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SUBJECT: Gelation of Small Diameter, Thoria Sol Microspheres in Non-Flowing Alcohols

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ABSTRACT

A study was made to determine the parameters affecting the mass transfer of water during gelation of 2 M thoria sols in non-flowing alcohol systems. The alcohols used were 2-ethyl-1-hexanol (2EH) and mixtures of isoamyl alcohol (iAA) and 2-ethyl-1-hexanol. The maximum gelled particle diameters were determined for 5- and 11-ft-long columns of alcohol. Mass transfer theory was used in the development of a computer program to predict the maximum diameters expected.

With an increase in iAA concentration, the maximum gelled particle diameter increased. Cracking and deformation were noted, primarily in the larger spheres, at iAA concentrations greater than 30 vol %. The predictions from correlations indicated the expected trends but were, in general, 10 to 20% low.

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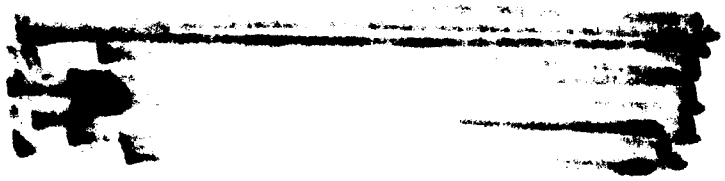

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1. SUMMARY

The Sol-Gel Process for producing nuclear fuels involves the gelation of aqueous sols, containing a fissile or breeding material, by extraction of water. The parameters affecting the mass transfer of water from thoria (a breeding material) sols were studied in a vertical column of non-flowing alcohol. An attempt was made to predict the maximum diameter of gelled spheres obtainable in several alcohol mixtures using the correlation proposed by Clinton (1).

Gelation of the 2 M thoria sols was studied in 5- and 11-ft columns of alcohol. The alcohols investigated were pure 2-ethyl-1-hexanol (2EH) and mixtures of 2-ethyl-1-hexanol and isoamyl alcohol (iAA) (up to 60% vol iAA). The gelled particles were observed with a microscope, and the maximum diameters were determined. Based on mass transfer correlations, a computer study was made to predict the maximum diameter of gelled particles to be expected. The physical parameters used were determined from the experimental measurements.

The results indicate that the diameter of the largest gelled particle increases with increased concentration of iAA. Cracking and deformation were noticed primarily in the larger spheres, at iAA concentrations greater than 30 vol %. (Gelled spheres of 270-290 μ can be produced using 30 vol % iAA without deformation.) When the maximum gelled diameter was plotted against the mole fraction of iAA, the slope of the curve increased with increasing mole fraction of iAA.

The maximum diameters were predicted with an expression of the form

$$N_{Sh} = A + B(N_{Sc}^{1/3} N_{Re}^{1/2})$$

For A = 5 and B = 0.814 proposed by Clinton, the results were, in general, too low. When the values 2 and 1 were assigned to A and B, respectively (the classical values) (6, 7), a better fit to the experimental results was obtained, but again the prediction was low. Although the correlation for the mass transfer coefficient did not quantitatively fit the data, it can be used as a first approximation in determining the alcohol system to be used.

Further study is necessary to determine the effect of surfactant on the gelation of thoria sol particles in the mixtures of alcohols studied. An attempt should be made to determine the values of the A and B constants for systems with surfactants so that predictions can be made from theory. Also a more accurate determination of the water concentration in the microspheres at gelation is critical to obtaining an accurate prediction of the largest microsphere that will gel in a given column.

2. INTRODUCTION

The Sol-Gel Process is used for preparing fuel breeding materials in the form of microspheres for use in nuclear reactors. Aqueous sols containing thorium oxide, uranium oxide, or their mixtures are gelled by extracting water with a suitable dehydrating agent, usually an alcohol. The microspheres are prepared by dispersing the sol into the alcohol; for large spheres a fluidized system is used; for small spheres a stagnant column of alcohol may be sufficient. The sol droplets remain in the dehydrating agent until enough water has been extracted to cause gelation. Once gelation is completed, the microspheres are dried and further processed. Since the dehydrating agent becomes "wet," it must be continuously removed, "dried," and recycled. It is important to minimize the necessary volume of dehydrating agent. In addition, it is important that the dehydrating system produce undistorted, spherically shaped gelled particles.

The object of this study was to define the parameters affecting the mass transport of water during gelation of small spheres. Since countercurrent alcohol flow cannot be used in the gelation of small spheres, a second object of this study was to determine whether Clinton's equation (1) (derived for alcohol upflow, larger spheres, and presence of surfactants) is applicable.

A theoretical analysis was carried out in an effort to determine the important parameters affecting mass transfer. Further, an experimental study was made using several different mixtures of alcohol dehydrating agents in an effort to determine the largest gelled particles obtainable in non-flow systems. A comparison was then made with the theoretical model to assess its value in predicting the mass transfer.

3. THEORETICAL APPROACH

The sol must remain in the dehydrating agent, usually an organic alcohol, until enough water has been extracted to cause gelation. S.D. Clinton (2) found that the controlling resistance to mass transfer from the sol droplets is in the alcohol. The interfacial mass transfer coefficient, K, is defined by the following relation:

$$N = K(C_i - C_o) \tag{1}$$

Clinton (1) was able to correlate his mass transfer data for 1000-2000 μ sol particles in a fluidized system (in the presence of surfactants) with the classical form of the mass transfer relation for rigid spheres (6, 7).

$$N_{Sh} = A + B(N_{Sc}^{1/3} N_{Re}^{1/2}) \tag{2}$$

where:

N_{Sh} = Sherwood number

N_{Sc} = Schmidt number

N_{Re} = Reynolds number

A and B are constants.

Clinton's work:

$A = 5,\ B = 0.814$

Classical relation:

$A \approx 2,\ B \approx 0.5 - 1$

From Eq. (2) the mass transfer coefficient is found to be a function of the following parameters:

$$K = \text{funct}(\rho,\ \mu,\ D_v,\ D_p,\ V_t) \tag{3}$$

By appropriately choosing the dehydrating agent and temperature, one can obtain the desired mass transfer coefficient.

If the solubility of water in the dehydrating agent is much greater than the solubility of the dehydrating agent in water, as must be the case for proper gelation, it is possible to relate the rate of volume decrease of the sol particle to the rate of mass flux across the aqueous phase-alcohol interface (see Appendix 10.1).

$$-\frac{dD_p}{d\theta} \propto N \tag{4}$$

Substituting for N and K from Eqs. (1) and (3), one finds that

$$\frac{dD_p}{d\theta} = \text{funct}'(\rho,\ \mu,\ D_v,\ D_p,\ V_t, \Delta C) \tag{4a}$$

The density, viscosity, and solubility of water in the dehydrating agent were determined experimentally (see Appendices 10.3 and 10.4).

The diffusivity was estimated using the Wilke-Chang correlation (11).

$$D_v = \beta\left[\frac{(\emptyset M)^{0.5}\ T}{V_b^{0.6}\ \mu}\right] \tag{5}$$

or

$$D_v = \alpha\ \frac{T}{\mu} \tag{5a}$$

where α, the proportionality factor, is now a function of the dehydrating agent. For alcohol mixtures an average molecular weight is used to predict D_v (see Appendix 10.5 for a sample calculation (12).

The settling velocity for rigid spheres can be determined from

$$V_t = \sqrt{\frac{4g\ D_p(\rho_p - \rho)}{3\rho\ C_d}} \tag{6}$$

where C_d is the drag coefficient.

$$C_d = \frac{24}{N_{Re}} \qquad \text{Stoke's Law region } (N_{Re} < 0.3) \tag{7}$$

The use of V_t can be extended to $N_{Re} = 7$ using (10)

$$C_d \approx \frac{26.5}{N_{Re}^{0.914}} \qquad 0.3 < N_{Re} \leq 2 \tag{8}$$

$$C_d \approx \frac{25.58}{N_{Re}^{0.809}} \qquad 2 < N_{Re} \leq 7 \tag{9}$$

From a mass balance around the sol particles, the diameter of the sol particle can be related to the density of a particle by

$$D_p = D_{pf}\left(\frac{\rho_{pf} - 1}{\rho_p - 1}\right)^{1/3} \tag{10}$$

Substitution of the foregoing equations allows the calculation of the length required to gel a particular sol diameter.

$$L_{gel} \propto \int_{\rho_{pi}}^{\rho_{pf}} \frac{V_t \; d\rho_p}{K(C_i - C_o)} \tag{11}$$

(A more detailed presentation of the theoretical analysis is given in Appendix 10.1.)

The present study did not use surfactants and therefore there is some question as to whether the assumption of rigid spheres used in Eq. (6) is valid. According to Harmathy (9) liquid particles flowing in immiscible liquid systems can be considered increasingly rigid as the Eotvos number, N_{oe}, approaches zero. The largest Eotvos number in the present study was

$$N_{oe} = \frac{g \; \Delta\rho \; D_p^2}{\sigma} = \frac{(980 \text{ cm/sec}^2)(0.66 \text{ g/cm}^3)(9 \times 10^{-2} \text{ cm})^2}{15 \text{ dynes/cm}}$$

$$= 0.35$$

which is close to zero. Further, liquid particles with $N_{oe} < 15$ are considered spherical in shape.

According to Perry (10) the settling velocities of non-rigid spheres can be approximated by Eq. (6) for $N_{Re} \lesssim 10$. Further, as the sol begins to gel, the particle tends to become more and more rigid. Therefore, to a first approximation, the sol can be considered rigid.

A computer program was written to aid in the evaluation of the integral of Eq. (11) (see Appendix 10.2). The trapesoidal rule was used with a step size equal to one hundredth of the difference between the final and initial particle density. Successive iteration allows the determination of the maximum diameter particle obtainable in a given length column. The physical properties used in the simulation correspond to the properties of the alcohols experimentally determined.

4. EXPERIMENTAL APPROACH AND APPARATUS

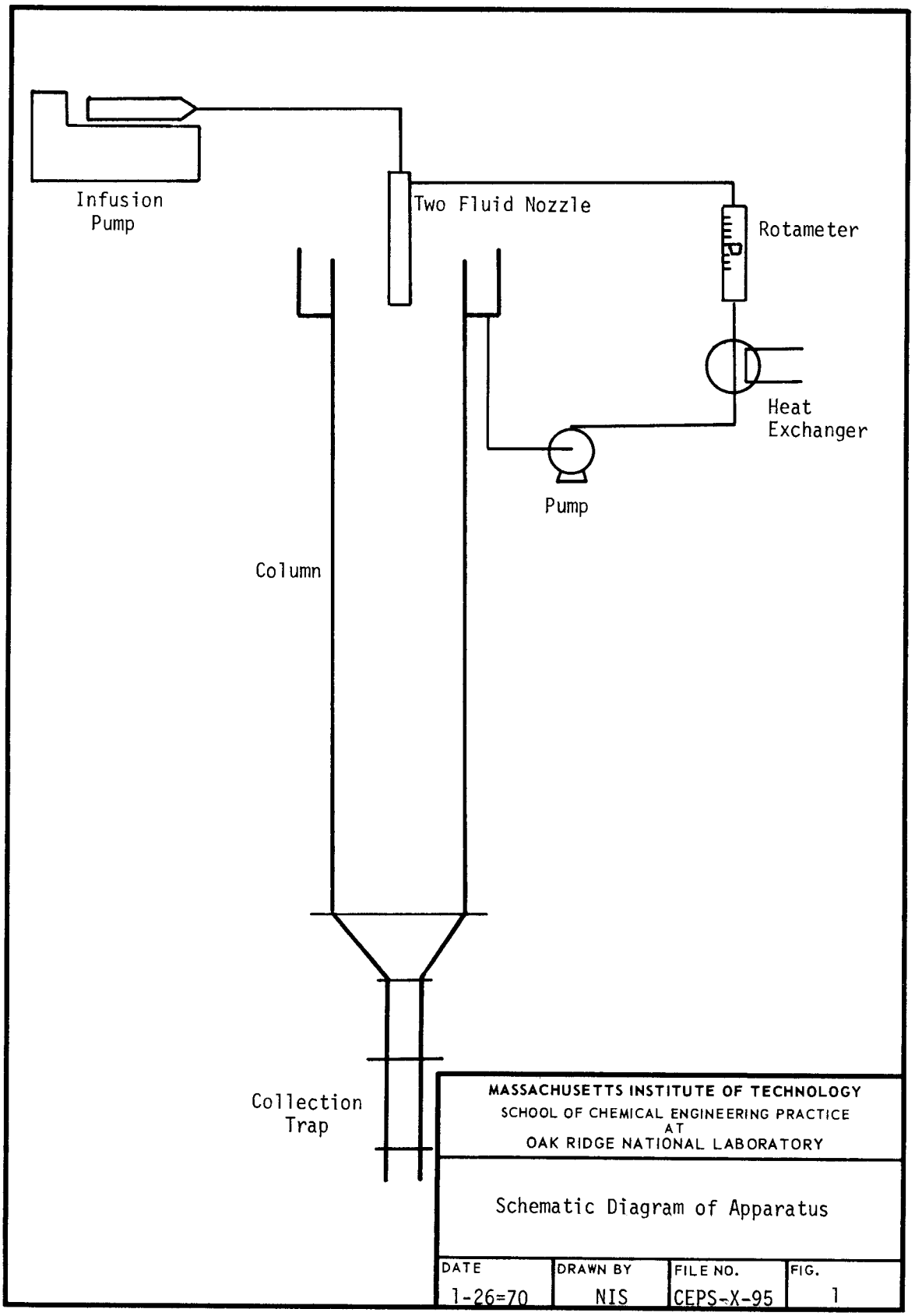
The experimental approach was to determine the diameter of the largest gelled sphere obtained for various column heights and alcohol mixtures. A 2 M ThO_2 sol was used in this study. The column heights investigated were 5 and 11 ft. The alcohols used were 2EH and mixtures of iAA and 2EH. The effect of increased temperature was also studied for 2EH in the 11-ft column.

The experimental apparatus is shown in Fig. 1. It consisted of a 4- or 10-ft-long, 2-in.-ID, vertical glass column. The bottom of the column was attached to a 1-ft-long, tapered fitting which led to a length of 3/4-in.-ID Tygon tubing. This tubing was clamped at two places and formed the collection trap. The top of the column was fitted with an expansion joint and weir to collect the alcohol overflow. The sol was dispersed into the nozzle with an infusion pump. The alcohol drive fluid was recirculated with an impeller pump, metered with a rotameter,and kept at room temperature with a heat exchanger.

The alcohol mixtures used, in volume percent, were:

Mixture No.	Percent iAA	Percent 2EH
1	0	100
2	10	90
3	20	80
4	30	70
5	40	60
6	50	50
7	60	40

The range of alcohol mixtures studied corresponds to the range in physical properties shown in Table 1. The density, viscosity, and solubility of water in the alcohol mixtures were experimentally determined,and the apparatus and results are given in Appendices 10.3 and 10.4. Experiments were conducted for 68% iAA and 32% 2EH, and the solubility of water in this mixture was extrapolated (see Fig. 7) for use in the computer program. Diffusivities were calculated (see Appendix 10.5).


Infusion
Pump
Two Fluid Nozzle
Rotameter
Heat
Exchanger
Pump
Column
Collection
Trap
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Schematic Diagram of Apparatus
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FIG.
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Table 1. Range of Property Values Used in Experimentation

Physical Property	Range
Alcohol density	0.815 to 0.828 gm/ml
Viscosity	4.8 to 7.4 cp
Concentration driving force	22.8 to 41.5 mg H_2O/ml corresponding to pure 2EH and 60% iAA - 40% 2EH, respectively
Diffusivity	2.56×10^{-6} to 3.48×10^{-6} cm^2/sec

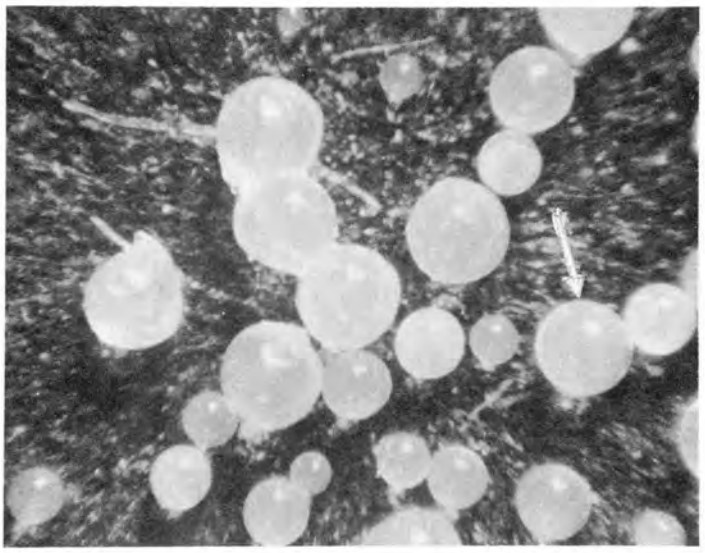
Since only small amounts of the sol were used during an experimental run, it was assumed that the concentration driving force was constant throughout the run (maximum percentage decrease in driving force was 3%). The gelled spheres were collected from the trap and dried by drawing air over them in a sintered glass funnel. Drying took about 15 min. The dried spheres were then analyzed using a binocular microscope with one eye piece fitted with a measuring grid. The largest diameter particles were measured only if they were spherical in shape, and not cracked or pitted on the surface. However, at concentrations above 30% iAA by volume, a particle was considered acceptable if spherical or only slightly eliptical in shape. Typical photomicrographs for the 11-ft-column are shown in Fig. 2.

One set of experimental runs was carried out at a temperature of 48°C. The column was insulated with 1-in.-thick fiberglas insulation. The alcohol, pure 2EH, was preheated to 60°C and poured into the column. About 30 min was allowed for equilibration. Collection was carried out in the same manner as described above.

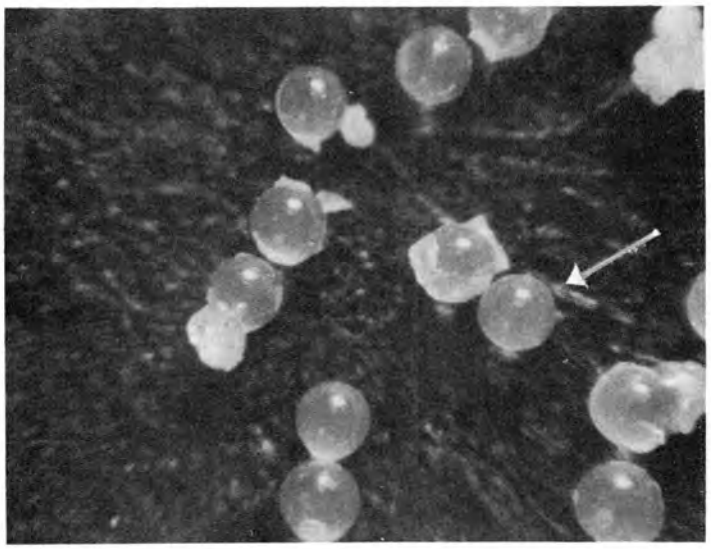
5. RESULTS

The results obtained from the experimental investigation are shown in Figs. 3 and 4. The range of diameters associated with each point corresponds to the accuracy to which the microscope scale could be read.

Examination of the largest obtainable spheres revealed little or no cracking and deformation for alcohol mixtures up to 40 vol % iAA in the 5-ft-column. Cracking and deformation was found to increase as the concentration of iAA was increased above 40 vol %. Similar affects were noted in the 11-ft-column. The largest obtainable spheres in the 11-ft-column were 270-290 μ at 30 vol % iAA. At an alcohol concentration of 60 vol % iAA in the 11-ft-column, the largest particles were slightly deformed. Particles

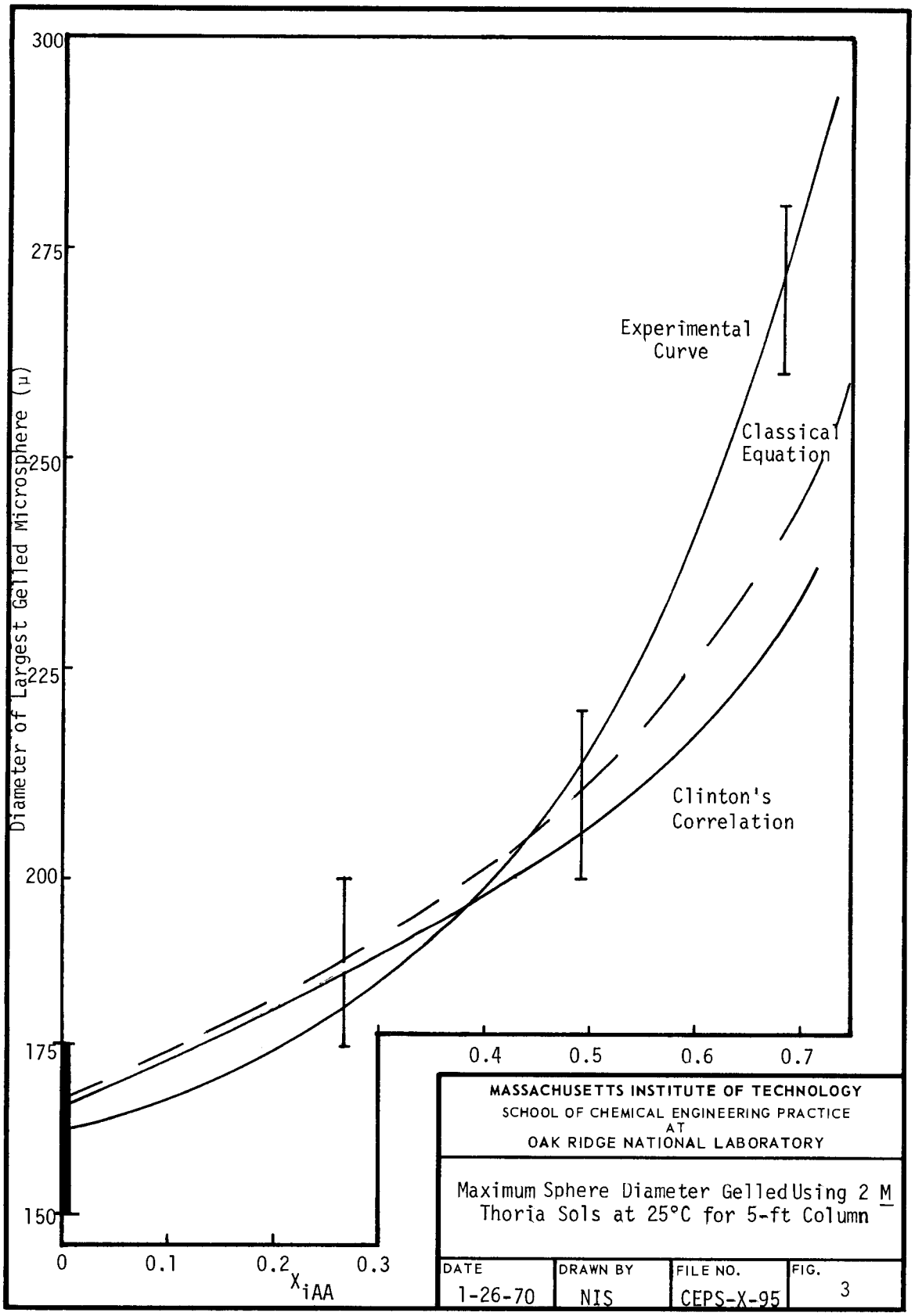


$X_{iAA} = 0.6$



$X_{iAA} = 0.4$

Fig. 2. Typical Photomicrographs for 11-ft-Column (White arrow indicates largest acceptable microsphere.)



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Maximum Sphere Diameter Gelled Using 2 M Thoria Sols at 25°C for 5-ft Column

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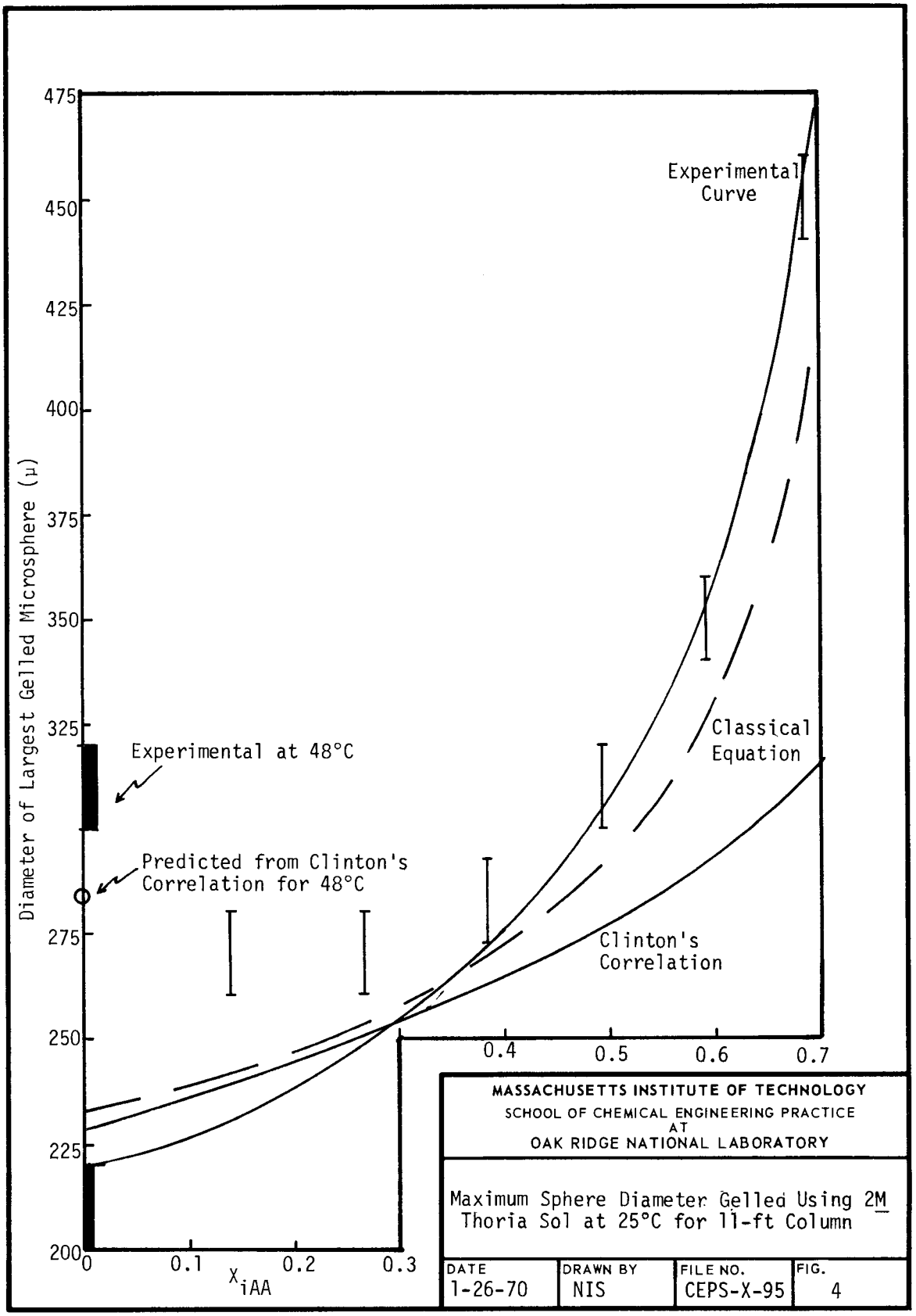
475
450
425
400
375
350
325
275
250
225
200
Diameter of Largest Gelled Microsphere (μ)
Experimental Curve
Classical Equation
Clinton's Correlation
Experimental at 48°C
Predicted from Clinton's Correlation for 48°C
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
X_{iAA}
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Maximum Sphere Diameter Gelled Using 2M Thoria Sol at 25°C for 11-ft Column
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FIG. 4

of 240-260 μ, however, were found to be completely spherical. An experimental run was carried out at 48°C in the 11-ft-column using pure 2EH, and the result is shown in Fig. 4.

Shown also in Figs. 3 and 4 are the maximum gelled diameters predicted by using Clinton's mass transfer correlation and the classical correlation. The classical equation gave a better fit to the data than Clinton's correlation although at high iAA concentrations, both predictions were low. Within the range of data taken, use of the classical equation resulted in predictions less than 20% low.

6. DISCUSSION OF RESULTS

The experimental results show that an increase in iAA concentration causes an increase in the size of the largest gelled microsphere. This result is to be expected since the addition of iAA increases the mass transfer coefficient (primarily through an increase in diffusivity and velocity, and a decrease in viscosity) and the concentration driving force. Although this study is concerned with the largest gelled microsphere producible, from a practical point of view, this sphere may have little value due to slight deformation. It is apparent that the iAA alcohol concentrations greater than 30 vol % give a mass transfer rate that is too great or spheres too large to maintain a spherical shape.

If one compares the predicted diameter with those found experimentally, it is seen that the experimental values are greater, in general, than predicted. This indicates that the predicted mass transfer rate may be too small. Clinton's correlation for the mass transfer coefficient was based on data for single thoria sol spheres, 1000-2000 μ in diameter, in the presence of surfactant in fluidized systems. There is some question as to the validity of applying the expression to smaller spheres in stagnant systems with no surfactant present. Clinton (3) showed that sol particles without surfactant had a greater initial mass transfer rate than sol particles with surfactants. After some gelation had occurred, the mass transfer rate for the sol without surfactant approached that of the sol with surfactant. He found, however, that the total gelation time was considerably reduced. Thus the different surfactant concentration is a possible reason for the discrepancy found between the experimental and the predicted results based on Clinton's correlation.

The classical mass transfer equation (A = 2 and B = 1) was also used to predict the largest microspheres produced in the 5- and 11-ft-columns for the different alcohol mixtures. It was found that the experimentally observed particle diameters agreed better with the classical mass transfer equation than with Clinton's equation. This tends to substantiate the claim that the discrepancy between the observed maximum gelled sphere diameter and the predicted maximum sphere diameter using Clinton's equation is due to the absence of surfactant in the system.

The discrepancy between the experimental results and those predicted from the classical mass transfer correlation might be explained by possible errors in the values of the diffusivity and settling velocity terms. The diffusivity was determined using the Wilke-Chang correlation. Application of the correlation to a multicomponent system has been shown to have errors as high as 20% associated with it (11). This uncertainty results in approximately an 8% error in maximum diameter. The settling velocity equations could be somewhat in error if the sol particles do not act as a rigid sphere. However, this error is small if the particle Reynolds number is less than 10 (10). The increase in velocity due to the particle not behaving as a rigid sphere would tend to increase the rate of mass transfer and decrease the gelation time. The residence time would, however, also decrease thus tending to nullify the effect.

In all computer simulations the initial sol particle density was taken as 1.476 gm/ml (2 M); the final gelled particle density was taken as 4.808 gm/ml (16 M), the average density found by Clinton (5) for gelled thoria spheres. During the present study the gelled particle density was not determined. It is believed, therefore, that some error could be attributed to differences in final density; however, no effort was made to quantify it.

Even though the classical equation does not predict the maximum particle diameters exactly, it can still be of great value as a first approximation in predicting the effects of different alcohol systems. As the results indicate, the effect of increased concentration of iAA can be estimated.

The effect of temperature as seen by the prediction using Clinton's correlation can be estimated by the classical equations, but unfortunately the predicted value was not obtained due to limitations of the computer routine.

7. CONCLUSIONS

1. It is possible to obtain gelled spheres of 270-290 μ in diameter without distortions using an 11-ft-column filled with 30 vol % iAA - 70 vol % 2EH. The largest gelled spheres obtained in the 5-ft-column were 200-220 μ using a 40 vol % iAA - 60 vol % 2EH alcohol mixture. It is possible to make larger spheres in either column by increasing the concentration of iAA, but an increase in cracking and deformation results.

2. The computer predictions in most cases gave results 10-20% less than was found experimentally.

3. Although the predictions from the equations developed are not exact, they show the proper indication of the effects of the several parameters on the mass transfer. The simulation can, therefore, be used as a guide for choosing the proper alcohol dehydrating agent.

8. RECOMMENDATIONS

1. Further experimentation should be carried out using single thoria sol microspheres with and without surfactants in stagnant alcohol systems in an effort to obtain a more accurate correlation for mass transfer.

2. Experimentation should be made using different alcohol and sol systems to determine the generality of the mass transfer correlation.

3. A study should be made to determine a correlation to predict cracking and deformation.

4. An experimental method should be developed which allows a determination of the molarity of the final gelled particle.

9. ACKNOWLEDGEMENTS

The authors wish to thank P.A. Haas and S.D. Clinton for their help throughout this project. We would also like to thank the Analytical Chemistry Division for their assistance.

10. APPENDIX

10.1 Theory

The expression for the mass flux is

$$N = K(C_i - C_o) \tag{1}$$

where K is correlated by

$$N_{Sh} = A + B(N_{Sc}^{1/3} N_{Re}^{1/2}) \tag{2}$$

Solving for K, one obtains

$$K = \frac{AD_v}{D_p} + B\left(\frac{\mu^{1/3}}{\rho^{1/3} D_v^{1/3}} \frac{\rho^{1/2} V_t^{1/2} D_p^{1/2}}{\mu^{1/2}} \frac{D_v}{D_p}\right)$$

$$= \frac{AD_v}{D_p} + B\left(\frac{\rho^{1/6} V_t^{1/2} D_v^{2/3}}{\mu^{1/6} D_p^{1/2}}\right)$$

N can be related to the rate of volume decrease as follows:

$$- d(\rho_w \frac{\pi}{6} D_p^3) = N \pi D_p^2 d\theta \tag{12}$$

$$- \frac{\rho_w}{2} \frac{dD_p}{d\theta} = K(C_i - C_o)$$

From a mass balance on the sphere, the density of the particle can be related to the molarity of the particle (4).

$$\rho_p = 0.238 \underline{M}_p + 1 \tag{13}$$

The density of water is assumed equal to 1 gm/ml.

The molarity of the dehydrating sol drop can be eliminated using the equation

$$\left(\frac{D_{pf}}{D_p}\right)^3 = \frac{M_p}{M_{pf}} \tag{14}$$

Therefore,

$$\left(\frac{D_{pf}}{D_p}\right)^3 = \frac{\rho_p - 1}{\rho_{pf} - 1} \tag{10}$$

The velocity can be expressed using

$$V_t = \sqrt{\frac{4g\ D_p(\rho_p - \rho)}{3\rho\ C_d}} \tag{6}$$

where:

$$C_d = \frac{24}{N_{Re}} \qquad N_{Re} < 0.3 \tag{7}$$

$$C_d = \frac{26.5}{N_{Re}^{0.914}} \qquad 0.3 < N_{Re} < 2 \tag{8}$$

$$C_d = \frac{25.58}{N_{Re}^{0.809}} \qquad 2 < N_{Re} < 7 \tag{9}$$

Finally the diffusivity can be calculated using the Wilke-Chang correlation (11).

$$D_v = \frac{7.4 \times 10^{-10}}{2.3}\left(\frac{(\phi M)^{0.5}\ T}{V_b^{0.6}\ \mu}\right) \tag{5}$$

or

$$D_v = \alpha\ \frac{T}{\mu}$$

where

$$\alpha = \frac{7.4 \times 10^{-10}}{2.3}\left(\frac{(\emptyset M)^{0.5}}{V_b^{0.6}}\right)$$

The gel time can be calculated from Eqs. (10) and (12) and simplifying,

$$d\theta_{gel} = \frac{\rho_w D_{pf}(\rho_{pf} - 1)\, d\rho_p}{6K(C_i - C_o)(\rho_p - 1)^{4/3}} \tag{15}$$

where:

$$K = \frac{A\alpha T}{D_{pf}\mu}\left(\frac{\rho_p - 1}{\rho_{pf} - 1}\right)^{1/3} + B\left[\frac{\rho^{1/6}\, V_t^{1/2}(\alpha T)^{2/3}}{\mu^{3/6}\, D_{pf}^{1/2}}\left(\frac{\rho_p - 1}{\rho_{pf} - 1}\right)^{1/6}\right]$$

and V_t is given by Eq. (6).

The length of column required to gel a sphere of final diameter D_{pf} is

$$L_{gel} = \int_{\rho_{pi}}^{\rho_{pf}} V_t\left(\frac{d\theta_{gel}}{d\rho_p}\right) d\rho_p \tag{11}$$

where $d\theta_{gel}/d\rho_p$ is obtained from Eq. (15) and the limits of integration are the sol particle density initially and the particle density at gelation. In the present work

$$\rho_{pi} = 1.476 \text{ gm/ml } (2\ \underline{M})$$

$$\rho_{pf} = 4.808 \text{ gm/ml } (16\ \underline{M})\ (\underline{5})$$

10.2 Computer Simulation

A computer code was developed to calculate the final densities and diameters of the microspheres and properties of the dehydrating alcohol as well as the time and distance of fall necessary for proper gelling. The Stokesian relation for particle Reynolds numbers less than 0.2 was used to determine the free fall velocity, and for Re $\leq$ 2.0 a modified form was applied. A listing of the code and the user supplied input can be found in the calculation file.

10.3 Viscosity and Density Determination

The viscosity of the dehydrating agent was determined at 25°C and various alcohol mixtures with the use of an Ostwald Viscometer. The same volume of alcohol was drawn into a reservoir and allowed to flow, by gravity, through a capillary tube for each run. The time was determined for the liquid to flow by two marks. From the relation: $\mu = C\rho t$(5), the viscosity can be determined. (t = time (sec), ρ = density (gm/cc), and C = a constant.)

C was determined by the use of pure 2EH, whose viscosity was obtained from P.A. Haas (8);the density and time were experimentally determined. The temperature of the viscometer was held between 24-25°C with the use of a Dewar flask filled with 25°C water. The results are shown in Fig. 5.

Density measurements were made using a hydrometer on excess samples of alcohols used in the viscosity measurements. The results can be seen in Fig. 6.

10.4 Solubility of H_2O in Alcohol Mixtures

The solubility of water in the alcohol mixtures used was obtained from samples submitted to the Analytical Chemistry Division. The determinations were carried out using the Carl Fischer titration method at 25°C. The results can be seen in Fig. 7.

10.5 Sample Calculation of Diffusivities

From Reid and Sherwood (11),

$$D^{\circ}_{12} = 3.22 \times 10^{-8} \left[(\Phi M_2)^{1/2} T/\mu_2 V_b^{0.6}\right]$$

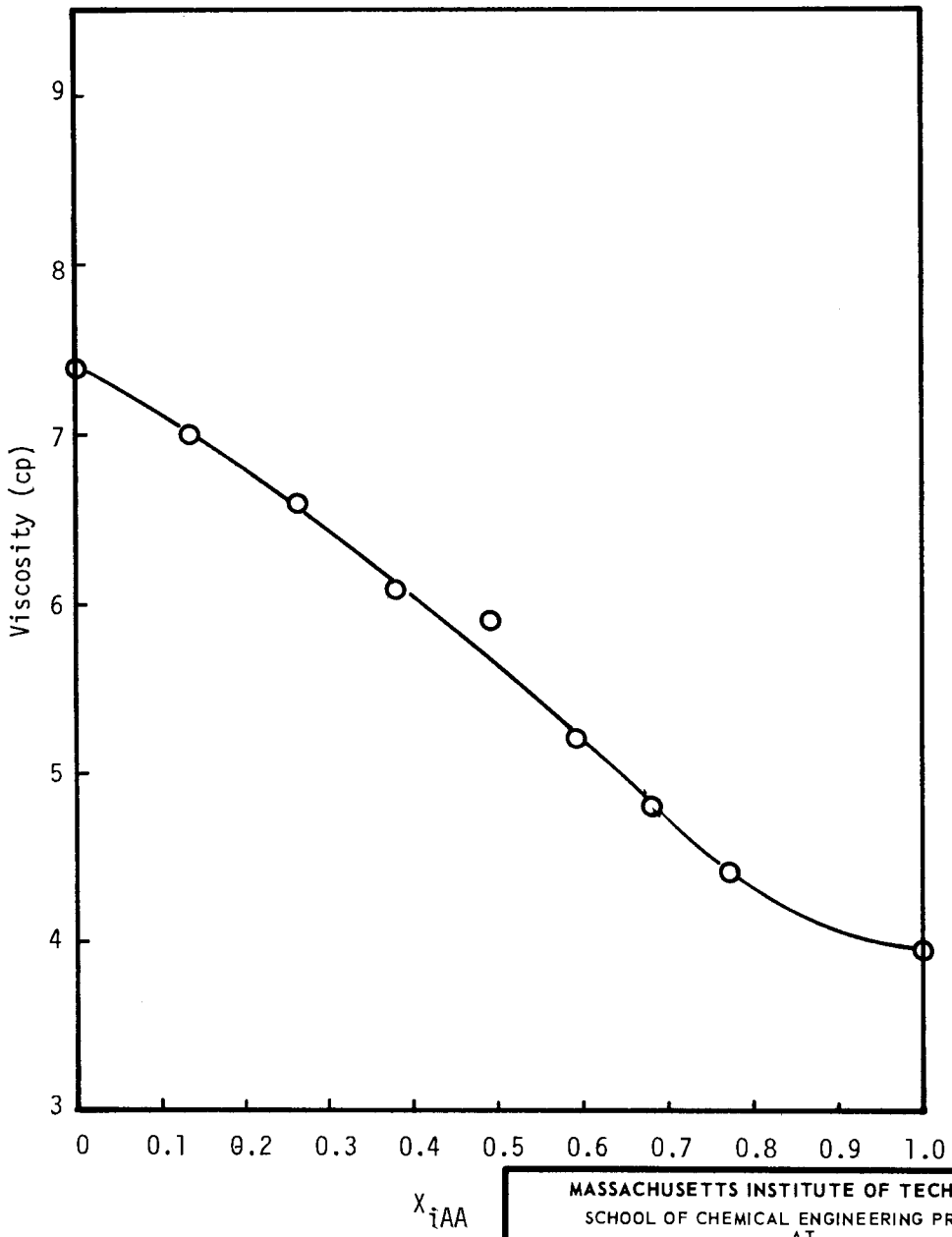
where:

D°_{12} = mutual diffusion coefficient of solute 1 in solvent 2 at very low solute concentrations, cm^2/sec

M_2 = molecular weight of solvent, g/gmole

T = temperature, °K

μ_2 = viscosity of solution (solvent), cp



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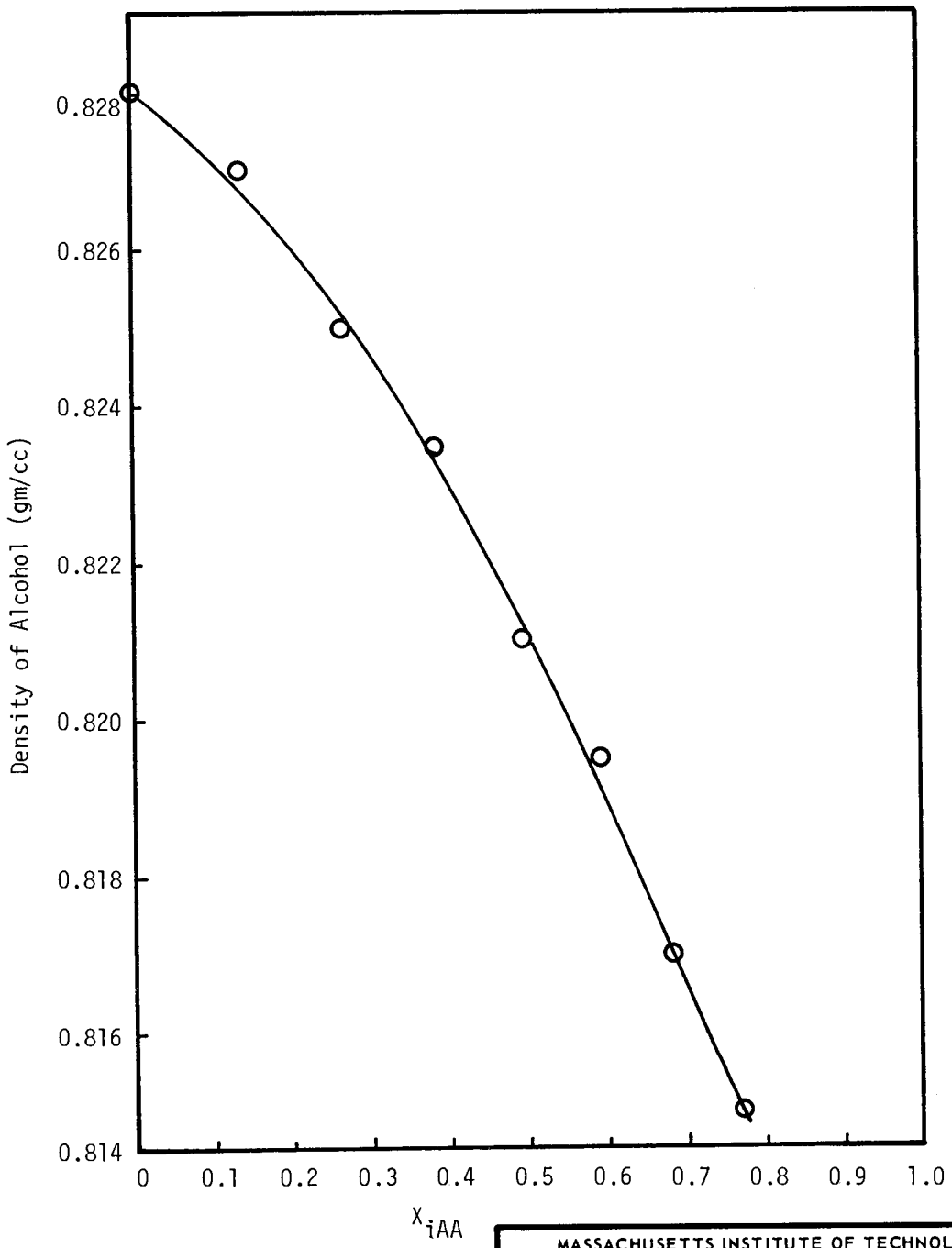
Viscosity of 2EH-iAA Mixtures
at 25°C

DATE	DRAWN BY	FILE NO.	FIG.
1-26-70	NIS	CEPS-X-95	5



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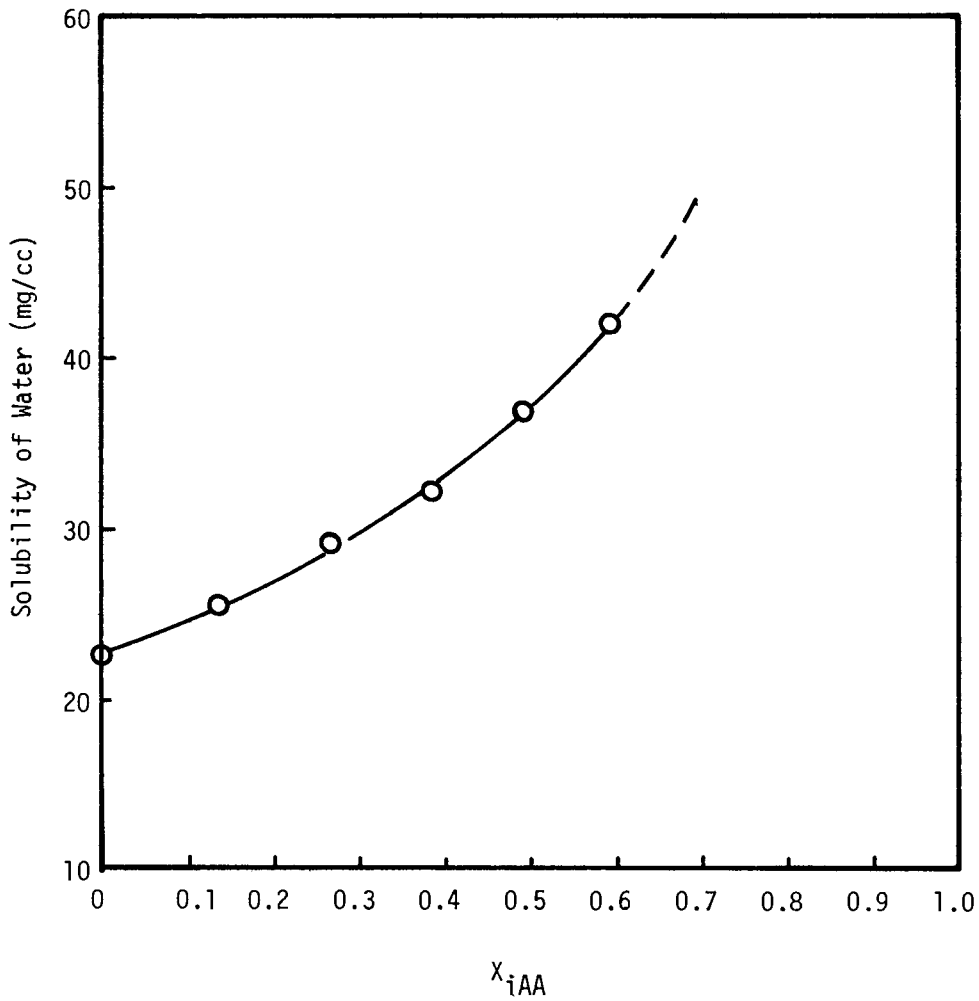
Density of Mixtures of
2EH-iAA at 25°C

DATE	DRAWN BY	FILE NO.	FIG.
1-26-70	NIS	CEPS-X-95	6



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Solubility of Water in Mixtures of 2EH and iAA at 25°C

DATE	DRAWN BY	FILE NO.	FIG.
1-26-70	NIS	CEPS-X-95	7

V_b = molal volume of the solute at its normal boiling point, $cm^3/gmole$

Φ = association parameter of solvent

In this case water is the diffusing solute. For water

$$V_b = 18.9\ cm^3/gmole$$

For the alcohol systems studied, $\Phi = 1$.

10.5.1 Calculation of Diffusion Coefficient for the Diffusion of Water in 2EH

Molecular weight of alcohol = 130.23 g/gmole

$T = 298°K$

Viscosity of 2EH = 7.4 cp (see Appendix 10.3)

$$D^{\circ}_{12} = 3.22 \times 10^{-8}[(1 \times 130.23)^{1/2}(298)/(7.4)(18.9)^{0.6}]$$
$$= 2.56 \times 10^{-6}\ cm^2/sec$$

10.5.2 Calculation of Diffusion Coefficient for the Diffusion of Water in a 60% iAA - 40% 2EH by Volume Mixture

$X'_{iAA} = 0.6852$

$X'_{2EH} = 0.3148$

$M_{iAA} = 88.15$ g/gmole

$M_{2EH} = 130.23$ g/gmole

$T = 298°K$

$\mu = 4.82$ cp (see Appendix 10.3)

$$\overline{M} = X'_{iAA}M_{iAA} + X'_{2EH}M_{2EH}$$

$$= (0.6852)(88.15) + (0.3148)(130.23) = 101.4 \text{ g/gmole}$$

$$D^{\circ}_{12} = (3.22 \times 10^{-8})[(1 \times 101.4)^{1/2}(298)(4.82)(18.9)^{0.6}]$$

$$= 3.48 \times 10^{-6} \text{ cm}^2/\text{sec}$$

10.6 Location of Original Data

Original data for this project are located in ORNL Databook A-5571-G, pp. 29-49, on file at the M.I.T. School of Chemical Engineering Practice, Bldg. 1000, ORNL. Results of the computer simulation are also on file at this location.

10.7 Nomenclature

A	constant in classical mass transfer correlation
B	constant in classical mass transfer correlation
C	constant used in alcohol viscosity determination
C_d	drag coefficient, dimensionless
C_i	saturated concentration of water in alcohol at sol-alcohol interface, gm/ml
C_o	concentration of water in bulk alcohol, gm/ml
ΔC	concentration driving force of water, gm/ml
D_p	sol particle diameter at any time, cm
D_{pf}	final diameter of gelled particle, cm
D_v	diffusivity, cm^2/sec
g	acceleration of gravity, cm/sec^2
K	mass transfer coefficient, cm/sec
L_{gel}	length of column required for gelation, cm
M	molecular weight of alcohol dehydrating agent, gm/gmole

M_{iAA}	molecular weight of iAA, g/gmole
M_{2EH}	molecular weight of 2EH, g/gmole
$\overline{M}$	average molecular weight, g/gmole
$\underline{M}_p$	molarity of the sol particle, gmole/liter
$\underline{M}_{pf}$	final molarity of the gelled particle, gmole/liter
N	mass flux of water from sol drop, gm/cm^2-sec
N_{oe}	Eotvos number, $\frac{g\ \Delta\rho\ D_p^2}{\sigma}$, non-dimensional
N_{Re}	sphere Reynolds number, $\frac{\rho\ V_t\ D_p}{\mu}$, non-dimensional
N_{Sc}	Schmidt number, $\frac{\mu}{\rho\ D_v}$, non-dimensional
N_{Sh}	Sherwood number, $\frac{K\ D_p}{D_v}$, non-dimensional
T	temperature, °K
V_t	terminal velocity of sol particles, cm/sec
V_b	molal volume of water at its boiling point, cc/gmole
X_{iAA}	volume fraction of iAA
X'_{iAA}	mole fraction of iAA
α	proportionality factor for Eq. (5)
β	constant used in Wilke-Chang correlation
θ	time, sec
θ_{gel}	gelation time, sec
μ	viscosity of alcohol, cp or poise
ρ	density of alcohol, gm/ml
ρ_p	density of particle at any time, gm/ml
ρ_{pi}	initial density of particle, gm/ml
ρ_{pf}	final density of gelled particle, gm/ml

ρ_w density of water, gm/ml

$\Delta\rho$ density difference between particle and fluid, gm/ml

σ interfacial tension, dynes/cm

$\emptyset$ association parameter, non-dimensional

10.8 Literature References

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