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SUBJECT: Study of Operating Characteristics of a Rotating Bowl for Production of Microspheres

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ABSTRACT

A study was made of the operating characteristics of a rotating bowl for the production of uniform microspheres. The system was investigated using first water, and later a modeling fluid which approximated the properties of a nitrate-stabilized urania sol. Previously reported data (8) on similar bowl systems indicated very good uniformity of droplet diameters within a plume projected from the bowl, but the results of this study did not confirm such performance, at least in the case of water. It was found that droplet diameter varied about 50% of the mean in the total spray and about 20% of the mean in the plume for the water tests. The modeling fluid was found to give better uniformity of droplet size across the plume, the average diameter variation from the plume mean being around 11% or less. Droplet size was found to depend directly on the flow rate, inversely on the angular velocity and bowl diameter. The droplet size data were successfully correlated by dimensional analysis. It was found that from 70 to 90% of the collected spray was within a band of fairly uniform drop size. However, an overall mass balance accounted for 70% or less of the feed. These results indicate that if the device is used for the production of microspheres, that some recycling of fluid will be necessary.

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1. SUMMARY

Studies were conducted on a rotating bowl disperser to assess its usefulness for the production of microspheres having diameters of 150 to 500 μ in the ORNL Sol-Gel Process. Plexiglas bowls 3.4 and 5.08 cm in diameter with nozzle diameters of 254 and 635 μ were rotated with frequencies of 3000 to 12,000 rpm. The fluids used in the experiments were water, a surfactant solution, and a modeling fluid approximating the properties of a urania sol. Droplet size was determined from photographs of drops collected in silicone oil on vertical slides close to the bowl and in horizontal petri dishes some distance from the bowl.

Droplet size was found to vary about 50% of the mean in the total spray. However, from 70 to 90% by weight of the recovered fluid was found in a band of fairly uniform drop size. Droplet size varied about 20% of the mean within this band. Mass balances on the recovered droplets accounted for 70% or less of the feed fluid.

Mean droplet size in the band was correlated dimensionally with a driving force number, the ratio of radial to tangential momentum of the droplet at the edge of the bowl. The correlation held fairly well for various combinations of angular velocities, bowl diameter, nozzle diameter, flow rates, viscosities, densities, and surface tensions of the fluids.

In addition to extension of this study to actual sols, further studies on the effect of suspected droplet coalescence and on the reason for nonclosure of the mass balance are recommended. Further studies are also recommended on the possibility of recycle of droplets falling outside of the limits of the band.

2. INTRODUCTION

2.1 Background

Ceramic microspheres of the oxides, carbides, and nitrides of thorium, uranium, plutonium, zirconium, and mixtures of the above are being developed at Oak Ridge National Laboratory as candidate materials for fuels in high temperature gas cooled reactors. The fuel material is first converted to an aqueous sol, dispersed into small droplets, gelled in an immiscible fluid, dried, sintered, and finally pyrolytically coated with carbon. Two-fluid nozzles, vibrating capillaries, spinning disk atomizers, and rotary dispersers have been investigated (13) as apparatus for dispersing the microspheres. Each of these methods involves one or more of the following disadvantages in the production of small (50 to 500 μ in diameter) microspheres: (1) low capacity, (2) poor droplet uniformity, and (3) poor variability over the range of droplet size. These three points are then to be considered when examining ways to produce microspheres.

Roberts and Devemy, in an earlier study (13) of possible ways for improving microsphere production techniques, recommended a study of the rotating bowl. The rotating bowl developed by E.N. Brown (8) was described as capable of producing uniform droplets (5 to 10% deviation from the mean) of any desired size between 80 and 400 μ at throughputs up to 26 cc/min. With this background, the present study was undertaken to examine the characteristics of a rotating bowl for production of microspheres.

Brown used a plexiglas bowl which was rotated at high speed about a vertical axis. A hole or nozzle was drilled in the side of the bowl, and liquid fed into the top of the bowl was accelerated through the nozzle and was thrown out of the bowl in a horizontal stream of drops. Brown's bowl was 3.6 cm diameter with one nozzle of about 635 μ diameter in the wall. He presented data giving the dependence of the droplet size on the angular velocity and the flow rate into the bowl. He concluded that the droplet size varies inversely with the angular velocity, and directly with the flow rate into the bowl. He also stated that the effect of the hole size on the droplet diameter was not significant up to a critical 400 μ drop size. He concluded also that the drop size should be inversely proportional to the bowl diameter, in agreement with earlier work done by Walton and Prewett (14) using a rotating disk disperser. Brown stated that his best results were obtained using small diameter holes, low liquid flow rates, and high angular velocities. Brown has, since his original study, done more work on the bowls, using bowls with more than one hole, and reports using up to 32 holes in a bowl without adversely affecting drop size distributions (9). Brown did not study effects of the properties of fluids other than water.

2.2 Method of Approach

The first step in this study was to find the drop size as a function of angular velocity of the bowl, flow rate into the bowl, bowl diameter, and nozzle diameter. An attempt was made to generate data comparable to those of Brown (8) under similar conditions, especially to reproduce his narrow drop size distributions. The second step was to try to determine how the sol itself might be dispersed by the bowl. One sol material of current interest is an aqueous urania dispersion of the approximate chemical formula $U(NO_3)_{1.5}(COOH)_{0.3}(OH)_{1.5}$. The important properties of the fluid with respect to droplet production are thought to be the viscosity, the surface tension, and the density. The urania sols presented too many safety difficulties to be used directly in the study; so a modeling fluid which approximated these fluid properties of the sol was tested.

3. APPARATUS AND PROCEDURE

The bowls used in this study (Fig. 1) were made of plexiglas, similar to those of Brown (8). Two bowl diameters, 1.325 and 2.000 in. were tested with holes of two diameters, 254 and 635 μ. The bowl was driven by an

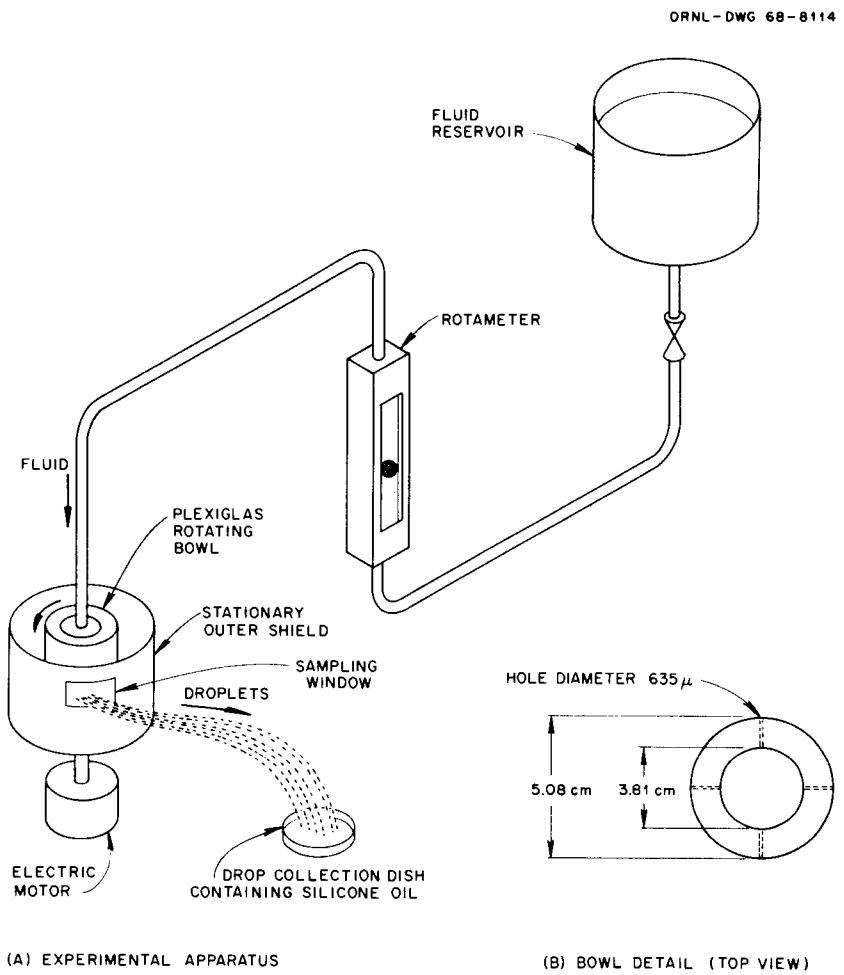

Fig. 1. Experimental Apparatus

electric motor capable of 3000 - 12,100 rpm. Feed was introduced to the center of the bowl at a controlled rate, 4.8 or 10.1 cc/min per hole. The spray was sampled through a window in the cylindrical shield enclosing the apparatus. A summary of experimental conditions is shown in Table 1.

Droplets issuing from the sampling window were collected in two ways - on coated slides or petri dishes. The petri dishes, containing a thin layer of silicone oil covered by a layer of kerosene, were placed on a horizontal surface 5.5 in. below and 4 to 40 in. away from the sampling window. The droplets impinged on the layer of kerosene and sank to the surface of the silicone oil where they assumed a nearly spherical shape. The layer of kerosene minimized evaporation. Droplets were also collected on slides coated with silicone oil held just outside the sampling window at a slight angle to the vertical. An electric timer was used for measuring the exposure times for mass balance calculations.

As soon as a sample of the droplets was taken, the petri dish or slide was placed on a microscope stage under a Polaroid camera and lighted from beneath. Magnification of 10 to 24x was achievable with the 36 mm lens used. The droplet sizes were measured from the photographs with millimeter rulers or with a Zeiss semiautomatic drop counting device available through the courtesy of T.E. Wilmarth of ORNL.

Three fluids were tested in the rotating bowl - water (the bulk of the experiments), a surfactant solution (KNOX-60 laboratory glass cleaner in water), and a modeling solution containing sucrose and zinc chloride in water, the properties of which approximated those of an ORNL urania sol. Properties of the three solutions are compared to the properties of a urania sol in Table 2.

4. RESULTS

4.1 General Observations

The droplets in these experiments always formed a plume which fell on the collection area as a band as reported by Brown (8). The distance of the band from the bowl never exceeded 36 in. in a horizontal direction at a distance of 15 cm below the holes in the bowl. The determination of the bandwidth is somewhat subjective, depending on what the experimentalist chooses to call the band and where he judges the band to lie. The bands described in this paper were found by placing paper towels on a table 15 cm below the bowl, and observing a dark band on the towels due to the wetting by droplets. The width of the band appeared to be smallest in the modeling fluid tests and in tests with the small bowl diameter. Bandwidth did not correlate strongly with angular velocity or flow rate, but smaller bands were generally obtained at higher angular velocities and lower flow rates. The flow from the holes of the bowl always appeared to be in the form of a jet when observed with a strobe light, but it was not certain that the flow was not many droplets which appeared to be a jet.

Table 1. Summary of Experiments

Test No.	Fluid	Bowl Diameter (in.)	Hole Size (microns)	Flow Rates (cc/min)	Angular Velocity (rpm)	Purpose
SCA 1-8	water	2.00	635	10.1	3000	Drop size distribution across a band
SCB 1-20	water	2.00	635	10.1	3000	Drop size and weight distribution over entire spraying area
SCC	water	2.00	635	10.1	3000	Drop size and weight distribution across a band
BC 1-11	water	2.00	635	4.8,10.1	3000 - 12,000	Tests for effect of angular velocity and flow rate
BDC 1-5	water	1.325	635	4.8,10.1	3000, 12,000	Test for effect of bowl diameter
HC 1-8	water	2.00	254	4.8,10.1	3000, 12,000	Test for effect of hole diameter
SC 1-2	surfactant solution	2.00	635, 254	10.1	3000	Test for effect of surfactant
SBC 1-5	modeling solution	1.325	635	4.8,10.1	3000, 12,000	Test of model solution
SGCM-1	modeling solution	1.325	635	4.8	3000	Weight distribution over entire spraying area for model solution
SGCM-2	modeling solution	1.325	635	4.8	12,000	Weight collected in band for modeling fluid

Table 2. Fluid Properties
(Temperature 25°C)

Fluid	Density (gm/cc)	Viscosity (centipoise)	Surface Tension (dynes/cm)
water	0.99804	0.8937	71.97
surfactant solution	-	-	40.5
Modeling fluid	1.36	2.87	79
Tested urania sol	1.2-1.3	2-5	72.6

Physical properties of water are from Ref. (13).

Urania sol densities and viscosities are from Ref. (12). Modeling fluid properties determined by analytical laboratory (surface tension at 22°C).

It is possible to operate the rotating bowl such that there is an annulus of liquid within the inside wall of the bowl. The bowl may also be operated under "dry bowl" conditions, in which there is no annulus. A comparison was made, using the same angular velocity, bowl diameter and hole size for two different flow rates, one sufficient to produce an annulus, and one which resulted in a dry bowl. The test with the annulus produced a much larger drop size, as would be expected, but the uniformity of the droplets was comparable. In two other cases, the flow rate into the bowl was held constant while the angular velocity was lowered to the point at which an annulus appeared in the bowl. In general, one could say that the operation of the bowl "full" does not produce drops which are significantly more or less uniform than those produced with a dry bowl. It would seem, then, that increasing the capacity of the device with higher flow rates is not detrimental to the uniformity.

4.2 Droplet Size Distribution

Droplet size distributions for the total spray area are given in Figs. 2 and 3. Figure 2 presents droplet sizes measured from a slide held just outside the cylinder which surrounded the rotating bowl. Thus, it might be expected to give a good sample of the total range of droplets produced by the device. The second distribution was obtained by collecting drops horizontally on a table 15 cm below the holes in the bowl. Figure 3 shows the droplet size distribution as a function of distance from the center of the bowl for the conditions of an angular velocity of 3000 rpm, a flow rate per hole of 10.1 cc/min, with a bowl of 2 in. outer diameter and 635 μ hole size. Mean droplet diameters and the standard deviation expressed as percent of the mean droplet diameter for all tests are presented in Appendix 8.1.

4.3 Mass Distributions and Mass Balances over Spraying Area

For two sets of conditions studied, one using water and one using the modeling fluid, the complete weight distribution in the spray area was found. The results are presented in Figs. 4 and 5. In addition, complete mass balances for these two cases have been calculated and are given in Table 3. For two other sets of conditions, mass balance calculations for the band area only were made and are presented in the same table. Another test was made by gathering the droplets at the edge of the metal cylinder surrounding the bowl. In this test, 98% of the predicted weight of water was found in the receiving container.

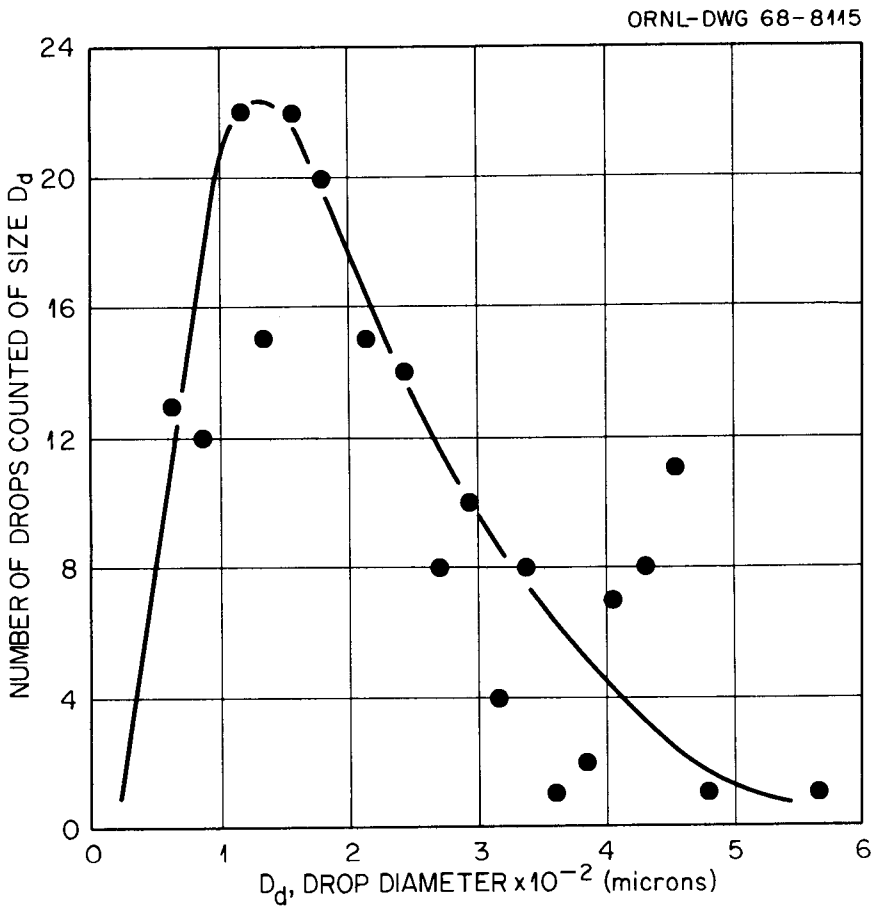

Fig. 2. Water Drop Size Distribution for a Sample Taken Just Outside the Bowl

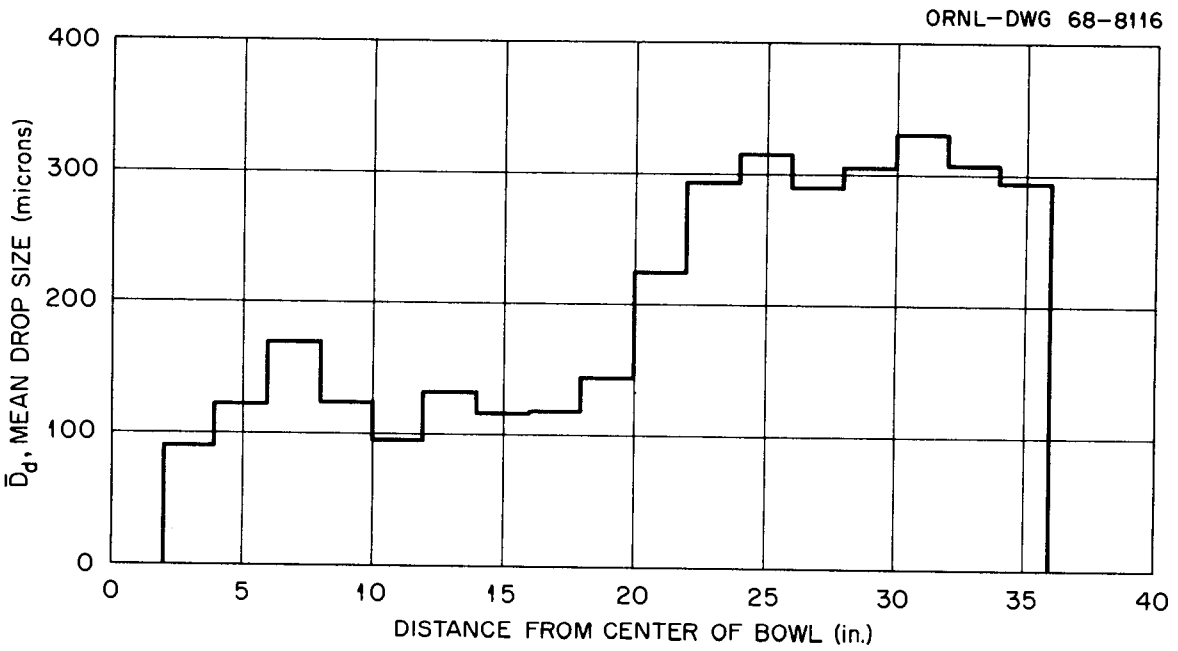

Fig. 3. Water Drop Size as a Function of Distance from Center of Bowl (SCB Tests)

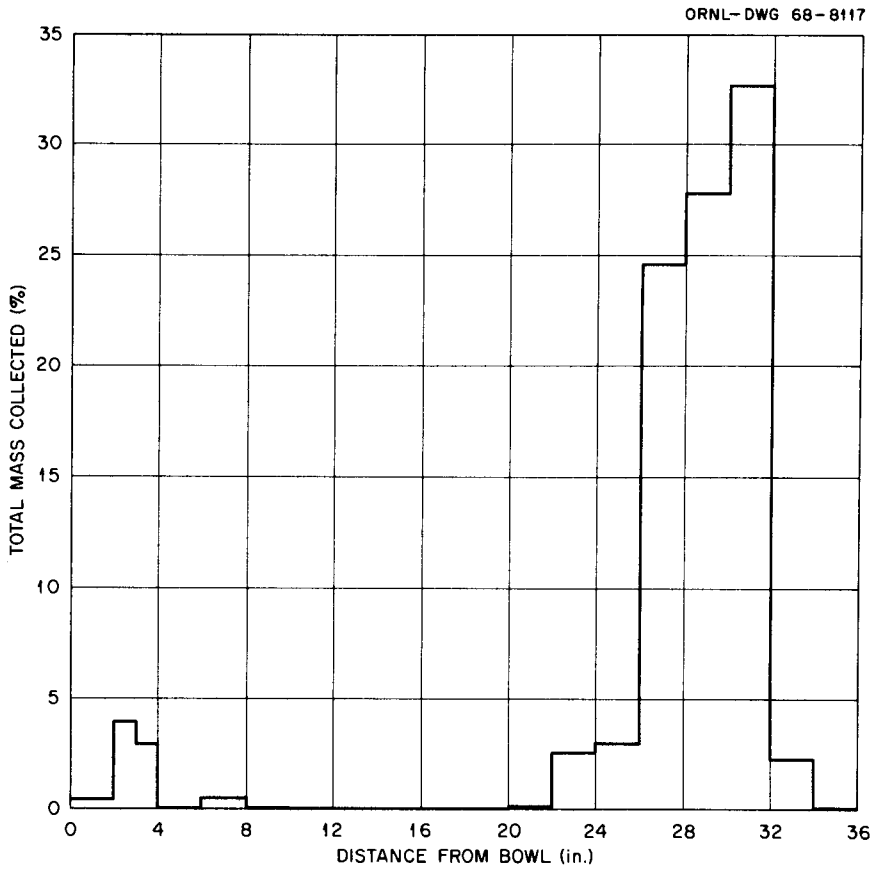

Fig. 4. Mass Distribution, Water Experiment SCB 1-20

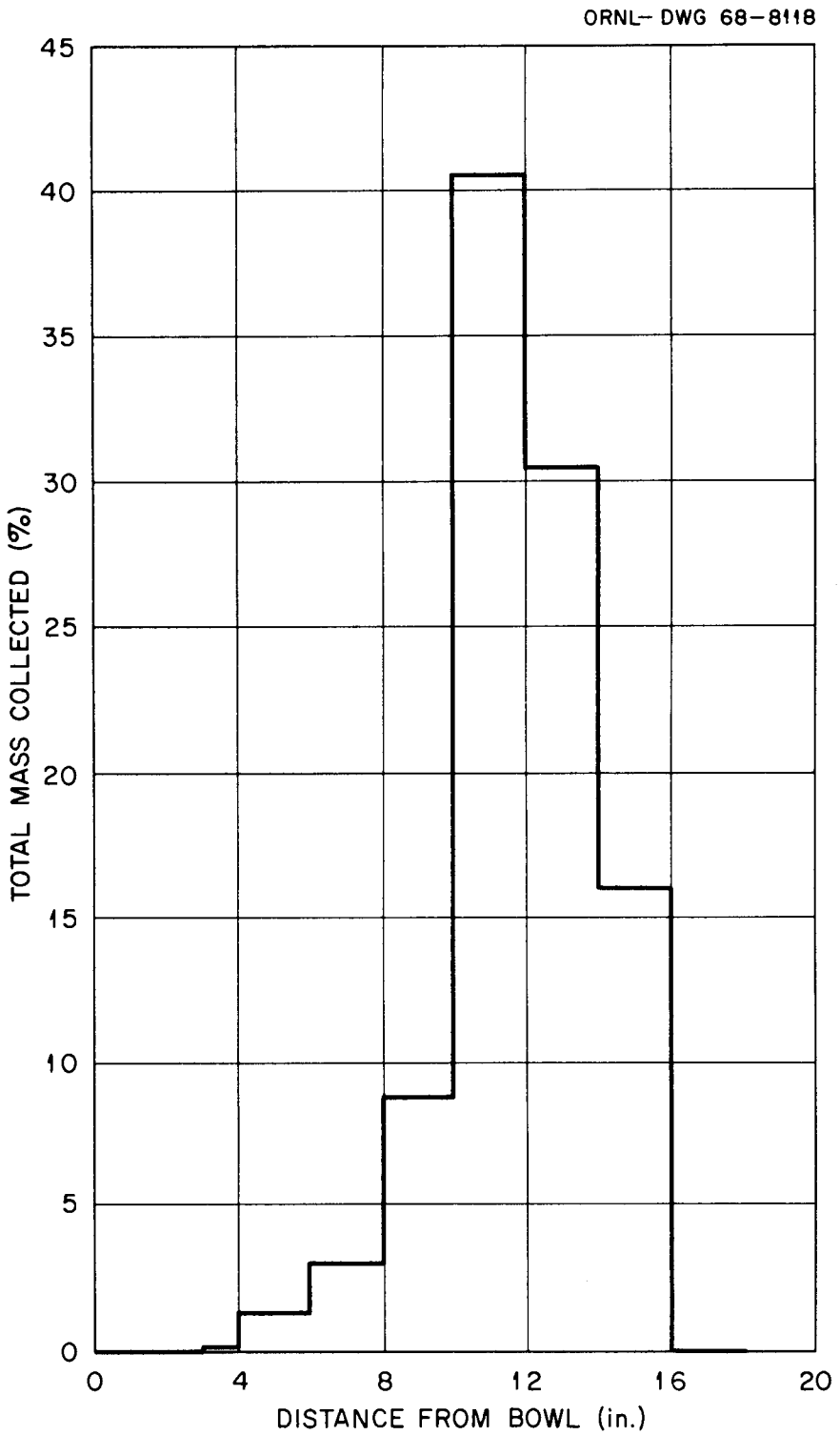

Fig. 5. Mass Distribution, Modeling Fluid, Experiment SGCM-1

Table 3. Mass Balance Results

Test No.	Fluid Tested	Angular Velocity (rpm)	Flow Rate Per Hole (cc/min)	Bowl Diameter (in.)	Total Weight Recovered, Percent of Flow Rate	Total Weight in Band, Percent of Flow Rate	Weight in Band, Percent of Total Weight Recovered
SCC	water	3000	10.1	2	-	57.4	-
SCB	water	3000	10.1	2	72.2	88.0	93.1
SGC	model	3000	4.8	1.34	54.5	38.9	71.2
SGC	model	12,100	4.8	1.34	-	39.0	-

Hole diameter for all cases is 635 μ.

4.4 Effects of Angular Velocity, Flow Rate, Bowl Diameter and Hole-Size on Mean Droplet Diameter

The experimental apparatus available made possible the investigation of the effects of angular velocity, flow rate, bowl diameter and hole size. Data obtained for tests made at conditions used by Brown in his tests are shown in Fig. 6. Table 4 shows the variation of drop size with bowl diameter.

Table 4. Effect of Bowl Diameter on Estimated Mean Droplet Size in Band

Test No.	Bowl Diameter (in.)	Rotational Speed (rpm)	Flow Rate (cc/min)	Mean Drop Diameter (micron)	Standard Deviation from Mean (Percent of Mean Diameter)
BDC 1-2	1.34	3000	10.1	415	16
BDC 3-4	1.34	12,100	10.1	185	14
SCB 12-16	2.00	3000	10.1	311	16
BC 7-8	2.00	12,100	10.1	212	12

A dimensional analysis on liquid jets breaking up into droplets, suggested by Roberts and Devemy (13), involved a correlation of droplet size to three dimensionless groups: (1) Reynolds number, Re, a ratio of inertial force to viscous shear, (2) the Ohnesorge number, Z_j, which includes surface, inertial, and viscous forces, and (3) the rotational driving force number, F_ω, a ratio of rotational to inertial forces.

$$F_\omega = \frac{\omega^2 x_o D_N}{V^2} \qquad (1)$$

where:

F_ω = rotational driving force number, dimensionless

ω = angular velocity, radians/sec

x_o = bowl diameter, cm

D_N = nozzle diameter, cm

V = velocity of fluid through nozzle, cm/sec

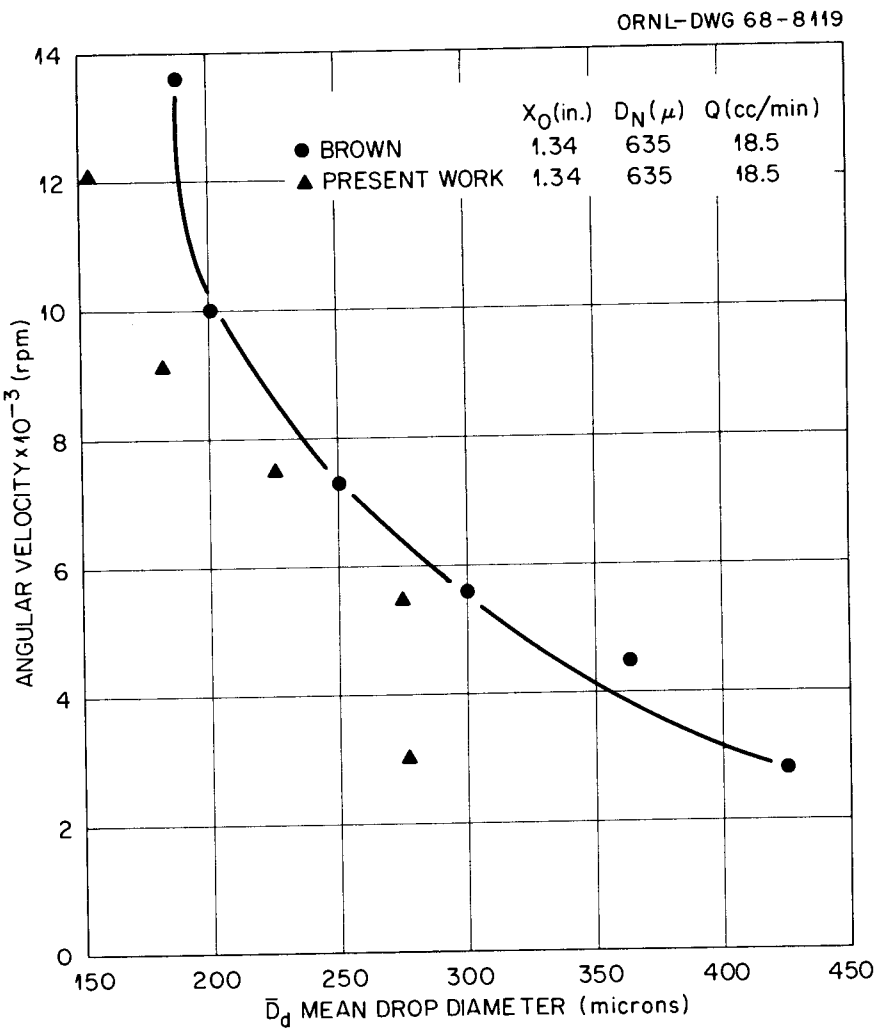

Fig. 6. Comparison of Present Work and Data of Brown

The quantity V is related to the flow rate, Q, by V = Q/area of hole.

In this study a correlation with a modified driving force number was found to give better agreement. The modified driving force is a ratio of tangential to radial momentum, and is defined by the equation:

$$F'_{\omega} = \left(\frac{\omega\ x_o}{V}\right) \qquad (2)$$

The correlation of all experimental data as outlined in Table 5 can be seen in Fig. 7. Plotted against F'^2_{ω} is the dimensionless mean droplet size, $\overline{D}_d/D_N$, the ratio of the mean droplet size sampled in the band to the diameter of the hole in the bowl.

4.5 Model Solution and Surfactant Test Results

A modeling fluid approximating the properties (see Table 2) of the urania sol was used in several experiments. A comparison of the mean droplet size for the tests using the modeling fluid with those for water under the same conditions is shown in Table 6. Since many sols of interest have surface tension considerably lower than that of water, two tests were made using a surfactant in water. The results of these tests are shown in Table 7.

5. DISCUSSION OF RESULTS

5.1 Droplet Size Distribution

Droplet size distributions over the range of the total water spray are presented in Figs. 2 and 3. Figure 2 gives the distribution for a silicone coated slide held vertically outside the sampling window of the device. The mean of this distribution is about 190 μ and the standard deviation is about 90 μ. Clearly the droplet size distribution would not be uniform enough for production of fuel microspheres. Consequently, some type of sorting and recycle procedure for the droplets would be necessary. The easiest way to effect the sorting might be to let the different drag characteristics of the particles separate them as they are projected outward.

Figure 3 shows the variation of mean droplet diameter as a function of the distance outward from the center of the bowl for the case of the 2 in. bowl at 3000 rpm and a flow rate of 10.1 cc/min. The droplet size distribution over the whole range varies widely, as is indicated by the results in Fig. 2, but it is interesting to note that the mean droplet diameters within the band between 22 and 36 in. from the bowl have much less spread.

Table 5. Legend for Figure 7

Run Number	Symbol	Flow Rate Per Hole (cm^3/min)	Speed at Which Annulus Formed (rpm)	Bowl Diameter x_o (cm)	Hole Diameter D_N (micron)	Viscosity μ (cp)	Density ρ (g/cm^3)	Surface Tension σ (dynes/cm)
SCA,SCB,BC(1-9)	○	10.1		5.08	635	1.0	1.0	72.6
SCC,BC(10-15)	■	4.8		5.08	635	1.0	1.0	72.6
HC(1-4)	□	10.1	3000	5.08	254	1.0	1.0	72.6
HC(5-7)	△	4.8		5.08	254	1.0	1.0	72.6
HC 8	◊	5.85	2500	5.08	254	1.0	1.0	72.6
SO 1	●	10.1		5.08	254	1.0	1.0	40.1
SO 2	▲	10.1		5.08	635	1.0	1.0	40.1
SGC(1-2)	♦	10.1	3000	3.4	635	2.87	1.346	80.14
SGC(3-5)	▽	4.8	3000	3.4	635	2.87	1.346	80.14
B_2C(1-12)	▼	18.5		3.4	635	1.0	1.0	72.6
BDC(1-5)	✚	10.1	1560	3.4	635	1.0	1.0	72.6
FB(1-4)	✜	10.1	680	5.08	635	1.0	1.0	72.6
Data of Brown (8)	+	18.5		3.6	711	1.0	1.0	72.6
Data of Brown (8)	✳	6.6		3.6	711	1.0	1.0	72.6
Data of Brown (8)	★	6.6		3.6	635	1.0	1.0	72.6
Data of Brown (8)	☆	18.5		3.6	635	1.0	1.0	72.6

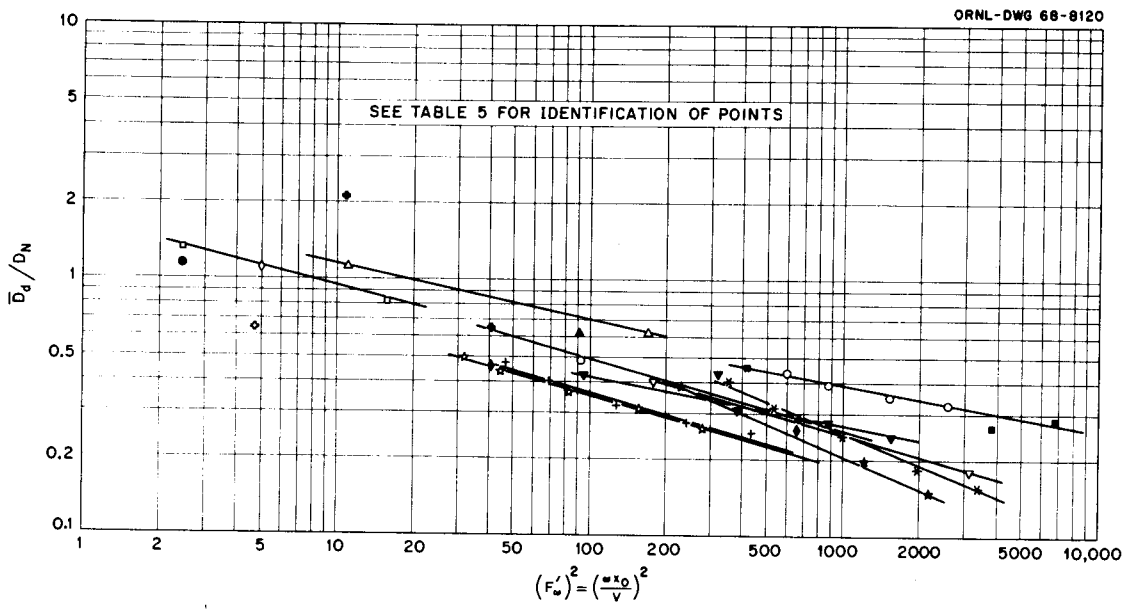

Fig. 7. Dimensionless Drop Size vs Driving Force Number

Table 6. Results of Modeling Solution Tests. Estimated Mean Diameter Within Band

Test No.	Fluid	Angular Velocity (rpm)	Flow Rate (cc/min)	Mean Diameter $\overline{D}_d$ (micron)	Standard Deviation from Mean (Percent of Mean Diameter)
SGC 1	model	3000	10.1	293	3
SGC 2	model	12,100	10.1	167	8
SGC 5	model	3000	4.8	258	12
SGC 3-4	model	12,100	4.8	113	6
BDC 1-2	water	3000	10.1	415	16
BDC 3-4	water	12,100	10.1	185	14

Table 7. Effect of Surface Tension

Fluid	Test No.	Hole Size D_N (micron)	Mean Drop Diameter $\overline{D}_d$ (micron)	Standard Deviation from Mean (Percent of Mean Diameter)
surfactant	SO 1	254	294	6
surfactant	SO 2	635	396	12
water	HC 3-4	254	333	29
water	SCB 12-14	635	311	15

For the data in Fig. 3, 93% of the mass collected fell in the band. The mean diameter of those droplets falling in the band was about 315 μ, and the standard deviation was about 45 μ, or about 15% of the mean. This is significantly higher than the 5-10% variation indicated by Brown (8). However, Brown's distributions were apparently measured only in the center of the band (9).

The lack of uniformity of the water drops produced is rather discouraging from the viewpoint of practical application of the device. Of particular note, however, are the results of a few tests made with the modeling fluid near the end of experimentation. For these tests (see Table 6) the highest value of the standard deviation within the band was 12% of the mean, and that value was for a very narrow band (2 in.). The rest of the standard deviations fell below 8% of the mean with narrow bands. Thus, the results for the modeling solution show significantly better droplet uniformity within the bands. About 71% of the total weight collected fell within the band for the test with modeling solution. Since the few results with the fluid whose properties approximate those of urania sols were more encouraging than the results with water, we recommend further investigation using real sols.

It was noticed during the course of the tests that two droplets sometimes coalesced on contact. The drop size distributions for a given sample from the band are fairly uniform; most drops are within 5% of the mean. However, in most samples a small percentage of larger drops appeared. Nearly all of the larger drops were about the size of a drop that would be formed by coalescence of two smaller drops (1.26 times the mean diameter). Obviously, droplet coalescence should be prevented or taken into consideration more fully in future studies.

5.2 Mass Distribution and Mass Balances

Information on the droplet distribution by mass throughout the spraying area is presented in Figs. 4 and 5 and Table 3. For the experiment SCB 1-20, Fig. 4, 93% of the total mass collected in the spraying area is at distances exceeding 22 in. For experiment SGCM-1, using the modeling solution, Fig. 5, 87% of the mass was found at distances exceeding 10 in., and about 71% was within the band from 10 to 14 in.

The data from the calculated mass balances summarized in Table 3 are disturbing. In the two complete mass balances made, the percent recoveries (based on the total feed rate to the bowl) were only 54.5% and 72.2%. The higher recovery is for a test using water, the lower using the modeling fluid.

One possible reason for the nonclosure of the balance is evaporation of the fluid. A calculation considering evaporation losses is presented in Appendix 8.2. From this calculation it appears that evaporation could contribute significantly to the nonclosure of the mass balance. However, the second complete mass balance, using the modeling fluid, should be expected

to close more completely if evaporation were the major reason for loss, because the zinc chloride and sucrose used in the modeling fluid should lower the vapor pressure of water considerably. However, closure of the material balance was worse with the modeling fluid than with water; only about half of the modeling fluid was collected. Possibly many of the droplets were carried away by air currents in the room. A test was made collecting the droplets just outside the rim of the bowl in a tared cup. It was found that the mass balance closed within 1.5%.

An error analysis showed that the calculation of the mass balance could be in error as much as 20% if all errors were maximum and additive, but this does not provide a very satisfactory explanation.

5.3 Effect of Angular Velocity, Flow Rate, Bowl Diameter and Hole Diameter on Mean Droplet Size

From Fig. 6 and Table 4 it can be seen that droplet size varied inversely with the angular velocity. This effect was found by Brown (8) in his investigation. At low speed the drop size decreased with increased bowl diameter, but in our work at high speed the drop size was largely unchanged when the bowl diameter was increased.

The only difference in the apparatus used in Brown's work and that used in this study is the angle at which the hole is bored into the bowl. Brown's holes were drilled on a secant line, while in this work the holes were radial. A comparison of the data of Brown at the same flow rate, bowl diameter, hole size, and over the same range of angular velocities, shown in Fig. 6, shows the same shape of curve, except for the test at 3000 rpm, which produced a droplet size 64% below that given by Brown. The curve generated in these experiments also was to the left of that of Brown, indicating smaller droplet size for the same conditions. The comparison of the physical characteristics of the two bowls is sufficiently close that the results of the experiments should be the same.

We initially believed from Brown's work that the hole diameter would not have a very great effect. However, our correlation of $F_\omega'^2$ and $\overline{D}_d/D_N$ seems moderately good and is obviously influenced by hole diameter. The correlation, given in Fig. 7 for conditions listed in Table 5, shows fairly straight lines of slope - 0.25 for tests of the present work. It is important to note that Fig. 7 correlates data in which ω, X_o, V, and D_N were all changed. Note that the driving force number F_ω' is the ratio of the tangantial to radial fluid momenta. Two points at the left side of the figure which deviate widely from the correlation were made at very low angular velocities. Drop uniformity was found to be poor at such low rpm, in agreement with findings of Brown (8). Data for one flow rate used by Brown (also shown on Fig. 7) agree fairly well with data of the present work, but his data at low flow rate (6.6 cc/min) give a slope of -0.40.

5.4 Results with the Model Solution and Surfactant Solution

No difficulties were experienced in the shift from water to modeling solution; the operation of the bowl seemed qualitatively the same as that for water. The same conclusions with respect to the effect of flow rate and angular velocity on mean droplet size were drawn as for the water cases. As can be noted in Table 6, the droplet size was somewhat smaller for the modeling solution for the same set of operating conditions at low speed, but at high speed, the results for water and the modeling fluid are about the same.

The tests for the surfactant solutions, shown in Table 7, indicate that the hole diameter has a significant effect on the droplet size. The mean droplet size of 294 μ for the 254 μ hole case is within the measurement error of 333 μ, the value for water, but the mean for the 635 μ hole case differs significantly from the corresponding value for water, 311 μ. The effect of surfactant is not clear; further investigation is needed here.

Problems encountered in this investigation - nonclosure of mass balance, evaporation, and droplet coalescence - might not be present in experiments with a real sol. If the sol droplets were caught in gelating solution, gelled, and sized when solid, a more straightforward assessment of the applicability of the rotating bowl to the ORNL Sol-Gel Process would be possible.

6. CONCLUSIONS AND RECOMMENDATIONS

6.1 Conclusions

1. The rotating bowl has been found to produce droplets in the 90 to 400 μ range with high throughputs.

2. The data indicate the drop uniformity produced by the rotating bowl to be less than that reported by Brown. A possible reason for the poor uniformity is the coalescence of drops.

3. The drop producing performance of the bowl correlated by dimensionless groups, agrees with Brown's findings (8) that the drop size varies directly with the flow rate and inversely with bowl diameter and angular velocity.

4. For the modeling solution drop uniformity was improved.

6.2 Recommendations

Because of the high throughputs obtained while producing droplets in the 90 to 400 μ range, the rotating bowl is attractive in spite of the rather poor drop size uniformity with water. Further experiments should be performed using real sols instead of water and modeling fluids. Problems of sampling, drop coalescence, and evaporation might be eliminated if the sol droplets were gelled and the rigid microspheres sized. If further experiments with water or modeling solution are attempted, the above difficulties must be faced.

7. ACKNOWLEDGEMENTS

We wish to thank our consultant, P.A. Haas, for his help and advice. Thanks also go to S.D. Clinton for helping us with the photographic methods and to the Analytical Chemistry Division for their fast service.

8. APPENDIX

8.1 Summary for all Tests

In Table 8 are presented the test conditions and the results for all experiments conducted.

8.2 Sample Calculation of Evaporation Rate from Droplets

The rate of mass transfer from a droplet is given by

$$N = k_x (x_i - x_\infty)$$

where:

N = flux of water by evaporation per unit interfacial area, mole/cm^2-sec

k_x = mass transfer coefficient based on mole fractions, mole/cm^2-sec

x_i = mole fraction water in the air at the interface, dimensionless

x_∞ = mole fraction water in the air far away from the droplets, dimensionless

The mass transfer coefficient can be found from correlation in Bird, Stewart, and Lightfoot (6) as:

$$k_x = \frac{cD^*}{D_d}\left[2.0 + 0.60\left(\frac{D_d\, v}{\nu}\right)^{1/2}\left(\frac{\nu}{D^*}\right)^{1/3}\right] \qquad (3)$$

where:

D_d = diameter of drop, cm

c = molar density of air, mole/cm^3

D^* = diffusivity of water in air, cm^2/sec

v = droplet velocity, cm/sec

ν = kinematic viscosity of air, cm^2/sec

Table 8. Data Summary for All Tests

Test No.	Fluid	Bowl Diameter X_o (in.)	Hole Diameter D_N (microns)	Flow Rate Q $(\frac{cc}{min\text{-}hole})$	Rotational Speed ω (rpm)	Band Width (in.)	Sample Distance (in.)	Mean Drop Diameter $\bar{D}_d$ (microns)	Standard Deviation from Mean (Percent of Mean)
SCA-6	water	2.00	635	10.1	3000	20-26	20-21	354	13.8
SCA-7	water	2.00	635	10.1	3000	20-26	21-22	280	4.9
SCA-8	water	2.00	635	10.1	3000	20-26	22-23	389	7.4
SCA-9	water	2.00	635	10.1	3000	20-26	23-24	341	7.65
SCA-10	water	2.00	635	10.1	3000	20-26	24-25	358	16.0
SCA-11	water	2.00	635	10.1	3000	20-26	25-26	329	2.8
SCA-12	water	2.00	635	10.1	3000	20-26	26-27		
SCA-13	water	2.00	635	10.1	3000	20-26	27-28		
SCB-1-4	water	2.00	635	10.1	3000	24-32	1-4	90.0	69.2
SCB-5	water	2.00	635	10.1	3000	24-32	4-6	127.5	48.3
SCB-6	water	2.00	635	10.1	3000	24-32	6-8	153	21.7
SCB-7	water	2.00	635	10.1	3000	24-32	8-10	138	33.6
SCB-8	water	2.00	635	10.1	3000	24-32	12-14	78.0	34.8
SCB-9	water	2.00	635	10.1	3000	24-32	10-12	97.9	27.0
SCB-10	water	2.00	635	10.1	3000	24-32	14-16	107.5	38.6
SCB-11	water	2.00	635	10.1	3000	24-32	16-18	106.5	51.9
SCB-12	water	2.00	635	10.1	3000	24-32	18-20	145	28.2
SCB-13	water	2.00	635	10.1	3000	24-32	20-22	227	12.0
SCB-14	water	2.00	635	10.1	3000	24-32	22-24	296	27.0
SCB-15	water	2.00	635	10.1	3000	24-32	24-26	312	28.6
SCB-16	water	2.00	635	10.1	3000	24-32	26-28	289	27.1
SCB-17	water	2.00	635	10.1	3000	24-32	28-30	300	15.5
SCB-18	water	2.00	635	10.1	3000	24-32	30-32	349	9.1
SCB-19	water	2.00	635	10.1	3000	24-32	32-34	310	10.5
SCB-20	water	2.00	635	10.1	3000	24-32	34-36	293	24.4
SCC-1	water	2.00	635	10.1	3000	12-26	20-22	138	9.4
SCC-2	water	2.00	635	10.1	3000	12-26	22-24	159	10.6
SCC-3	water	2.00	635	10.1	3000	12-26	24-26	171	17.8
SCC-4	water	2.00	635	10.1	3000	12-26	26-28	169	5.8
SCC-5	water	2.00	635	10.1	3000	12-26	28-30	164	18.8
SCC-6	water	2.00	635	10.1	3000	12-26	21-23	148	17.6
SCC-7	water	2.00	635	10.1	3000	12-26	27-29	199	12.7
SCC-10	water	2.00	635	10.1	3000	12-26	16-18	110	17.3
SCC-11	water	2.00	635	10.1	3000	12-26	18-20	143	13.3
BDC-1	water	1.325	635	10.1	3000	17-21	21-23	408	14.9
BDC-2	water	1.325	635	10.1	3000	17-21	23-25	423	17.1
BDC-3	water	1.325	635	10.1	12,100	15.5-18.5	19.5-21.5	182	20.8
BDC-4	water	1.325	635	10.1	12,100	15.5-18.5	20.5-22.5	189	8.25
BDC-5	water	1.325	635	10.1	1560	4-6.5	8-10	1410	40.3

Table 8. (Continued)

Test No.	Fluid	Bowl Diameter X_o (in.)	Hole Diameter D_N (microns)	Flow Rate Q $(\frac{cc}{min\text{-}hole})$	Rotational Speed (rpm)	Band Width (in.)	Sample Distance (in.)	Mean Drop Diameter $\bar{D}_d$ (microns)	Standard Deviation from Mean (Percent of Mean)
HC-1	water	2.00	254	10.1	12,100	16.5-27.5	20.5-22.5	1410	10.3
HC-2	water	2.00	254	10.1	12,100	16.5-27.5	29.5-31.5	176	11.7
HC-3	water	2.00	254	10.1	3000	16.5-22.5	20.5-22.5	241	2.4
HC-4	water	2.00	254	10.1	3000	16.5-22.5	24.5-26.5	303	6.1
HC-5	water	2.00	254	4.8	3000	13-15	17-19	362	28.6
HC-6	water	2.00	254	4.8	12,100	11-25	17-19	282	38.0
HC-7	water	2.00	254	4.8	12,100	11-25	23-25	139	10.95
HC-8	water	2.00	254	4.8	2500	14.5-16.5	18.5-20.5	178	19.2
SO-1	surfactant	2.00	635	10.1	3000	16-19.5	21-23	294	6.3
SO-2	surfactant	2.00	254	10.1	3000	22.5-25.5	23.5-25.5	396	12.2
SGC-1	model	1.325	635	10.1	3000	15-16.5	14.5-16.5	293	2.5
SGC-2	model	1.325	635	10.1	12,100	17-19	17-19	167	7.8
SGC-3	model	1.325	635	4.8	12,100	6-10.5	6-8	113	4.4
SGC-4	model	1.325	635	4.8	12,100	6-10.5	8.5-10.5	121	5.8
SGC-5	model	1.325	635	4.8	3000	11-13	11-13	258	11.6
BC-1	water	2.00	635	10.1	9100	24-35	24-26	236	19.7
BC-2	water	2.00	635	10.1	9100	24-35	28-30	290	25.6
BC-3	water	2.00	635	10.1	9100	24-35	33-35	304	17.7
BC-4	water	2.00	635	10.1	7500	25.5-32	25-27	290	21.5
BC-5	water	2.00	635	10.1	7500	25.5-32	27.5-29.5	271	4.2
BC-6	water	2.00	635	10.1	7500	25.5-32	30-32	286	5.4
BC-7	water	2.00	635	10.1	12,100	24-31	24-26	212	12.0
BC-8	water	2.00	635	10.1	12,100	24-31	27-29	251	8.9
BC-9	water	2.00	635	10.1	3000	24-31	30-32	247	13.6
BC-10	water	2.00	635	10.1	3000	14.5-18.5	14.5-16.5	174	17.9
BC-11	water	2.00	635	10.1	3000	14.5-18.5	16.5-18.5	182	24.7
BC-12	water	2.00	635	4.8	7500	18-22	18-20	213	12.2
BC-13	water	2.00	635	4.8	7500	18-22	20-22	209	21.6
BC-14	water	2.00	635	4.8	3000	16.5-20.5	16.5-18.5	287	3.8
BC-15	water	2.00	635	4.8	3000	16.5-20.5	18.5-20.5	293	9.1
B2C-1	water	1.325	635	18.5	3000	7.5-9.5	7.5-9.5	275	6.65
B2C-3	water	1.325	635	18.5	3000	8.5-10.5	8.5-10.5	272	12.3
B2C-4	water	1.325	635	18.5	3000	8-9.5	8-10	277	14.1
B2C-5	water	1.325	635	18.5	3000	8-10	8-10	238	8.9
B2C-6	water	1.325	635	18.5	3000	7-8.5	6.5-8.5	221	6.65
B2C-8	water	1.325	635	18.5	9100	12-14	12-14	180.6	5.7
B2C-9	water	1.325	635	18.5	12,100	10-14	11-13	157.5	12.9
B2C-10	water	1.325	635	18.5	7500	9-14	11-13	198	3.8
B2C-11	water	1.325	635	18.5	5500	9-11.5	9.5-11.5	275	9.4
B2C-12	water	1.325	635	18.5	3000	26-28.5	26-28	736	30.1

For a stationary drop in stagnant air, v = 0, and Eq. (3) reduces to:

$$k_x = \frac{2cD^*}{D_d} \tag{4}$$

We shall consider evaporation from a 100 μ drop moving at 3180 cm/sec, the tangential velocity at the rim of a bowl of radius 1 in. rotating at 12,100 rpm. The temperature in the room is 26°C and the relative humidity is 50%. At this temperature the vapor pressure of water is 25.209 mm of mercury, or 3.32×10^{-2} atm (15).

Therefore,

$$x_i = 3.32 \times 10^{-2}$$

$$x_\infty = 1.66 \times 10^{-2} \text{ at 50\% relative humidity}$$

From the ideal gas law:

$$c = \frac{P}{RT} = \frac{1 \text{ atm}}{83 \frac{\text{cm}^3\text{-atm}}{\text{mole-°K}}(299°\text{K})} = 4.08 \times 10^{-5} \text{ mole/cm}^3$$

Also,

$$D^* = 0.25 \text{ cm}^2\text{/sec (10)}$$

$$\nu = 0.15 \text{ cm}^2\text{/sec (7)}$$

$$v = 3180 \text{ cm/sec}$$

$$k_x = \frac{(2)(4.08 \times 10^{-5})(0.25)}{10^{-2}}\left[2.0 + 0.60\left(\frac{(10^{-2})(3180)}{0.15}\right)^{1/2}\left(\frac{0.15}{0.25}\right)^{1/3}\right]$$

$$= 1.09 \times 10^{-2} \text{ mole/cm}^2\text{-sec}$$

and

$$N = (1.09 \times 10^{-2})(3.32 \times 10^{-2} - 1.66 \times 10^{-2})$$

$$= 1.81 \times 10^{-4} \text{ mole/cm}^2\text{-sec}$$

A 100 μ drop has a surface area of 3.14×10^{-4} cm^2, so that the rate of evaporation per droplet is

$$(3.14 \times 10^{-4}\ cm^2)(1.81 \times 10^{-4}\ mole/cm^2\text{-}sec) = 5.69 \times 10^{-8}\ mole/sec$$

The amount of water in the droplet is

$$5.24 \times 10^{-7}\ g = 2.9 \times 10^{-8}\ moles$$

If the droplet traveled 60 cm to the collection bowl at a constant velocity of 3180 cm/sec, it would have traveled for 0.0187 sec and would have lost

$$(5.69 \times 10^{-8}\ moles/sec)(1.87 \times 10^{-2}\ sec) = 1.06 \times 10^{-9}\ moles$$

or about 1/30 of its mass. Some additional evaporation might take place while the droplet was on the collection plate before being photographed.

On the basis of 100 μ drop size, evaporation could perhaps contribute to the non-closure of the balance, if the drops were exposed to air of 50% humidity and temperature 26% for only one or two seconds. However, it is difficult to understand the observed non-closure of the sol (with lower vapor pressure) mass balance in terms of evaporation.

8.3 Calculations for Droplets Appearing to be Coalesced

Ratio of diameters for two drops of volumes v and 2v is

$$\frac{\sqrt[3]{2}}{\sqrt[3]{1}} = 1.26$$

For test SO-2 the droplet sizes counted are (in millimeters at 24X magnification):

Drop No.	Size	Drop No.	Size
1	9.0	10	7.3
2	9.0	11	9.4
3	9.0	12	11.0
4	8.2	13	9.7
5	8.9	14	11.0
6	8.9	15	9.4
7	9.0	16	9.5
8	11.3	17	9.0
9	12.0		

$\overline{D}_d = 9.51$ mm

Standard deviation is 12.2% of mean.

Notice that drops number 8, 9, 12, and 14 appear abnormally large. If one deletes these droplets, the mean becomes 8.95 mm, and the standard deviation 6.95%. Multiplying 8.95 by 1.26, one obtains 11.3 mm for the size of a coalesced droplet. This value is close to the measured values for drops 8, 9, 12, and 14, and so the assumption of droplet coalescence seems reasonable.

8.4 Nomenclature

c molar density of air, moles/cm^3

D^* diffusivity of water vapor in air, cm^2/sec

D_d droplet diameter, cm or μ as noted

$\overline{D}_d$ mean droplet diameter, μ

D_N nozzle diameter, cm or μ as noted

F_ω rotational driving force number $= \dfrac{\omega^2 x_o D_N}{V^2}$, dimensionless

F'_ω modified driving force number $= \dfrac{\omega x_o}{V}$, dimensionless

k_x mass transfer coefficient based on mole fraction, moles/cm^2-sec

N flux of water by evaporation per unit droplet interfacial area, moles/cm^2-sec

Q flow rate to bowl, cm^3/min

Re Reynolds number $= \dfrac{V D_N \rho_j}{\mu_j}$, dimensionless

V velocity of fluid through nozzle, cm/sec

v droplet velocity in air, cm/sec

We Weber number $= V\sqrt{\dfrac{\rho_j D_N}{\sigma}}$, dimensionless

x_o bowl diameter, cm

x_i mole fraction water in the air at the water-air interface, dimensionless

x_∞ mole fraction water in the air free stream

Z_j Ohnesorge number of the jet $= \dfrac{\mu_j}{\sqrt{\rho_j D_N \sigma}}$

μ_j jet viscosity, gm/cm-sec

ν kinematic viscosity of air, cm^2/sec

ρ_j jet density, gm/cm^3

σ surface tension of jet, dynes/cm

ω angular velocity, radians/sec

8.5 Location of Original Data

The experimental data are recorded in ORNL Notebook A-5253-G, pp. 27-59, and Notebook A-5258-G, pp. 43-78, on file at the M.I.T. School of Chemical Engineering Practice, Bldg. 1000, ORNL.

8.6 Literature References

1. Applebey, M.P., "Viscosity of Aqueous Solutions of Weak Electrolytes and of all Organic Acids and Bases," "Internation Critical Tables of Numerical Data, Physics, Chemistry and Technology," E.M. Washburn, editor, Vol. V, p. 22, McGraw-Hill, London (1928).

2. Bearce, H.W., G.C. Mulligan, and M.P. Maslen, "Density of Certain Aqueous Organic Solutions," ibid., Vol. III, p. 116.

3. Bates, F.J., "Polarimetry, Saccharimetry and the Sugars, National Bureau of Standards Circular 440," pp. 627-29, United States Government Printing Office, Washington (1942).

4. Ibid., p. 671.

5. Bates, S.J., and W.P. Baxter, "Viscosity of Aqueous Solutions of Strong Electrolytes," in "International Critical Tables of Numerical Data, Physics, Chemistry, and Technology," E.M. Washburn, editor, Vol. V, p. 14, McGraw-Hill, London (1928).

6. Bird, R.B., W.E. Stewart, and E.N. Lightfoot, "Transport Phenomena," pp. 642-649, John Wiley and Sons, Inc., New York (1960).

7. Ibid., p. 8.

8. Brown, E.N., "Rotating Bowl for the Production of Uniform Drops," Rev. Sci. Instr., 32, 914 (1961).

9. Brown, E.N., personal communication, National Center for Atmospheric Research, Boulder, Colo., May 19, 1968.

10. Emmert, R.E., and R.L. Pigford, "Gas Absorption and Solvent Extraction," in "Chemical Engineers' Handbook," J.H. Perry, editor, pp. 14-23, McGraw-Hill, New York (1963).

11. Gillespie, L.J., special editor, "Density and Thermal Expansion of Aqueous Solutions of Inorganic Substances and of Strong Electrolytes," in "International Critical Tables of Numerical Data, Physics, Chemistry and Technology," E.W. Washburn, editor, Vol. III, p. 65, McGraw-Hill, London (1928).

12. Haas, P.A., personal communication, Oak Ridge National Laboratory, Oak Ridge, Tenn., May 8, 1968.

13. Roberts, D.J., and J.P. Devemy (Consultant: S.D. Clinton), "Dispersion of Uniform Liquid Microspheres," ORNL-MIT-49, M.I.T. School of Chemical Engineering Practice, Oak Ridge National Laboratory, Oak Ridge, Tenn. (March 27, 1968).

14. Walton, W.H., and W.C. Prewett, "The Production of Sprays and Mists of Uniform Drop Size by Means of Spinning Disc Type Sprayers," Proc. Phys. Soc., B62, 341 (1949).

15. Weast, R.C., editor, "Handbook of Chemistry and Physics," 47th ed., Chemical Rubber Company, Cleveland (1966).

16. Young, T.F., and W.D. Harkins, "Surface Tension Data for Certain Pure Liquids Between 0 and 360°C and for all Types of Solutions at all Temperatures," in "International Critical Tables of Numerical Data, Physics, Chemistry and Technology," E.W. Washburn, editor, Vol. IV, p. 446, McGraw-Hill, London (1928).

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