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STUDIES OF SMALL PARTICLE SUSPENSIONS

FOR L.M.F.R.

PART 1 - FLUID FLOW WITH SUSPENSIONS

SIMULATING THE U-Na SYSTEM

by

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Velocities of 2.5 to 2.9 feet per second, (76 to 88 centimeters per second), are needed to prevent settling of tungsten powder in a 1-in. i.d. horizontal pipe, from aqueous suspensions containing 6.1 to 7.0 percent by weight of tungsten. In one instance a narrow moving bed was observed at a velocity of 3.8 feet per second for a suspension containing 5.0 percent of tungsten, but the formation of a moving bed was not reproducible.

Settling has been observed at Reynolds numbers as high as 36,300.

The equation of Dallavalle, (4), (5), suitably modified, predicts settling velocities in a horizontal pipe of the same order as those found experimentally for dense solid, micron-sized particle suspensions.

For fully suspended flow, the friction factors lie approximately 10 percent above the smooth tube, normal liquid curve. It was not found necessary to consider non-Newtonian relationships to correlate the pressure drop data.

At mean Reynolds numbers above approximately 35,000 "streamlines" have been observed in the lower half of the pipe for fully suspended flow. This phenomena is discussed, but a full quantitative theoretical explanation is needed together with further experimental work.

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1. INTRODUCTION

This work was commenced to provide data for the LMFR project on the pumping, handling and settling properties of dense solid suspensions.

One fuel system under consideration for the LMFR consists of a mixture of uranium powder and sodium with an atomic ratio of 1 to 100 of uranium to sodium.

Tungsten metal powder as a suspension in water was chosen to simulate the uranium sodium system because the density ratios of the two systems are comparable. This ratio is important in determining settling velocities. Furthermore, the fluid viscosities of the two systems are of the same order, and also tungsten metal powder is readily available in sub-sieve size ranges.

There is a complete lack of experimental information in the literature on the dynamic behaviour of suspensions of the type which would be analogous to the uranium sodium system.

Various authors, (6), (13), claim that suspensions of particles less than 20- 30 microns behave as normal liquids above the normal critical Reynolds number range of 2000 - 3000. It is also stated in the literature that these particles do not settle out in a horizontal pipe as long as the flow is turbulent. However, no experimental confirmation of these statements can be found in the literature for solid to liquid density ratios of the order of 20 to 1.

There is no method by which the uranium sodium system can be predicted as being Newtonian or non-Newtonian in behaviour. The type of behaviour exhibited by a suspension is extremely important in pipe, pump and heat exchanger design and layout. If suspensions such as this can be shown to be normal or Newtonian, then equipment problems for fully suspended flow are very much simplified.

If the suspension is non-Newtonian it would mean that relationships such as those of Hedström, (7), or Metzner and Reed, (11), would be required to define the system.

The initial step therefore, in the simulated suspension work is to determine whether the suspension being studied is behaving as a normal liquid in fully suspended flow. At the same time, it is necessary to know the settling conditions in a horizontal pipe before studying the more difficult problem of settling in fittings, concentration gradients due to settling, and rate and amount of deposition of solid.

2. EXPERIMENTAL APPARATUS

The loop, called No. 1 tungsten water loop, consisted of a main tank, pump, industrial Pyrex glass tubing, horizontal metal metering tube, flow diverter, measuring vessel, control valves and manometers. The general arrangement is shown in Figure 1.

The main tank was a galvanised steel 45 gallon drum with a side outlet near the bottom of the drum. A side outlet was used to prevent clogging of the pump when the loop was closed down.

The pump was an Ajax CKH 1 bronze centrifugal pump with a 1-in. outlet which could deliver 30 gallons per minute against a head of 75 feet.

The metering tube consisted of an eleven foot length of 0.995 ± 0.001 in. i.d. copper tubing, to the end of which was butted a 15-in. length of industrial Pyrex glass tubing selected with the same diameter. This end section was used to determine the settling conditions in the copper tube. The distance between the pressure tappings in the copper tube was four feet and ample upstream and downstream lengths from these tappings were provided as calming sections.

Flow diversion was originally by flexible rubber hose to a weighing vessel. For the suspension runs, except for the lowest loop velocities, this was replaced by a measuring tank and flow diverter which consisted of a connected system of quick acting gate valves. A single arm movement diverted the flow instantly from the main tank to the measuring tank. The measuring tank was calibrated with water.

Considerable difficulty was experienced in maintaining the powder in suspension in the main tank. For the high velocity runs, a by-pass line from the pump as well as a 1 H.P. propeller type agitator were found necessary to keep the solids suspended in the main tank.

Two-fluid manometers were used to measure the pressure drop across the tappings. To cover the full flow range, mercury, carbon tetrachloride and chloro-benzene were used as metering fluids.

The tapping heads were connected to the metering tube as shown in Figure 2. The hole diameter in the metering tube was drilled to 0.154 in. and the internal surface was deburred. The tapping heads were designed to prevent the suspension travelling up the manometer lines after resetting the flow. The tapping heads were flushed out when necessary with clean water.

Occasionally, small air bubbles entrained in the main tank, entered the system and collected in the manometer lines. To avoid this, the metering tube was turned so that the centre line of the pressure-tapping holes made an angle of 30° with the vertical. However, in no instance during measurement was the concentration of air bubbles circulating significant.

The tungsten powder used was the HR1 grade supplied by Murex Ltd., Rainham, Essex, U. K. The material was supplied in seven lots and the mean particle size distributions as determined by the Andreasen pipette were as follows (3)

Stokes' Diameter Microns	Percentage Undersize	
	Samples A & B	Samples C, D, E, F and G.
15	87	90
10	78	83
5	60	62
4	54	53
3	45	42
2	33	29
1	18	15

3. RESULTS

To verify the experimental technique the apparatus was run with clean water. The results of the pressure drop runs are given in Table I and plotted in Figure 3. The results compare favourably with the standard curve for smooth tubes (15).

For an atomic ratio of uranium 233 to sodium of 1 to 100, the volume ratio at 500°C is 1 to 224, assuming a uranium density of 18.9 g/cc. The important comparable criterion is the volume concentration and for the tungsten powder-water system at 25°C the mass ratio corresponding to the 1 to 224 volume ratio is 1 to 11.6 of tungsten to water, or 7.9 percent by weight of tungsten.

Attempts were made to keep the mean concentration circulating at 7.9 percent, determined by analysis of the discharge. Considerable difficulty was experienced in achieving this as the tungsten rapidly settled in the main tank. With too vigorous agitation in the tank, considerable quantities of air entered the system.

The results for the settling velocities in the horizontal sections are given in Table II. Immediately before settling, streaming occurred. This appeared as striations close to, but not on the bottom of the tube. In some instances, the deposited solid could be seen moving as a bed along the bottom of the tube, but this condition was not reproducible. The settled condition was taken as the point at which stationary solid was first visually observed on the bottom of the sighting tube. This was determined by circulating the suspension at progressively smaller valve openings and taking a timed discharge before and after settling. Concentrations from 6.1 to 7.0 percent of tungsten were circulated, for the settling velocity determinations.

Table II gives the mean velocities before and after settling at each of the concentrations studied.

The pressure drop results for the suspensions are shown in Table III. For each pressure drop run Reynolds numbers and friction factors were calculated and plotted on Figures 6a and 6b. The densities used in the suspension runs were the mean densities calculated from the tungsten and water densities. The viscosity was calculated from the Orr and Dallavalle, (14), equation for suspensions -

$$\mu_s = \frac{\mu_1}{(1 - x_v/x_{vb})^{1.8}} \quad (1)$$

Where μ_s = viscosity of suspension, centipoise

μ_1 = viscosity of liquid, centipoise

x_v = volume fraction of solids in suspension

x_{vb} = "settled volume", or volume fraction occupied by the solid component in a bed formed by gravity sedimentation from the liquid.

The "settled volume" of tungsten in water was determined by shaking a weighed quantity of tungsten powder in a measured volume of water and allowing to stand until no further settling occurred. The results for "settled volume" are given in Table IV.

In the initial stages, it was noticed that the supernatant liquid from the suspension often had a blue coloration. This was shown to be colloidal tungsten.

No evidence of this blue coloration was seen during subsequent running, but the colour of the supernatant liquid at the completion of the runs was dark brown.

This supernatant liquid was subsequently analysed and contained 11.5 percent w/v tungsten as tungstate. Iron and zinc cations were present also.

DISCUSSION

(a) Settling Velocity in a Horizontal Pipe:

From Table II it can be seen that settling in a horizontal pipe occurs at velocities of 2.5 to 2.9 feet per second for the tungsten suspensions investigated.

Attempts were made to verify existing correlations for settling in a horizontal pipe.

Durand and Condolios, (6), derived a correlation for coal, sand and gravel for closely graded sizes greater than 50 microns.

$$V_L = F_L \left[2gD \frac{(\rho' - \rho)}{\rho} \right]^{1/2} \quad (2)$$

Where V_L = limiting velocity for deposition, cm/sec

g = acceleration due to gravity, cm/sec²

D = pipe diameter, cm

ρ' = density of solid, g/cc

ρ = density of water, g/cc

and F_L is a constant depending only on the concentration and size of the particles. This is not applicable in the present system because Durand's graph for F_L is not suitable for sub-sieve size particles.

Spells, (16), developed an equation for settling velocities for particles with specific gravities ranging from 2.0 to 2.6 g/cc, and particle size ranging from 80 micron to 820 micron.

$$v_m^{1.225} = 0.0251 g d \left[\frac{D \rho'}{\eta} \right]^{0.775} \frac{(\rho' - \rho)}{\rho} \quad (3)$$

Where v_m = minimum velocity to prevent settling, cm/sec

g = acceleration due to gravity, cm/sec²

d = particle diameter such that 85% by weight are smaller than d cm

D = pipe diameter, cm

- ρ' = density of slurry, g/cc
- η = slurry viscosity (taken equal to the viscosity of the medium), g/(cm)(sec)
- δ = density of solid, g/cc
- ρ = density of medium, g/cc

Substituting the 85 percent under size value for the diameter, as determined by the Andreasen pipette, (3), yields a velocity of 29.6 centimeters per second or 0.97 feet per second.

Newitt, et alia, (13), have developed equations for transition velocities for the various regions they define.

The regions defined are :-

1. Homogeneous Flow - concentration uniform over pipe cross section.
2. Heterogeneous Flow - concentration not uniform over pipe cross section.
 - (a) Fully suspended Flow - all particles in fully suspended state
 - (b) Suspended flow with moving bed
 - (c) Suspended flow with layer of particles sliding over a stationary deposit - particles moving in saltation.
 - (d) Stationary deposit with superimposed ripples
 - (e) Isolated deposits.

These authors studied materials with specific gravities ranging from 1.18 to 4.60 and with closely graded particle sizes ranging from minus 240 mesh to 3/16in. but covered mainly the larger size ranges.

The transition velocity from homogeneous to heterogeneous flow, V_H , is given by Newitt as -

$$(V_H)^3 = 1800 gDW \quad (4)$$

where

g = acceleration due to gravity, cm/sec²

D = diameter of pipe, cm

W = terminal velocity, cm/sec

The transition velocity from heterogeneous flow to flow by sliding bed or saltation, V_B , is given by

$$V_B = 17W \quad (5)$$

In the present system, it was found that the flow did not follow all these regions as defined by Newitt. It was not possible to experimentally define region (b) or (d) in the present work. The equations derived by Newitt are not applicable for mixed sizes as a single terminal velocity cannot be assigned.

Murphy, et alia, (12), studied suspensions of glass beads, steel particles and lead particles in water. No explicit equation was found for the lower transition velocity, defined by the authors as the velocity at which a stationary layer of particles is formed. For the lead particles of diameter 0.0505 in. lower transition velocities of 2.9 to 4.8 feet per second in a $\frac{1}{2}$ -in. diameter pipe are reported.

Abraham, et alia, (1), investigated a UO_2 -NaK slurry system up to temperatures of $600^\circ C$ in two loops constructed of $\frac{1}{2}$ -in. stainless steel tubing. Using a radiometric technique, it was found that at velocities greater than 2.1 and 2.4 feet per second, between $25^\circ C$ and $450^\circ C$ respectively, all the UO_2 was in suspension. At temperatures above $500^\circ C$ however, fall out of UO_2 particles was observed. Slurry stability up to $600^\circ C$ was finally achieved by the addition of uranium to the system.

Hitchon, (8), used a radiometric method to determine the distribution of solids for $ThO_2 - H_2O$ slurries circulating in a $1\frac{1}{2}$ -in. nominal bore stainless steel pipe. The velocity at which settling was first detected within 15 minutes of reducing the flow rate, was 1.4 feet per second, for both the 0.27 and 0.61g $ThO_2/g H_2O$ slurries studied.

Dallavalle, (4), (5), has given the following equation for the transport velocity of cinders, carbon, anthracite and quartz in air :-

$$V = 6000 \left[\frac{\rho}{\rho + 1} \right] d^{2/5} \quad (6)$$

Where V = transport velocity, ft/min

ρ = specific gravity of solid

d = diameter of the largest particle, in.

The maximum particle specific gravity was 2.65 giving a density ratio of about 2000 to 1, and the particle sizes varied from 0.04 to 0.16 inches.

The d in equation (6) refers to the maximum particle diameter present. This diameter is not available from the Andreasen pipette analysis of the tungsten powder in the present work. However, substitution of the 85 percent undersize diameter of 12.5 microns in equation (6), gives a transport velocity of 4.5 feet per second.

The 85 percent undersize diameter of the UO_2 used by Abraham, et alia. (1), was 3.5 microns, determined by a sedimentation method. Substitution of this into equation (6) gives 2.6 feet per second.

In Hitchon's work, (8), the 80 percent undersize value of 3 microns is quoted. Using this as an approximation for the 85 percent value in their loop, yields a settling velocity of 2.4 feet per second, calculated from equation (6). In this work the particle size analysis was determined by a photoextinction sedimentometer.

The following table gives the experimental velocities and the calculated velocities obtained from the modified Dallavalle equation. These data apply only for suspensions of density ratios of greater than 9 to 1 and 85 percent undersize diameters of less than 20 microns.

	Experimental ft./sec.	Calculated ft./sec.
This investigation Tungsten water	2.5 - 2.9	4.5
Abraham, et alia. (1) UO_2 - NaK	2.1 - 2.4	2.6
Hitchon, (8) ThO_2 - water	1.4	2.4

Considerably more experimental data are needed to verify Dallavalle's equation and also to verify the use of the 85 percent undersize diameter. It appears that this method, however, is the best available to date for predicting settling velocities for micron-sized particles forming suspensions with density ratios greater than 9 to 1. No agreement was found between the experimental figures of Durand, Spells, Newitt and Murphy and the calculated figures obtained from the Dallavalle equation.

It must be remembered that the 85 percent undersize diameter may vary depending on the method used for particle size analysis. It appears, from work done by Jarrett and Heywood, (10), that sedimentation techniques such as the Andreasen pipette, photo extinction, hydrometer, divers and manometer methods, yielding Stokes' diameter, give similar results. There appears to be no correlation between sedimentation techniques and microscopic counting for micron-sized particles, and this is the subject of other investigation.

It is considered that the diameters of tungsten particles determined by the fall of the particles through water, as done in the Andreasen pipette, more likely approaches the effective diameters of the particles during pumping in the loop. However, the effect of pumping causing breakdown of any agglomerates is unknown.

A good deal of experimental work is needed to determine the relationships between settling velocities in a horizontal pipe and such variables as pipe diameter, particle size and distribution, densities of solid and liquid, concentration of solid and agglomeration of particles.

There is no theory available at present to predict quantitatively settling velocities. A theoretical development is planned for future attention.

(b) Settling Velocity in a Vertical Pipe :

Velocities required to lift particles are those given by the terminal velocities calculated from Stokes' or Newton's Laws, (2), for ideal conditions.

The terminal velocity calculated from Stokes' Law is proportional to the square of the diameter of the particle. If agglomeration or flocculation exists then the actual terminal velocity can be many times the calculated figure.

The terminal velocity of a spherical tungsten particle, 20 microns in diameter falling in water at 25°C is 0.013 feet per second. The lowest velocity used in the loop was 0.5 feet per second where severe settling of the tungsten powder occurred in horizontal sections. Although the equipment was not designed to study vertical settling velocities, it was observed that at these low loop velocities the particles were moving up the vertical sections in the direction of flow.

Worster and Denny, (18), found that for 3-in. coal in a vertical six inch pipe, a mean velocity of twice the settling velocity is adequate to transport the solid. They also state that velocities must be determined from the requirements of any horizontal or sloping sections in the pipelines. These velocities will always be large in comparison with those sufficient for safe vertical lifting. This has been the case in the present work.

(c) Pressure Drop :

Worster, (17), Durand and Condolios, (6), and Newitt, et alia, (13), observed for diminishing velocities, that the head loss for thin slurries reached a minimum and then increased compared to that for clean water. The minimum head loss corresponded to the settling condition.

Figure 4 shows the clean water runs plotted as head loss versus velocity. With the tungsten suspensions, divergence from the water curve was observed as shown in Figure 5, but no minimum velocity was obtained. The divergence commenced in the region where settling occurred.

The results were also plotted as friction factor versus Reynolds number in Figures 6(a) and (b). The fully suspended data are given in Figure 6 (a) and lie approximately 10 percent above the clean water curve.

The increase in friction factor for fully suspended flow could be explained by the presence of tungstates in solution. Dissolved solids increase the viscosity more than the density with a resultant reduced Reynolds number, with a nett effect of shifting the points closer to the normal liquid curve. This was partially confirmed by Experiment 7. In this experiment the tungsten had been washed by decantation and the points for suspended flow fell on the standard curve. This is also confirmed by Figure 5.

For settled conditions the results are less consistent and diverge rapidly, as seen in Figure 6 (b). This could be due to the following reasons which would cause the increased friction factors :-

- (a) Increased viscosity near the bottom of the tube due to settling of the solid to the bottom.
- (b) Reduced cross-sectional area for flow.
- (c) Wide velocity differences across the tube due to the concentration of tungsten being greater in the bottom half.

It may well be that (a) and (c) are the major factors, as what appeared to be laminar flow in the bottom half was observed as streaming at very high Reynolds numbers, as mentioned previously. Further work is needed on the variation of concentration across the tube to clarify this point.

Hitchon and Murdoch, (9), investigated the flow of aqueous thoria suspensions. They concluded that for Reynolds numbers between 80,000 - 500,000 and, that up to concentrations of 610g/Kg of water thoria slurries may be treated simply as true fluids with a density computed on the basis of the weights of thoria and water in a given volume, and with a viscosity equal to that of water at the same temperature. The pressure drop in a given pipe can then be calculated with a reasonable degree of accuracy from the usual friction factor versus Reynolds number correlation.

It is noticed that the clean water results obtained by Hitchon and Murdoch lie about 10 percent above the known standard for water, which would be somewhat unexpected. Also very few runs for the clean water and also the slurries were made to support their conclusions. Using the Orr and Dallavalle viscosity equation in their results for slurries gives closer agreement with the clean water standard curve.

(d) Streamline and Turbulent Flow :

Durand and Condolios, (6), and Newitt, et alia, (13), state that homogeneous transport occurs with particles of less than 20 or 30 microns. These workers claim that the suspensions of particles of these diameters, in water, flow like a homogeneous fluid and begin to settle under laminar conditions, where the Reynolds number is less than 2000-3000.

In the tungsten water suspensions, the Reynolds number corresponding to a settling velocity of 2.9 feet per second and a concentration of 6.2 percent, is 35,600. It appears that the classification of flow conditions for suspensions as proposed by Durand and Newitt is not valid in this case. Homogeneous transport is not a single function of diameter alone.

Newitt states that the critical value of the Reynolds number for the transition from streamline to turbulent flow for his homogeneous suspensions is the same as for liquids, provided the density and viscosity terms relate to the suspension.

It has been observed in the present work that layering or streaming near the bottom of the tube was occurring at Reynolds numbers above approximately 35,000, which is many times the pure liquid value for laminar flow. If this is interpreted as true streamline flow, then it appears as if both streamline and turbulent flow are occurring simultaneously as the settling condition is reached.

The "streamlines" observed were not uniform over the tube cross section and were never noticed in the upper half of the tube. It is possible that the denser medium near the bottom of the tube caused by the settling of the tungsten is flowing in streamline flow while the region above it is fully turbulent.

It is also likely however, that the particles of tungsten in the "streamlines" are moving at streamline velocities, while the liquid around them is fully turbulent.

It is hoped that equipment now being set up to measure the concentration gradient across the cross section of the tube by radiometric methods, will provide a better understanding of this problem, as well as enable a method of controlling the concentration changes to be developed.

5. FURTHER WORK

The following additional work should cover :-

- (a) Radiometric investigation of the concentration gradient across the pipe before and after settling.
- (b) Theoretical development for dense solid settling conditions in a horizontal pipe.
- (c) Studies of suspensions analogous to UBe_{13} in Na such as precipitated barium sulphate in water.

- (d) Comparison of sedimentation and microscopic methods of particle size analysis.

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7. REFERENCES

1. Abraham, B. M., H.E. Flotow and R.D. Carlson, "UO₂ - NaK Slurry Studies in Loops at 600° C". Amer. Soc. Mech. Eng., 2nd Nuclear Engineering and Science Conference, Paper No. 57 - NES C - 104, (1957).
2. Brown, G.G., "Unit Operations", 1st ed., p. 74, John Wiley and Sons, Inc., (1950).
3. deBruin, H. J., and R.C. Cairns, "Particle Size Analysis of Tungsten Metal Powder". - AAEC/E. 12, (1957).
4. Dallavalle, J. M., "Determining Minimum Air Velocities for Exhaust Systems". Heat, Pip. and Air Cond., 4, 639, (1932).
5. Dallavalle, J. M., "The Theory and Practice of Pneumatic Conveying", Heat. and Ventil., 39, 28, (1942).
6. Durand, R., and E Condolios, "Proceedings of a Colloquium on the Hydraulic Transport of Coal". (National Coal Board, London), p. 39, (1952).
7. Hedstrom, B.O.A., "Flow of Plastic Materials in Pipes". Ind. Eng. Chem., 44, 651, (1952).
8. Hitchon, J.W., "Use of a Radiometric Technique to Investigate the Settling of Aqueous Thoria Slurries". AERE. CE/M 188, (1956).
9. Hitchon, J.W., and R. Murdoch, "The Pumping of Aqueous Thoria Slurries". AERE CE/M 189, (1957).

10. Jarrett, B.A. and H. Heywood, "A Comparison of Methods for Particle Size Analysis", Brit. J. Appl. Phys., Suppl. No. 3, S21, (1954).
11. Metzner, A.B. and J.C. Reed, "Flow of Non-Newtonian Fluids - Correlation of the Laminar, Transition and Turbulent-flow Regions". A.I.Ch. E. Journal, 1, 434, (1955).
12. Murphy, G., D.F. Young and R.J. Burian, "Progress Report on Friction Loss of Slurries in Straight Tubes". USAEC, ISC-474, (1954).
13. Newitt, D.M., J.F. Richardson, M. Abbott and R.B. Turtle, "Hydraulic Conveying of Solids in Horizontal Pipes", Trans. Instn. Chem. Engrs. 33, 93, (1955).
14. Orr, C., and J.M. Dallavalle, "Heat Transfer Properties of Liquid-Solid Suspensions". Chem. Eng. Progress Symposium Series, Vol. 50, No. 9, p. 29, (1954).
15. Perry, J.H. "Chemical Engineers' Handbook", 3rd ed., p. 382, McGraw-Hill Book Company Inc., (1953).
16. Spells, K.E., "Correlations for use in Transport of Aqueous Suspensions of Fine Solids through Pipes". Trans. Instn. Chem. Engrs., 33, 79, (1955).
17. Worster, R.C., "Proceedings of a Colloquium on the Hydraulic Transport of Coal". (National Coal Board, London), p. 5, (1952).
18. Worster, R.C. and D.F. Denny, "The Hydraulic Transport of Solid Material in Pipes". Proc. Inst. Mech. Eng., 169, 563, (1955).

