MASS TRANSFER OF WATER FROM SINGLE AQUEOUS SOL DROPLETS
FLUIDIZED IN A PARTIALLY MISCIBLE ALCOHOL*

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ABSTRACT

MASTER

During the preparation of sol-gel microspheres, droplets of the aqueous sol must be suspended or fluidized in an organic phase until enough water is extracted from the sol to cause gelation. The rate of mass transfer was investigated by fluidizing single water or thorium sol droplets in a partially miscible alcohol and measuring their diameters as a function of time. Three alcohols were studied: 2-ethyl-1-hexanol, 2-methyl-1-pentanol, and *n*-hexanol. The first alcohol was studied at 25, 40, and 50°C, whereas the remaining two were studied at 25°C only. The Schmidt numbers for these systems ranged between 5,000 and 36,000, and the sphere Reynolds number varied from 0.2 to 30.

A surface-active agent, Ethomeen S/15, was added to the organic phase to produce a rigid liquid drop with no induced circulation. By relating the tangential velocity at the drop interface to the sphere drag coefficient, an expression for the Sherwood number was derived and then confirmed experimentally for both water and sol drop data.

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An explicit relationship for the mass transfer coefficient was shown to be independent of the drop diameter, and an expression was obtained to predict the gelation times of thorium sol droplets from known sol and solvent properties.

INTRODUCTION

The sol-gel process which was developed at the Oak Ridge National Laboratory is a method for preparing a wide variety of ceramic fuel materials for use in nuclear reactors. As compared with conventional preparation procedures, the sol-gel process has the distinct advantage of low calcination temperature for obtaining particles of near-theoretical density.¹

The original objective of the sol-gel process was to produce thorium-uranium fragments suitable for vibratory compaction in metal tubes; however, during the past ten years, emphasis has been shifted toward the preparation of spherical particles, or microspheres. Pyrolytic-carbon-coated microspheres of thorium and uranium oxides, 200 to 400 microns in diameter, are the proposed fuel materials for certain high-temperature, gas-cooled reactor designs. These microspheres are prepared by dispersing uniformly sized drops of the appropriate sol into a partially miscible alcohol such as 2-ethyl-1-hexanol (2EH). The sol droplets must be fluidized in the organic phase until enough water is extracted from the aqueous sol to cause gelation. After drying, the gelled microspheres are calcined at 1150°C to produce an oxide product having a density within 1% of theoretical.²

The transfer of mass from systems of spheres to a surrounding fluid has been investigated rather extensively by studying single spheres suspended in a controlled flow of liquid or gas. Mass transport between a suspended fluid sphere and its surroundings can usually be separated into three categories: (1) transfer within the sphere, (2) transfer through the interface, and (3) transfer

outside the sphere. The aim of this study is to characterize mass transfer from a fluidized liquid drop in which the rate limiting resistance is located outside the sphere.

Many experimental data on forced-convection mass transfer from solid spheres have been correlated by a relationship for the Sherwood number involving the product of $N_{Re}^{1/2}$ and $N_{Sc}^{1/3}$.³⁻⁶ Although the exponent for the Reynolds number can be assumed to be constant and equal to 1/2, there is an accumulation of evidence to indicate that the exponent for the sphere Reynolds number varies with N_{Re} , increasing from 1/3 in the Stokes' law regime to 2/3 in the fully turbulent region.⁷ Despite the extensive investigations with single spheres, the available information is inadequate to establish a universal relationship for the mass transfer coefficients.^{8,9}

As compared with solid spheres, forced-convection mass transfer from liquid drops is complicated because conditions at the interface can induce internal circulation of the drop. Although numerous relationships are available for varying degrees of mobility of the drop surface,¹⁰ the actual mass transfer process can be very complex and may involve effects of interfacial turbulence varying from mild circulation near the interface to spontaneous emulsification.¹¹ In the past, some investigators have tried to attribute unexpected mass transfer results to a significant interfacial resistance caused by the accumulation of high-molecular-weight materials at the interface.¹² From later studies with liquid-liquid systems, however, it has become increasingly evident that, instead of acting as interfacial barriers, surface-active agents merely modify the drop hydrodynamics.¹³

In contrast with the situation for gases, the theory of the liquid state is only partly developed; and the existing theories do not provide as good a basis for the prediction of diffusion coefficients as the available empirical equations. Sherwood and Reid¹⁴ have recommended that the diffusivity of water in organic solvents be estimated by calculating a value from the Wilke-Chang relationship¹⁵ and then dividing the results by 2.3. Previously, the comparison of calculated and experimental values for the diffusivity of water in different organic solvents had led Olander¹⁶ to suggest this factor of 2.3. Olander concluded that, as a diffusing species in an organic solvent, water has a molar volume four times larger than expected due to polymerization of the molecules.

In the present study,¹⁷ single water or thoria sol drops were fluidized in three different alcohols: 2EH, 2-methyl-1-pentanol (2MP), and n-hexanol (nH). The droplet diameter and the fluidizing flow rate were measured as a function of time. The controllable variables in each run were initial drop diameter, initial sol molarity, organic-phase temperature, and water concentration in the organic phase. A surface active agent was added to the organic phase to produce a rigid aqueous drop. The objectives of this study were to determine mass transfer coefficients for the transport of water into the organic phase and to develop a correlation to predict drop gelation times from known sol and organic-phase properties.

THEORETICAL ANALYSIS

At 25°C the controlling mass transfer resistance for a thoria sol droplet fluidized in 2EH was shown to be in the organic-phase film surrounding the drop.¹⁸ With this model, the mass transfer coefficients for a water drop and a sol drop will be equal under identical hydrodynamic conditions. The amount of solvent that can be transferred into the aqueous drop is inconsequential; therefore, the change in drop diameter is a direct measure of the amount of water transferred into the organic phase. The mass flux can be related to the measured change in drop diameter with time, as follows:

$$N = - \frac{\rho_w}{\pi D_p^2} \left(\frac{\pi}{6} \right) \frac{d(D_p^3)}{dt} = - \frac{\rho_w dD_p}{2dt} = K(C_s - C_o) \quad (1)$$

During gelation the mass transfer resistance within a thoria sol drop will be negligible if water can be supplied to the interface rapidly enough to maintain C_s at saturation.

When the controlling mass transfer resistance is in a thin continuous-phase film surrounding the drop, the steady-state, convective-diffusion equation for the solute in natural curvilinear coordinates (see Figure 1) is:

$$u \frac{\partial C}{\partial x} + v \frac{\partial C}{\partial y} = D_v \frac{\partial^2 C}{\partial y^2} \quad (2)$$

In Equation (2), tangential diffusion is assumed to be negligible. In the immediate vicinity of the drop, the continuity equation for the continuous phase is:

$$\frac{\partial(ur)}{\partial x} + \frac{\partial(vr)}{\partial y} = 0 \quad (3)$$

Friedlander¹⁹ has suggested that the thin concentration boundary layer approximation is valid for Peclet numbers greater than 10^2 . Since the Schmidt numbers in this study are greater than 5,000, the approximation should apply for Reynolds number values greater than 0.02.

Because of the large Schmidt numbers, the concentration boundary layer thickness should be an order of magnitude thinner than the corresponding momentum boundary layer. Consequently, the entire concentration gradient will occur in the immediate vicinity of the drop surface, and for a rigid interface, the tangential velocity can be represented approximately by:

$$u = u_1' y \quad (4)$$

After substituting Equation (4) into (3) and integrating with respect to y , the following relationship is obtained for the radial velocity component:

$$v = -\frac{1}{r} \frac{\partial}{\partial x} \left(r u_1' \frac{y^2}{2} \right) \quad (5)$$

Using Equations (4) and (5) for the tangential and radial velocity components, Equation (2) can be integrated to obtain the solute concentration profile in the thin continuous-phase film. By the application

of Fick's law, the resulting concentration gradient at the drop interface can be related to the Sherwood number as follows:^{20,21}

$$N_{Sh} = 0.641 N_{Pe}^{1/3} \left[\int_0^\pi \left(\frac{D_p u_i'}{2U} \right)^{1/2} (\sin \theta)^{3/2} d\theta \right]^{2/3} \quad (6)$$

By defining an average value of u_i' which can be treated as a constant, Equation (6) can be integrated to give:

$$N_{Sh} = 0.738 N_{Pe}^{1/3} \left[\left(\frac{D_p}{U} \right)^{1/2} (u_i')_{avg}^{1/2} \right]^{2/3} \quad (7)$$

For the Reynolds number range of interest in this study (0.2 to 30), an expression for $(u_i')_{avg}$ can be approximated from basic fluid-flow data. The drag coefficient for flow around a sphere is defined by the following relationship:

$$F = \frac{f_D}{g_c} \left(\frac{\rho_s U^2}{2} \right) \left(\frac{\pi}{4} D_p^2 \right) \quad (8)$$

Since the drag coefficient for a sphere, f_D , is a function of Reynolds number only, the following expression can be used to describe the drag coefficient for a limited Reynolds number range:

$$f_D = A N_{Re}^a \quad (9)$$

where A is a constant (24 for the Stokes' law region) and the exponent, a , varies from -1 for $N_{Re} < 0.1$ to 0 for $N_{Re} > 1000$. The force exerted on the sphere by the flowing fluid can be separated into two categories: form drag and frictional drag. By summing the tangential shear stress over the drop surface in the direction of flow, the drag force due to friction for a Newtonian fluid is:

$$F_{\text{friction}} = \frac{\pi \mu_s}{2g_c} D_p^2 \int_0^\pi u_i' \sin^2 \theta d\theta \quad (10)$$

By defining a second average value of u_i' , Equation (10) can be integrated to give:

$$F_{\text{friction}} = \frac{\pi^2 \mu_s}{4g_c} D_p^2 (u_i')_{\text{AVG}} \quad (11)$$

For the Stokes' law region, the ratio of friction drag to the total drag force is 2/3. Jenson²² has calculated this ratio to be 0.66, 0.64, 0.63, and 0.60 for sphere Reynolds numbers of 5, 10, 20, and 40, respectively. Assuming $F_{\text{friction}} = (2/3)F$, Equations (8), (9), and (11) can be combined to give:

$$(u_i')_{\text{AVG}} = \frac{A N_{Re}^a \rho_s U^2}{3 \pi \mu_s} \quad (12)$$

For the Stokes' law region where $u_i' = (3 U/D_p) \sin \theta$, it can be shown that $(u_i')_{\text{AVG}} = 1.05 (u_i')_{\text{avg}}$. Assuming that the averaging techniques

can be extrapolated to larger Reynolds numbers, the combination of Equation (7) and (12) gives:

$$N_{Sh} = 0.344 A^{1/3} N_{Re}^{(2+a)/3} N_{Sc}^{1/3} \quad (13)$$

Since the numerical coefficient in Equation (13) has been calculated using two assumptions from the Stokes' law region, it would appear logical to let $A = 24$; then Equation (13) would become:

$$N_{Sh} = 0.99 N_{Re}^{(2+a)/3} N_{Sc}^{1/3} \quad (14)$$

where the value of a must be determined from experimental drag coefficient data.

A simplified expression can be obtained for the mass transfer coefficient by eliminating the fluidizing velocity from Equation (13). With F equal to the weight of the aqueous drop in the continuous phase, the following expression is obtained from Equation (8) for the solvent fluidizing velocity:

$$U^2 = \frac{4g D_p (\rho_p - \rho_s)}{3 f_D \rho_s} \quad (15)$$

Substituting Equation (9) into Equation (15) for the drag coefficient, an explicit relationship can be obtained for the fluidizing velocity. Using this result in Equation (13) gives:

$$K = 3.76 \frac{D_p^{2/3}}{\mu_s^{1/3}} (\rho_p - \rho_s)^{1/3} \quad (16)$$

For a given system and temperature, Equation (16) predicts that the mass transfer coefficient for a fluidized rigid sphere is dependent only on the density difference of the drop and the continuous phase.

By expressing the drop diameter and density as a function of the sol molarity, an expression can be derived from Equations (1) and (16) to predict the gelation time of any sol drop when the controlling mass transfer resistance is in the continuous phase. For a temperature range of 25 to 50°C, the density difference of the thorica sol drop and the solvent for the alcohol systems in this study is:

$$\rho_p - \rho_s = 0.238 M_p + 0.175 \quad (17)$$

The diameter of a sol drop can be related to the molarity of the thorica sol by the following equation:

$$D_p = \left(\frac{M_c}{M_p} \right)^{1/3} D_c \quad (18)$$

where the calculated value of M_c is 37.9. By using Equations (16), (17), and (18), Equation (1) can be integrated to give the following expression for gelation time (in minutes):

$$\theta_G = 2.5 \times 10^{-3} \frac{\mu_s^{1/3} D_c}{D_v^{2/3} (C_s - C_o)} \int_{M_I}^{M_G} \frac{d M_p}{M_p^{4/3} (0.238 M_p + 0.175)^{1/3}} \quad (19)$$

The sol molarity integral function in Equation (19) was evaluated, and values of the integral are tabulated in Table 1. For a desired theoretically dense sphere size, Equation (19) can be used to determine the required fluidization time for a given initial sol molarity and concentration of water in the bulk solvent. Since the value of the integral function is relatively small for the variation in molarity from 12 to 20, the exact gel molarity for a particular system is not necessary in order to obtain a reasonable estimate of the gelation time.

EXPERIMENTAL DATA

The primary purpose of the experimental equipment used in this study was to measure the diameter of a fluidized aqueous drop as a function of time. Basically, the equipment consisted of a tapered Plexiglas column, a light source, a photomacro lens positioned to magnify the drop by a factor of 20 on a ground-glass viewing screen, a Polaroid attachment, a timer, a centrifugal pump to circulate the alcohol, a rotameter, a heat exchanger, and a constant-temperature circulating water system.

The 8-in.-long tapered column was machined from a block of Plexiglas with a convergent-divergent flow channel. A vertical, laminar velocity profile was developed in the convergent section so that each aqueous drop could be fluidized along the central axis of the divergent column

with no horizontal motion. The vertical position of the fluidized drop was controlled by the alcohol flow rate, and all measurements were obtained at an inside column diameter of 0.635 cm.

The general procedure for each experimental run was to form and fluidize a single aqueous sol drop in the desired alcohol and then to measure the alcohol flow rate and drop diameter (to within ± 10 microns) until shrinkage no longer occurred. The independent variables in each experimental run were: initial drop diameter, 0.08 to 0.22 cm; initial sol molarity, 0 to 2.5 M thoria; solvent, 2EH, 2MP, or nH; solvent temperature, 25 to 50°C; and water concentration in the bulk solvent, 0 to 50% of saturation. A surface-active agent, Ethomeen S/15 (a tertiary amine marketed by Armour Industrial Chemical Company), was added to the organic phase in sufficient concentration (0.01 to 1.0 vol %) to produce a rigid liquid drop having no induced circulation.

The diameters of fluidized drops of water and thoria sol were measured, as a function of time, in a series of 131 runs. Typical data obtained for single water drops fluidized in nH at 25°C are shown in Figure 2. The concentration of water in the nH was 27 mg/cc, and the surfactant concentration was 1.0 vol %. The constant slope of the water-drop-diameter data with time indicates a constant mass flux of water from the drop.

Typical diameter data for single thoria sol drops fluidized in 2EH at 50°C are shown in Figure 3. The initial molarity of the thoria sol was 1.1, the concentration of water in the 2EH was 11 mg/cc, and the surfactant concentration was 0.01 vol %. The slope of the diameter data, which is proportional to the mass flux or the mass transfer coefficient, increases with increasing sol molarity. At the gelation

time, the apparent mass transfer rate becomes zero. The thoria gelation concentration varied from 13 to 18 M and was a function of the particular alcohol system and surfactant concentration.

For each diameter measurement, a characteristic fluidizing velocity was calculated by the following equation:

$$U = \frac{8 Q}{\pi(D^2 - D_p^2)} \quad (20)$$

The diffusion coefficients of water in the different alcohols were calculated by using the recommendation of Reid and Sherwood.¹⁴ Values of mass diffusivity, Schmidt number, and water solubility for the alcohol systems are given in Table 2.

RESULTS

The mass transfer data for the water and sol drop runs were correlated by plotting the logarithm of the Sherwood number divided by the Schmidt number raised to the 1/3 power against the logarithm of the Reynolds number (see Figure 4). The sphere Reynolds number varied from 0.2 to 30, and the Schmidt numbers for the different alcohol systems ranged between 5,000 and 36,000. A least-squares line was calculated for each of the 43 water drop runs (1785 data points), and the solid line in Figure 4 was obtained by averaging all of the least-squares constants. The equation for the solid line is:

$$N_{Sh} = 0.980 N_{Re}^{0.417} N_{Sc}^{0.333} \quad (21)$$

and the standard deviations in the coefficient and the Reynolds number exponent are 0.010 and 0.015, respectively. In Figure 4, all of the water

drop data are enclosed by the two parallel dashed lines which indicate a ± 10 percent variation from Equation (21). The data points in Figure 4 represent all of the sol drop runs at thoria molarities from 0.85 to 18. Despite the increased scatter in the thoria data, the mass transfer coefficients for both sol and water drops appear to be equal at a particular value of the sphere Reynolds number.

To test the validity of Equation (16), which is an explicit expression for the fluidized-drop mass transfer coefficient, the experimental rigid-drop data are shown in Figure 5. The ordinate is the product of the mass transfer coefficient and the solvent viscosity raised to the $1/3$ power, divided by the mass diffusivity to the $2/3$ power; the abscissa is the density difference for the ^{μ} _A aqueous drop and the solvent. The 43 water drop runs are located within a density difference of 0.17 to 0.18 g/cc, and the 408 sol drop data points are located within a density difference range of 0.4 to 4 g/cc. The equation of the least-squares line through all of the mass transfer data is:

$$K = 3.48 \frac{D_v^{2/3}}{\mu_s^{1/3}} (\rho_p - \rho_s)^{0.347} \quad (22)$$

In Figure 5, the solid line is Equation (22) and the two parallel dashed lines indicate a ± 10 percent variation. The single dotted line in Figure 5 is the theoretical prediction of Equation (16).

As predicted by Equation (19), the experimentally determined gelation times for the sol drop runs were plotted against the proposed function of sol and solvent properties (see Figure 6). The solid line in Figure 6 was determined by applying the method of least squares to the 88 data points. The slope and the intercept of the least-squares line are

2.72×10^{-3} and -0.06 min, respectively. The average error between the least-squares line and the experimental gelation times for the sol drop runs is ± 4.5 percent.

DISCUSSION OF RESULTS

The hydrodynamic data for the fluidized rigid water drop runs were correlated to determine the coefficient and exponent in Equation (9). A least-squares line was calculated for each run, and the following equation is the result of averaging the least-squares constants for all 43 runs:

$$f_D = 33.7 N_{Re}^{-0.749} \quad (23)$$

The standard deviations in the coefficient and the exponent of Equation (23) are 4.3 and 0.046, respectively. In this study, the ratio of the drop diameter to the column diameter ranged from 0.06 to 0.35. The flow restriction imposed by the column wall effectively decreases the infinite medium fluidizing velocity, resulting in an increased drag coefficient for a given sphere Reynolds number.

By applying the experimental data of Equation (23) to Equation (13), the predicted Reynolds number exponent for mass transfer is $(2 + a)/3 = 0.417 \pm 0.015$, which agrees precisely with the average measured result [see Equation (21)]. In general, the measured Reynolds number exponent for mass transfer is greater than the average value for the N_{Re} range of 2 to 30 and less than the average value for N_{Re} between 0.2 and 3. These results support the theory that the Reynolds number exponent for mass transfer varies with N_{Re} , increasing from $1/3$ where Stokes' law is applicable to $2/3$ for N_{Re} greater than 1,000.

With Equation (13) the value of the coefficient predicted from hydrodynamic data is $0.344 A^{1/3} = 1.11 \pm 0.05$, whereas the average measured value is 0.980 ± 0.048 [see Equation (21)]. From Equation (14) with $A = 24$ (Stokes' law region with unbounded flow), the value of the predicted coefficient is 0.99. Since the coefficient in Equation (13) is based on a constant fraction, $2/3$, of the total drag for unbounded flow, then the fraction of total drag due to friction should be reduced for flow restricted by a column wall. Consequently, the use of $A = 24$ in deriving Equation (14) is more justifiable than using an experimentally determined value with bounded flow (i.e., $A = 33.7$).

The preceding reasoning is also consistent with the theoretical and experimental mass transfer coefficient expressions, Equations (16) and (22). If the fluidizing velocity of a drop is reduced by 20 percent as the result of the effects of a column wall, then Equation (14) predicts that the mass transfer coefficient will be reduced by 8 percent ($1.20^{0.417} = 1.08$) independent of the value of the equation coefficient. Conversely, the functional variables in Equations (16) and (22) are independent of the fluidizing velocity, and the effect of the column wall must appear in the equation coefficients. Equation (16) theoretically predicts the mass transfer coefficient for a fluidized drop in an unbounded flow field, whereas Equation (22) is the experimental mass transfer coefficient correlation for a drop fluidized within a column wall. Consequently, the experimentally determined equation coefficient of 3.48 is 8 percent lower than the theoretical value of 3.76. Comparison of Equations (16) and (22) over the experimental density difference range (0.17 to 4 g/cc) shows that the 4 percent variation in the density different exponent contributes a maximum variation of only 2 percent in the predicted and measured mass transfer coefficient values.

The greater variation in the sol drop data for mass transfer can be attributed, for the most part, to the nonspherical shape achieved by the drops during some stage of the gelation process. Shape distortion was most apparent in runs where a large sol drop diameter and a high concentration of surfactant in the alcohol were used. As compared with 2EH, 2MP and nH required a higher concentration of Ethomeen S/15 to prevent drop circulation. Although a higher Ethomeen S/15 concentration has little apparent effect on the water-alcohol interface, the interfacial tension at the sol-alcohol interface is significantly lowered. With increased surfactant concentration, the resulting decrease in interfacial tension permitted a fluidized sol drop to become somewhat ellipsoidal during gelation. The higher Ethomeen S/15 concentration in the alcohol has no effect on the observed mass transfer rate; however, the molarity of the gelled product is apparently decreased.

In spite of the sphericity problem associated with sol drops, about 90 percent of the sol drop mass transfer data lies within the limits of the water drop data (see Figure 4). The agreement of sol and water drop Sherwood numbers for identical Reynolds numbers confirms that the limiting mass transfer resistances for fluidized-rigid sol and water drops are equal.

The gelation times of sol drops can be predicted from sol and solvent properties with sufficient accuracy to justify any engineering design calculations. The increased slope, 2.7×10^{-3} , in Figure 6 as compared with the predicted value of 2.5×10^{-3} in Equation (19) can be attributed directly to the 8 percent variation between the theoretical and experimental expressions for the mass transfer coefficient, Equations (16) and (22). Theoretically, the time intercept should approach zero as the value of D_c becomes infinitesimally small. Since the experimental

gelation times were determined to the nearest even tenth of a minute, the calculated intercept of -0.06 min for the data in Figure 6 can be neglected.

CONCLUSIONS

The following conclusions can be drawn from this study with regard to the mass transfer of water from aqueous droplets fluidized in a partially miscible alcohol:

1. The addition of a surface-active agent to the organic phase can produce a rigid aqueous drop with no induced circulation.
2. During the process of gelation, the controlling mass transfer resistance for a thorin sol drop is located in the organic phase surrounding the drop.
3. The forced convection mass transfer correlation for a fluidized drop can be predicted from theoretical considerations and hydrodynamic data.
4. The mass transfer coefficient for a rigid fluidized drop is a function of the mass diffusivity, the solvent viscosity, and the drop and solvent density difference.
5. The required gelation time for a fluidized rigid sol drop can be predicted from the properties of the aqueous sol and the solvent.

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LIST OF SYMBOLS

a	exponent in drag coefficient equation, dimensionless,
A	coefficient in drag coefficient equation, dimensionless,
C	local concentration of water in solvent, g/cc,
C ₀	concentration of water in bulk solvent stream, g/cc,
C _s	concentration of water in solvent at drop interface, g/cc,
D	column diameter, cm,
D _C	diameter of theoretically dense thorium sphere, cm,
D _p	diameter of aqueous drop, cm,
D _v	diffusion coefficient, cm ² /sec,
f _D	drag coefficient for flow around a sphere, dimensionless,
F	force exerted by fluid on sphere, dynes,
g	acceleration of gravity, cm/sec ² ,
g _C	dimensional constant, 1 g-cm/dyne-sec ² ,
K	mass transfer film coefficient, cm/sec,
<u>M_C</u>	molarity of theoretically dense thorium, moles/liter,
<u>M_G</u>	thorium molarity of gelled sphere, moles/liter,
<u>M_I</u>	initial thorium molarity of sol drop, moles/liter,
<u>M_p</u>	thorium molarity of sol drop, moles/liter,
N	mass flux of water from drop, g/cm ² -sec,
N _{Pe}	Peclet number, $\frac{D_p U}{D_v}$,
N _{Re}	sphere Reynolds number, $\frac{D_p \rho_s U}{\mu_s}$,
N _{Sc}	Schmidt number, $\frac{\mu_s}{\rho_s D_v}$,

N_{Sh}	Sherwood number, $\frac{KD_D}{D_V}$,
Q	volumetric alcohol flow rate, cc/sec,
r	radial distance from axis of symmetry to drop interface, cm,
r_p	radius of drop, cm,
t	time, sec,
u	tangential velocity component in x-direction, cm/sec,
u'_i	derivative of interfacial velocity with respect to y , 1/sec,
U	fluidizing velocity, cm/sec,
v	radial velocity component in the y -direction, cm/sec,
x	distance along drop from forward stagnation point, cm,
y	distance normal to interface in continuous phase, cm,
θ	polar angle measured from forward stagnation point, radians,
θ_G	gelation time, min,
μ_s	viscosity of solvent, g/cm-sec,
ρ_p	density of aqueous drop, g/cc,
ρ_s	density of bulk solvent, g/cc,
ρ_w	density of water, g/cc.

TABLE 1
VALUES OF THE SOL MOLARITY INTEGRAL FUNCTION
FOR THOPIA MOLARITIES FROM 0.1 TO 20

M (moles/liter)	$\int_M^{20} \frac{dM}{M_p^{4/3} (0.238 M_p + 0.175)^{1/3}}$
0.1	7.42
0.2	5.17
0.3	4.11
0.4	3.47
0.5	3.02
0.6	2.69
0.8	2.23
1.0	1.91
1.2	1.68
1.4	1.50
1.6	1.35
1.8	1.23
2.0	1.13
2.2	1.05
2.4	0.974
2.6	0.910
2.8	0.853
3.0	0.802
3.5	0.696
4.0	0.612
4.5	0.543
5.0	0.486
6.0	0.395
7.0	0.326
8.0	0.271
9.0	0.227
10.0	0.190
12.0	0.131
14.0	0.087
16.0	0.052
18.0	0.024
20.0	0.000

TABLE 2
PHYSICAL AND CHEMICAL DATA FOR THE ALCOHOLS

Alcohol	Temperature (°C)	D_v (cm ² /sec)	N_{Sc} ($\mu_s/\rho_s D_v$)	C_s (g/cc)
2EH	25	0.25×10^{-5}	35,700	0.020
2EH	40	0.47×10^{-5}	10,900	0.023
2EH	50	0.68×10^{-5}	5,450	0.025
2MP	25	0.33×10^{-5}	18,500	0.043
nH	25	0.39×10^{-5}	13,400	0.054

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Figure 1. Natural Curvilinear Coordinates for a Thin Region at the Surface of the Drop.

Figure 2. Diameter-Time Relationship for Water Drop Fluidized at 25°C in n-Hexanol Containing 27 mg of Water per cc.

Figure 3. Diameter-Time Relationship for 1.10 M Thoria Sol Drop Fluidized at 50°C in 2-Ethyl-1-hexanol Containing 11 mg of Water per cc.

Figure 4. Mass Transfer Correlation for Fluidized Rigid Thoria Sol Drops.

Figure 5. Correlation of Mass Transfer Coefficient for Fluidized Rigid Drops.

Figure 6. Correlation of Gelation Time for Fluidized Rigid Thoria Sol Drops.