, enter into the expression for β_I . The parameter n usually assumes a value of 2, which is the principle mode of oscillation. Equations (7) through (9) are confirmed to within 10% (10, 11). A drop falling through another fluid is different

from a solid sphere in that it may be deformed, and it will have a circulation setup within itself by the shearing effect of the relative motion of the two fluids. These are among the initial sources of disturbance which can cause the droplets to oscillate.

Another characteristic parameter for droplets from narrow jets is called the limiting velocity v_0 . Below this limiting velocity and for a given fluid, the drops will be uniform in size. In fact, Merrington and Richardson (12) found that for fine nozzles (below 1-mm diam) the constant drop size reached at low velocities is roughly twice the nozzle diameter. The ratio of droplet diameter D to jet diameter D_j has a mean value of 2.1 for nozzle diameters ranging from 0.30 to 0.65 mm. When jets issue from larger nozzles at low speeds, the drops are no longer uniformly twice the diameter of the nozzle. This limiting velocity v_0 is originally given as:

$$\frac{\mu}{\sqrt{\rho \sigma D_n}} = 2000 \left(\frac{\mu}{v_0 \rho D_n} \right)^{4/3} \quad (10)$$

Equivalently,

$$v_0 = 299 \left(\frac{\mu^2 \sigma^3}{\rho^5 D_n^5} \right)^{1/8} \quad (11)$$

To summarize, some characteristics of a natural Newtonian jet, such as the breakup length, are well-known. There are physical limitations for droplets to form from a jet, such as a minimum pressure and a minimum spacing required. A liquid drop has a characteristic natural frequency of vibration when it is disturbed. Also, a limiting velocity exists for droplets from a narrow jet. On the other hand, only a limited amount of literature focused on a liquid jet that has an imposed oscillation. In particular, the factors that affect the uniformity of droplets formed this way is largely unknown.

9.2 Accurate Calibration Curves

Calibration curves describing flow rate as a function of pressure upstream of the nozzle were developed for water and 20% aqueous glycerol for all nozzles. At several different flow rates, the droplet stream was divided into a beaker for a measured time and the contents were weighed. The volumetric flow rate was then calculated from the liquid density, and this measured mass flow rate. A typical example is given in Fig. 17. The remaining curves may be found in the calculation file.

- $D_n = 325 \mu\text{m}$
△ $D_n = 350 \mu\text{m}$
□ $D_n = 500 \mu\text{m}$
▽ $D_n = 586 \mu\text{m}$

MASSACHUSETTS INSTITUTE OF TECHNOLOGY
SCHOOL OF CHEMICAL ENGINEERING PRACTICE
AT
OAK RIDGE NATIONAL LABORATORY

FLOW RATE CALIBRATION FOR WATER

DATE
9-18-81

DRAWN BY
JCC

FILE NO.
CEPS-X-337

FIG.
17

9.3 Dimensional Analysis

A dimensional analysis was performed, in which the important dimensionless groups were identified as Reynolds number (Re), Weber number (We), Froude number (Fr), Power number (Po), and droplet spacing-to-nozzle diameter ratio (λ/D_n). The vibration frequency was defined as a function of the dimensional parameters as shown below:

$$f = F(\mu, \rho, \sigma, V, \lambda, D_n, P, g) \quad (12)$$

A power relationship was assumed:

$$f = F(\mu)^a (\rho)^b (\sigma)^c (V)^d (\lambda)^e (D_n)^g (P)^h (g)^k \quad (13)$$

and the dimensional units [in terms of mass, length, and time, {MLT}], of each of the parameters were then substituted into Eq. (13). This yielded:

$$\left(\frac{1}{T}\right) = F\left(\frac{M}{LT}\right)^a \left(\frac{M}{L^3}\right)^b \left(\frac{M}{T^2}\right)^c \left(\frac{L}{T}\right)^d (L)^e (L)^g \left(\frac{ML^2}{T^3}\right)^h \left(\frac{L}{T^2}\right)^k \quad (14)$$

Equating exponents of like terms gives:

$$L: 0 = -a - 3b + d + e + g + 2h + k \quad (15)$$

$$M: 0 = a + b + c + h \quad (16)$$

$$T: -1 = -a - 2c - d - 3h - 2k \quad (17)$$

Since there were eight unknowns, but only three equations to relate them, some assumptions were required to solve the system. Since we expect μ , σ , λ , and P to appear in separate numbers, we will derive preferred forms if we do not eliminate their exponents a , c , e , and h . Then,

$$\text{From Eq. (16): } b = -a - c - h$$

$$\text{From Eq. (17): } d = 1 - a - 2c - 3h - 2k$$

$$\begin{aligned} \text{From Eq. (18): } 0 = & -a - 3(-a - c - h) + (1 - a - 2c - \\ & 3h - 2k) + e + g + 2h + k \end{aligned}$$

So,

$$g = -a - c - e - 2h + k - 1 \quad (18)$$

Substituting this into Eq. (13) yields:

$$f = F(\mu)^a (\rho)^{-a-c-h} (\sigma)^c (V)^{1-a-2c-3h-2k} (\lambda)^e (D_n)^{-a-c-e-2h+k-1} (p)^h (g)^k \quad (19)$$

Grouping terms with like exponents gives:

$$\left(\frac{f D_n}{V}\right) = F\left(\frac{D_n V \rho}{\mu}\right)^{-a} \left(\frac{D_n V^2 \rho}{\sigma}\right)^{-b} \left(\frac{\lambda}{D_n}\right)^e \left(\frac{p}{\rho V^3 D_n^2}\right)^h \left(\frac{V^2}{D_n g}\right)^{-k} \quad (20)$$

Multiplying both sides by $\left(\frac{\lambda}{D_n}\right)^h$ and assuming that $V = f\lambda$ gives:

$$\left(\frac{f D_n}{V}\right) \left(\frac{\lambda}{D_n}\right)^h = F(Re)^{-a} (We)^{-b} \left(\frac{\lambda}{D_n}\right)^e (Fr)^{-k} \left(\frac{p}{\rho f^3 \lambda^2 D_n^3}\right)^h \quad (21)$$

or,

$$\left(\frac{\lambda}{D_n}\right)^{h-e-1} = F(Re)^{-a} (We)^{-b} (Fr)^{-k} (p_o)^h \quad (22)$$

Making the left-hand-side exponent equal to unity gives:

$$\frac{\lambda}{D_n} = F'(Re)^{\frac{a}{1+e-h}} (We)^{\frac{b}{1+e-h}} (Fr)^{\frac{k}{1+e-h}} (p_o)^{\frac{-h}{1+e-h}} \quad (23)$$

Finally,

$$\frac{\lambda}{D_n} = F' (Re)^A (We)^B (Fr)^C (p_o)^D \quad (24)$$

9.4 SAS Program Listing and Data Base

The following is a listing of the SAS computer program used and its data base:

```

//MIT001 JOB (20945), SAV66452,35 MII
//ROUTE PRINT PM145
//CLASS CPU33=103, IO=2
// EXEC SAS
//SAS.SYSIN DD *
=SASFIL.1
//
ENDINPUT

00100 DATA GLYCOL1;
00200 INPUT RUN NOZZLE DIAM FREQ QMIN LMIN;
00300 VISC=0.199;
00400 SURF=47.7;
00500 DENS=1.113;
00600 VEL=4.*QMIN/(3.1416*DIAM**2);
00700 RE=LOG(DIAM*VEL*DENS/VISC);
00800 WE=LOG(DIAM*VEL**2*DENS/SURF);
00900 FR=LOG(VEL**2/(980.*DIAM));
01000 SMIN=LOG(LMIN/DIAM);
01100 LIST;
01200 CARDS;
01300 8 4 0.0501 4169 1.635 0.1990
01400 10 4 0.0501 2979 1.193 0.2032
01500 12 4 0.0501 2400 0.919 0.1941
01600 14 4 0.0501 1677 0.644 0.1947
01700 15 4 0.0501 3633 1.385 0.1933
01800 17 2 0.0325 3802 0.387 0.1227
01900 19 2 0.0325 5410 0.523 0.1165
02000 21 2 0.0325 5550 0.593 0.1288
02100 23 2 0.0325 6500 0.695 0.1290
02200 25 3 0.0350 3900 0.572 0.1541
02300 27 3 0.0350 3729 0.689 0.1921
02400 29 3 0.0350 2900 0.796 0.2851
02500 31 3 0.0350 3887 0.379 0.1063

```

```

02600 DATA GLYCOL2;
02700 INPUT RUN NOZZLE DIAM FREQ QMAX LMAX;
02800 VISC=0.199;
02900 SURF=47.7;
03000 DENS=1.113;
03100 VEL=4.*QMAX/(3.1416*DIAM**2);
03200 RE=LOG(DIAM*VEL*DENS/VISC);
03300 WE=LOG(DIAM*VEL**2*DENS/SURF);
03400 FR=LOG(VEL**2/(980.*DIAM));
03500 SMAX=LOG(LMAX/DIAM);
03600 LIST;
03700 CARDS;
03800 9 4 0.0501 3271 1.631 0.2529
03900 11 4 0.0501 2439 1.175 0.2444
04000 13 4 0.0501 2004 0.917 0.2322
04100 14 4 0.0501 1677 0.644 0.1947
04200 16 4 0.0501 2906 1.365 0.2384
04300 18 2 0.0325 1846 0.457 0.2987
04400 20 2 0.0325 2956 0.497 0.2025
04500 22 2 0.0325 3254 0.610 0.2260
04600 24 2 0.0325 3550 0.698 0.2371
04700 26 3 0.0350 2085 0.578 0.2879
04800 28 3 0.0350 2174 0.687 0.3285
04900 30 3 0.0350 2830 0.797 0.2927
05000 32 3 0.0350 1664 0.434 0.2710
05100 DATA WATER1;
05200 INPUT RUN NOZZLE DIAM FREQ QMIN LMIN;
05300 VISC=0.01005;
05400 SURF=72.75;
05500 DENS=1.0;
05600 VEL=4.*QMIN/(3.1416*DIAM**2);
05700 RE=LOG(DIAM*VEL*DENS/VISC);
05800 WE=LOG(DIAM*VEL**2*DENS/SURF);
05900 FR=LOG(VEL**2/(980.*DIAM));
06000 SMIN=LOG(QMIN/DIAM);
06100 LIST;
06200 CARDS;
06300 39 4 0.0501 460 0.435 0.4797
06400 40 4 0.0501 710 0.366 0.2615
06500 42 4 0.0501 1000 0.410 0.2020
06600 44 4 0.0501 1200 0.508 0.2147
06700 46 4 0.0501 1400 0.671 0.2431
06800 48 4 0.0501 1550 0.790 0.2585
06900 49 3 0.0350 820 0.184 0.2332
07000 51 3 0.0350 1000 0.172 0.1788
07100 53 3 0.0250 1300 0.180 0.1439
07200 55 3 0.0350 1800 0.193 0.1114
07300 57 3 0.0350 2400 0.263 0.1139
07400 59 3 0.0350 3000 0.295 0.1622
07500 61 3 0.0350 3800 0.405 0.1108
07600 63 3 0.0350 4700 0.540 0.1194

```

07700	65	3	0.0350	5500	0.615	0.1162
07800	67	3	0.0350	6600	0.650	0.1024
07900	69	3	0.0350	7050	0.745	0.1098
08000	71	2	0.0325	800	0.150	0.2260
08100	73	2	0.0325	925	0.155	0.2020
08200	75	2	0.0325	1150	0.165	0.1730
08300	77	2	0.0325	1300	0.190	0.1762
08400	79	2	0.0325	1600	0.215	0.1620
08500	81	2	0.0325	1900	0.230	0.1459
08600	83	2	0.0325	3500	0.345	0.1188
08700	85	2	0.0325	5000	0.455	0.1097
08800	115	5	0.0586	575	0.400	0.2579
08900	117	5	0.0586	900	0.720	0.2966
09000	119	5	0.0586	1200	0.920	0.2843
09100	121	5	0.0586	1500	1.240	0.3065
09200	123	5	0.0586	1800	1.330	0.2740
09300	DATA WATER2;					
09400	INPUT RUN NOZZLE DIAM FREQ QMAX LMAX;					
09500	VISC=0.01005;					
09600	SURF=72.75;					
09700	DENS=1.0;					
09800	VEL=4.*QMAX/(3.1416*DIAM**2);					
09900	RE=LOG(DIAM*VEL*DENS/VISC);					
10000	WE=LOG(DIAM*VEL**2*DENS/SURF);					
10100	FR=LOG(VEL**2/(980.*DIAM));					
10200	SMAX=LOG(LMAX/DIAM);					
10300	LIST;					
10400	CARDS;					
10500	39	4	0.0501	460	0.435	0.4797
10600	41	4	0.0501	710	0.640	0.4573
10700	43	4	0.0501	1000	0.710	0.3602
10800	45	4	0.0501	1200	0.690	0.2917
10900	47	4	0.0501	1400	0.810	0.2935
11000	48	4	0.0501	1550	0.790	0.2585
11100	50	3	0.0350	820	0.265	0.3359
11200	52	3	0.0350	1000	0.335	0.3482
11300	54	3	0.0350	1300	0.420	0.3358
11400	56	3	0.0350	1800	0.560	0.3234
11500	58	3	0.0350	2400	0.680	0.2945
11600	60	3	0.0350	3000	0.765	0.2650
11700	62	3	0.0350	3800	0.845	0.2311
11800	64	3	0.0350	4700	0.910	0.2012
11900	66	3	0.0350	5500	0.865	0.1162
12000	68	3	0.0350	6600	0.915	0.1441
12100	70	3	0.0350	7050	0.920	0.1356
12200	72	2	0.0325	800	0.200	0.3014
12300	74	2	0.0325	925	0.215	0.2802
12400	76	2	0.0325	1150	0.295	0.3092
12500	78	2	0.0325	1300	0.395	0.3663
12600	80	2	0.0325	1600	0.355	0.2675
12700	82	2	0.0325	1900	0.430	0.2728
12800	84	2	0.0325	3500	0.745	0.2566
12900	86	2	0.0325	5000	0.940	0.2266

```

13000 116 5 0.0586 575 0.720 0.4643
13100 118 5 0.0586 900 1.230 0.5067
13200 120 5 0.0586 1200 1.510 0.4666
13300 122 5 0.0586 1500 1.810 0.4474
13400 124 5 0.0586 1800 1.930 0.3976
13500 DATA GLYCER1;
13600 INPUT RUN NOZZLE DIAM FREQ QMIN LMIN;
13700 VISC=0.01769;
13800 SURF=72.4;
13900 DENS=1.069;
14000 VEL=4.*QMIN/(3.1416*DIAM**2);
14100 RE=LOG(DIAM*VEL*DENS/VISC);
14200 WE=LOG(DIAM*VEL**2*DENS/SURF);
14300 FR=LOG(VEL**2/(980.*DIAM));
14400 SMIN=LOG(LMIN/DIAM);
14500 LIST;
14600 CARDS;
14700 90 2 0.0325 880 0.283 0.3877
14800 91 2 0.0325 1200 0.170 0.1708
14900 93 2 0.0325 1650 0.205 0.1498
15000 95 2 0.0325 2900 0.290 0.1205
15100 97 2 0.0325 3700 0.445 0.1450
15200 98 2 0.0325 5000 0.495 0.1193
15300 99 2 0.0325 6300 0.630 0.1205
15400 100 3 0.0350 825 0.203 0.2558
15500 101 3 0.0350 1250 0.270 0.2245
15600 103 3 0.0350 2100 0.295 0.1460
15700 105 3 0.0350 3600 0.570 0.1646
15800 106 3 0.0350 2800 0.413 0.1533
15900 107 3 0.0350 4200 0.545 0.1349
16000 108 4 0.0501 780 0.535 0.3479
16100 109 4 0.0501 1150 0.795 0.3507
16200 111 4 0.0501 1900 0.940 0.2510
16300 113 4 0.0501 1400 0.770 0.2790
16400 DATA GLYCER2;
16500 INPUT RUN NOZZLE DIAM FREQ QMAX LMAX;
16600 VISC=0.01769;
16700 SURF=72.4;
16800 DENS=1.069;
16900 VEL=4.*QMAX/(3.1416*DIAM**2);
17000 RE=LOG(DIAM*VEL*DENS/VISC);
17100 WE=LOG(DIAM*VEL**2*DENS/SURF);
17200 FR=LOG(VEL**2/(980.*DIAM));
17300 SMAX=LOG(LMAX/DIAM);
17400 LIST;
17500 CARDS;
17600 90 2 0.0325 880 0.283 0.3877
17700 92 2 0.0325 1200 0.375 0.3767
17800 94 2 0.0325 1650 0.535 0.3909
17900 96 2 0.0325 2900 0.767 0.3188
18000 100 3 0.0350 825 0.203 0.2558
18100 102 3 0.0350 1250 0.340 0.2827
18200 104 3 0.0350 2100 0.625 0.3093
18300 108 4 0.0501 780 0.535 0.3479
18400 110 4 0.0501 1150 0.995 0.4389
18500 112 4 0.0501 1900 0.995 0.2656
18600 114 4 0.0501 1400 0.855 0.3098

```

18700 DATA ALLMIN:
18800 SET GLYCOL1 WATER1 GLYCER1:
18900 DATA ALLMAX:
19000 SET GLYCOL2 WATER2 GLYCER2:
19100 DATA POWERMIN:
19200 INPUT RUN POWER:
19300 LIST:
19400 CARDS:
19500 8 2.03
19600 10 2.34
19700 12 3.21
19800 14 4.41
19900 15 2.57
20000 17 1.84
20100 19 1.58
20200 21 1.34
20300 23 1.40
20400 25 3.85
20500 27 3.96
20600 29 3.52
20700 31 2.16
20800 39 0.13
20900 40 6.24
21000 42 8.45
21100 44 8.22
21200 46 7.61
21300 48 7.35
21400 71 0.27
21500 73 0.56
21600 75 1.16
21700 77 0.23
21800 79 3.60
21900 81 0.89
22000 83 2.11
22100 85 3.77
22200 90 2.59
22300 91 1.98
22400 93 2.46
22500 95 2.74
22600 97 2.55
22700 98 2.65
22800 99 2.93
22900 100 0.02
23000 101 0.07
23100 103 4.8
23200 105 3.26
23300 106 0.13
23400 107 2.03
23500 108 2.17
23600 109 0.64
23700 111 3.78

```

23800 113 5.22
23900 115 0.58
24000 117 0.32
24100 119 0.25
24200 121 1.57
24300 123 0.46
24400 PROC SORT DATA=ALLMIN;
24500 BY RUN;
24600 PROC SORT DATA=POWERMIN;
24700 BY RUN;
24800 DATA FINALMIN;
24900 MERGE ALLMIN POWERMIN;
25000 BY RUN;
25100 PD=LOG(POWER/(DENS*FREQ**3*LMIN**2*DIAM**3));
25200 PROC GLM;
25300 MODEL SMIN=RE WE FR PD;
25400 OUTPUT OUT=LOWER PREDICTED=PSMIN RESIDUAL=RECID;
25500 PROC PRINT;
25600 PROC PLOT DATA=LOWER;
25700 PLOT RESID*SMIN=NOZZLE PSMIN*SMIN=NOZZLE SMIN*PSMIN=NOZZLE;
25800 DATA POWERMAX;
25900 INPUT RUN POWER;
26000 LIST;
26100 CARDS;
26200 9 1.58
26300 11 2.10
26400 13 2.39
26500 14 4.41
26600 16 2.13
26700 18 3.73
26800 20 1.79
26900 22 2.45
27000 24 2.48
27100 26 5.70
27200 28 2.38
27300 30 2.68
27400 32 2.81
27500 39 0.13
27600 41 0.03
27700 43 0.11
27800 45 0.42
27900 47 7.59
28000 48 7.35
28100 72 0.30
28200 74 0.83
28300 76 4.00
28400 78 0.53
28500 80 0.13

```

```

28600 82 0.20
28700 84 0.58
28800 86 3.67
28900 90 2.59
29000 92 0.07
29100 94 1.51
29200 96 0.33
29300 100 0.02
29400 102 0.06
29500 104 0.09
29600 108 2.17
29700 110 0.32
29800 112 3.75
29900 114 2.24
30000 116 0.03
30100 118 0.04
30200 120 0.03
30300 122 0.08
30400 124 0.27
30500 PROC SORT DATA=ALLMAX;
30600 BY RUN;
30700 PROC SORT DATA=POWERMAX;
30800 BY RUN;
30900 DATA FINALMAX;
31000 MERGE ALLMAX POWERMAX;
31100 BY RUN;
31200 PD=LOG(POWER/(DENS*FREQ**3*LMAX**2*DIAM**3));
31300 PROC GLM;
31400 MODEL SMAX=RE WE FR PD;
31500 OUTPUT OUT=UPPER PREDICTED=PSMAX RESIDUAL=RESID;
31600 PROC PRINT;
31700 PROC PLOT DATA=UPPER;
31800 PLOT RESID*SMAX=NOZZLE PSMAX*SMAX=NOZZLE SMAX*PSMAX=NOZZLE;
31900 DATA FINAL;
32000 MERGE FINALMIN FINALMAX;
32100 BY RUN;
32200 PROC PRINT DATA=FINAL;

```

9.5 Sample Calculations

The following sample calculations are based on the data of run 101.

Total pressure upstream of the nozzle is given by:

$$\begin{aligned}
 P_{\text{total}} &= P_{\text{air}} + P_{\text{static head}} \\
 &= [(\text{cm Hg})(13.6) + (\text{level})(2.54)(\rho_{\text{glycerol-water}})] \frac{98.07}{1000} \\
 &= [(4.9)(13.6) + (6.5)(2.54)(1.0469)] \frac{98.07}{1000} \\
 &= 8.23 \text{ kPa}
 \end{aligned}$$

Droplet diameter is given by (18):

$$D = \left(\frac{6 Q}{\pi f} \right)^{1/3} = \left(\frac{6 (0.27)}{\pi (1250)} \right)^{1/3} = 744 \text{ } \mu\text{m}$$

The natural frequency of oscillation of the droplets is calculated using Eqs. (7) through (9) of Appendix 9.1. For 20% glycol, substituting the values for all the constants into Eq. (7), this reduces to:

$$\beta_I = 66.5029 \left(\frac{6}{R^3} \right)^{1/2} - \frac{0.3124}{R} \left(\frac{6}{R^3} \right)^{1/2}$$

and $R = 0.0372 \text{ cm}$ or $D = 0.0744 \text{ cm}$. Therefore,

$$\beta_I = 3262 \text{ Hz}$$

The velocity of the jet is given by:

$$V = \frac{Q}{S} = \frac{Q}{\left(\frac{\pi D_n^2}{4} \right)} = \frac{4 Q}{\pi D_n^2} = \frac{(4)(0.27)}{(\pi)(0.0350)^2} = 280.63 \text{ cm/s}$$

The droplet spacing is given by:

$$\lambda_{\text{calculated}} = \left(\frac{V}{f}\right)(10) = \frac{280.63}{1250}(10) = 2.245 \text{ mm}$$

and

$$\lambda_{\text{observed}} = \frac{25.4}{N_d - 1} = \frac{25.4}{14 - 1} = 1.954 \text{ cm}$$

where

N_d = number of drops observed in 1-in. length of jet

9.6 Location of Original Data

The original data are located in ORNL Databook A-9185-G, pp. 1-56, on file at the MIT School of Chemical Engineering Practice, Bldg. 1505, ORNL. A copy of the original data, a calculation file, and a literature file are also on file.

9.7 Nomenclature

A, B, C, D, E, F, F'

a, b, c, d, e, g, h, k empirical constants

D droplet diameter, cm or μm

D_j jet diameter, cm or μm

D_n nozzle diameter, cm or μm

E voltage, volt

f frequency, Hz

g gravitational acceleration, cm/s

I current, amp

HMTA hexamethylenetetramine

L length

M mass

N number of orifices

N_d number of drops, drops/in.

n principle model of oscillation, usually $n=2$

p	pressure, kPa
P	power, W
Q	flow rate, cm^3/s
R	droplet radius, cm or μm
S	orifice cross-sectional area, cm^2
SRP	Savannah River Plant of the Department of Energy
t	time, s
T	time
TCE	trichloroethylene
V	liquid jet linear velocity, cm/s
v_0	limiting jet velocity, cm/s

Greek Symbols

β_I	natural frequency, Hz
β^*	frequency correction in Eq. (8), Hz
Γ	density weighting factor in Eq. (9), g/cm^3
$\lambda_{\text{obs}}, \lambda_{\text{calc}}$	observed and calculated center-to-center droplet spacing, cm or μm
ρ	density, kg/m^3
μ	viscosity, Pa-s
σ	surface tension, N/m
π	3.14159...

Dimensionless Groups

Fr	Froude number (V^2/gD_n)
Po	Power number ($P/\rho f^3 \lambda^2 D_n^3$)
Re	Reynolds number ($D_n V \rho / \mu$)
We	Weber number ($D_n V^2 \rho / \sigma$)

λ/D_n dimensionless droplet spacing

D/D_n diameter ratio

9.8 Literature References

1. Advanced Dynamic Instruments, "Installation and Operating Instructions, 25 Watt Solid State Oscillator/Amplifier," Serial No. 293, Dec. 18, 1980.
2. Beer, J.M., and N.A. Chigier, Combustion Aerodynamics, pp. 165-161, Wiley, New York, 1972.
3. Bond, W.D., personal communication, "Contribution to Alternative Waste Forms Progress Report," Aug. 25, 1981.
4. Cline, H.E., and T.R. Anthony, "The Effect of Harmonics on the Capillary Instability of Liquid Jets," J. Appl. Phys., 49(6), 3202 (1978).
5. Grace, J.R., T. Wairegi, and J. Brophy, "Break-Up of Drops and Bubbles in Stagnant Media," Can. J. Chem. Engr., 56, 3 (February 1978).
6. Grant, R.P., and S. Middleton, "Newtonian Jet Stability," AIChE J., 12(4), 669 (1966).
7. Haas, P.A., "Formation of Liquid Drops with Uniform and Controlled Diameters at Rates of 10^3 and 10^5 Drops Per Minute," AIChE J., 21, 383 (1975).
8. Haas, P.A., personal communication, "Formation of Gel Particles," Aug. 18, 1981.
9. Haas, P.A., J.M. Begovich, A.D. Ryon, and J.S. Vavruska, "Chemical Flowsheet Conditions for Preparing Urania Spheres by Internal Gelation," I&EC Prod. Res. Dev., 19, 459 (1980).
10. Loshak, Jonathan, "Forced Oscillation of Drops in a Viscous Medium," M.S. Thesis, Univ. of Rochester, Rochester, NY, 1969.
11. Loshak, Jonathan, and C.H. Byers, "Forced Oscillation of Drops in a Viscous Medium," Chem. Eng. Sci., 28, 149 (1973).
12. Merrington, A.C., and E.G. Richardson, "The Break-Up of Liquid Jets," Proc. Phys. Soc. London, 59, 1 (1947).
13. Pimbley, W.T., and H.C. Lee, "Satellite Droplet Formation in a Liquid Jet," IBM J. Res. Dev., 21, 21 (Jan. 1977).
14. Rayleigh, Lord, "On the Instability of Jets," Proc. London Math. Soc., 10, 4 (1878).

15. SAS Users's Guide, 1979 Edition, SAS Institute, Inc., Box 8000, Cary, N.C. 27511.

16. Stone, J.A., S.T. Goforth, and P.K. Smith, "Preliminary Evaluation of Alternative Forms for Immobilization of Savannah River Plant High-Level Waste," DP-1545 (December 1979).

17. Tiegs, S.M., P.A. Haas, and R.D. Spence, "The Sphere-Cal Process: Fabrication of Fuel Pellets from Gel Microspheres," ORNL/TM-6906 (Sept. 1979).

18. Williams, D.F., and C.C. Haws, "Design of and Operating Procedures for an Interim Facility for Preparing Kilogram Quantities of UO_2 Spheres by Internal Gelation," ORNL/TM-6950 (1980).