DOE/PC/90012-4

Hydrodynamics of Three-Phase Slurry Fischer-Tropsch Bubble Column Reactors

Quarterly Technical Progress Report
for the Period 1 April 1988 - 30 June 1988

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July 29, 1988

Prepared for the Pittsburgh Energy Technology Center,
the United States Department of Energy Under Contract No. DE-AC22-86PC90012
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NOTICE

PATENT STATUS

TECHNICAL STATUS

TABLE OF CONTENTS

I. Abstract

During the past quarter all modifications to the small stainless steel column circuit were completed. The pump was rebuilt and reinstalled. A new purgeless DP system was designed and fabricated. Initial results using this system with cold-flow runs indicated no operational problems. A new procedure was developed for analyzing the solids concentration in slurry samples withdrawn from hot-flow runs. It reduced the time required for analysis significantly. The percent difference between the actual solids concentration and the solids concentration determined from the new procedure was less than 3% (relative) for all samples analyzed.

Work on Task 3 (Measurement of Hydrodynamic Properties by Conventional Techniques) was continued during the past quarter. Four additional cold flow runs were conducted to test the performance of the new DP system, following which hot-flow studies were initiated. A total of thirteen experiments with FT-300 wax were conducted. Ten were conducted with solids and 3 were conducted without solids. Of the ten experiments performed using solids, nine were made using 0-5 μm iron oxide particles, and one with 45-53 μm iron oxide particles. Results from these experiments indicate that for a given system, the superficial liquid velocity has a greater effect on the average gas hold-up, compared to the effect of solids concentration. For experiments conducted in the batch mode of operation the axial solids distributions decreased with an increase in height above the distributor; whereas for experiments conducted in the continuous mode of operation the distributions remained essentially uniform. The axial gas hold-ups increased with height above the distributor for all runs. The increase was more pronounced for experiments conducted in the batch mode of operation. Some problems with solids settling were encountered during these runs. Minor modifications have been initiated to improve solids suspension in the storage tank.

Operational problems with the pump were encountered following the run with large

iron oxide. At the beginning of the next run, the seal separating the slurry from the cooling water failed, flooding the system with the coolant. The problem was corrected and the pump was brought back into service. However, during the next experiment, with 0-5 μm silica particles, it was not possible to obtain the desired liquid flow rate with the pump. Inspection of the pump revealed a chipped idle gear. A new replacement gear has been ordered.

We were unable to have the large iron oxide and silica particles sized by an outside company within our budgeted costs. We therefore initiated sizing on our own using a Sweco vibrating classification unit. Preliminary tests have been conducted to determine the rate at which the raw material can be classified into the desired size range.

Work on Task 4 was continued during the past quarter. All items needed to construct the movable assembly mechanism have been ordered, and majority of these items have been received. Fabrication of the support structure for the axial movement system has been completed. An application for a license permitting the use of radioactive material has been submitted. The high energy (Cesium-137) source holder was modified so that it can be mounted perpendicular to the column. The source holders have been approved for use by the Radiological Safety Office. A new data acquisition system has been ordered.

II. Objective and Scope of Work

The overall objective of this contract is to determine the effects of bubble column diameter, solids loading and particle size, and operating conditions (temperature, gas and liquid flow rates) on hydrodynamics of slurry bubble columns for Fischer–Tropsch synthesis, using a molten wax as the liquid medium. To accomplish these objectives, the following specific tasks will be undertaken.

Task 1 – Project Work Plan

The objective of this task is to establish a detailed project work plan covering the entire period of performance of the contract, including a detailed program schedule, analytical procedures, and estimated costs and manhours expended by month for each task.

Task 2 – Design and Construction of the Experimental Apparatus

The existing glass and stainless steel columns (0.051 m and 0.229 m in diameter, 3 m tall) that were constructed under our previous DOE contract (DE–AC22–84PC70027), will be modified and additions made in order to study the effect of continuous upward liquid flow. After the procurement of equipment and instrumentation, and construction of the unit is completed, a shakedown of test facilities will be made to verify achievement of planned operating conditions.

Task 3 – Measurement of Hydrodynamic Parameters by Conventional Techniques

In this task, the effects of operating conditions (liquid and gas superficial velocities, temperature), gas distributor, column diameter, and solids concentrations and particle size on hydrodynamic parameters in the glass and stainless steel columns will be determined. All experiments will be conducted using nitrogen at atmospheric pressure.

The hydrodynamic parameters that will be determined as a function of the independent variables mentioned above are: average gas hold–up, axial solids distribution, axial gas hold–up, flow regime characterization, and qualitative information on bubble size distribution.

Task 4 – Application of a Gamma Radiation Density Gauge for Determining Hydrodynamic Parameters

The objective of this task is to determine hydrodynamic parameters for the three–phase system using a nuclear density gauge apparatus. A movable assembly mechanism and positioning racks for the two nuclear density gauges and detectors will be designed and constructed. Following the interfacing of the apparatus with an on–line microprocessor, the gauges will be calibrated using pure components (liquid wax and solid particles), and with known proportions of liquid and solid. After calibration, the following parameters will be obtained from experiments in the large stainless steel column: axial gas hold–up, axial concentration of solids, and qualitative information on flow regimes and bubble size distributions.

III. Summary of Progress

During the past quarter, a new larger slurry storage tank for the small columns was installed and the 1/2" slurry feed lines were replaced with 1" lines. A calibration tank for slurry flow rate measurements was installed. The pump was rebuilt and mounted vertically (below the new storage tank) to prevent the settling of solids. Several different DP cell configurations were tested and a purgeless system appeared to perform the best. The high temperature Sensotron transducer was received and mounted on the small stainless steel column.

A new procedure (based on the Archimedean Principle) was developed to determine the weight fraction of solids in slurry samples obtained from hot-flow studies. The procedure was tested with samples having known quantities of solids (both silica and iron oxide). Solids concentrations determined by the new procedure were within 3% (relative) of the actual weight fractions. The percent relative differences were higher at low solids concentrations and were less than 1% for concentrations higher than 15 wt%. The new procedure is much simpler and faster than the filtration method previously employed.

Work on Task 3 (Measurement of Hydrodynamic Properties by Conventional Techniques) was continued during the past quarter. A total of seventeen runs were conducted in the small stainless steel column. The first four runs used tap water as the liquid medium, and the primary purpose of these experiments was to test the new purgeless DP system. No operational problems with the DP system were encountered during these experiments. Average gas hold-ups from these runs compared favorably with those obtained from similar experiments conducted in the glass column during the previous quarter. Following the successful testing of the DP system we initiated hot-flow studies in the small stainless steel column.

Thirteen hot-flow experiments were conducted in the small stainless steel column.

The first three experiments were conducted using FT-300 wax with no solids. One of these was conducted in the batch mode, and the other two in the continuous mode of operation using superficial liquid velocities of 0.005 and 0.02 m/s, respectively. The same format was repeated with slurries containing 10, 20, and 30 wt% of 0-5 μm iron oxide in FT-300 wax. The last experiment was conducted at a liquid velocity of 0.005 m/s with 10 wt% 45-53 μm iron oxide particles in FT-300 wax. All experiments were conducted using superficial gas velocities of 0.02, 0.04, 0.06, 0.09, and 0.12 m/s at a temperature of 265 °C. The 2 mm orifice plate distributor was used for all runs. A spreadsheet program was developed for analyzing the data.

Regardless of the system studied (i.e. different solids concentrations), the average gas hold–up was significantly higher for experiments conducted in the batch mode of operation. The gas hold–ups obtained at a liquid velocity of 0.02 m/s were consistently lower than the gas hold–ups obtained at a liquid velocity of 0.005 m/s; however, the difference was not significant. Axial gas hold–ups increased with an increase in height above the distributor. There was a substantial increase in the axial gas hold–up in the uppermost section of the column for experiments conducted in the batch mode which may be attributed to the presence of foam. In general, the average gas hold–up decreased with an increase in solids concentration for all gas velocities and slurry velocities employed. The difference in gas hold–ups between experiments without solids and with 10 wt % solids was large; however, the decrease in hold–up between 10 and 20 wt % or between 20 and 30 wt % was fairly small. For experiments conducted in the batch mode, the axial solids concentration decreased with an increase in height above the distributor. For experiments conducted in the continuous mode of operation, the solids concentration profiles remained fairly uniform. The superficial gas velocity did not have a significant effect on the solids concentration profiles.

The experiment with large iron oxide particles indicated a significant amount of solids settling. Solids recovery was very low, both in the storage tank and in the column. The maximum weight fraction of solids in the system was approximately 6 % for a slurry having 10 wt% solids. This was observed at the bottom of the column at a gas velocity of 0.12 m/s. Following this experiment, another experiment was initiated using the same system except a liquid velocity of 0.02 m/s was employed. Upon starting the experiment, the seal in the pump separating the cooling water from the slurry failed and coolant flowed into the system. Upon examination of the pump, it appeared that some iron oxide particles became wedged between the graphite bushing and metal seal plate. The graphite bushing and metal seal plate were cleaned and reinstalled.

Another experiment was initiated with 0 – 5 μm silica particles. Initially, the pump seemed to function properly. However, four hours after the pump was turned on, we could no longer maintain the required liquid flow rate, regardless of the pump setting. Thus, the experiment was terminated. Upon examining the pump, the idle gear was found to be chipped. We have ordered a new one.

Some minor modifications to the storage tank have been initiated to try and improve the solids suspension. These modifications include the use of nitrogen purge lines to help suspend the solids.

The surface tension apparatus was modified to accommodate high temperature measurements. We have measured the surface tension of fresh FT–300, used FT–300 from two of the experiments conducted last month, fresh FT–200, used FT–200 from our previous two–phase project, fresh Sasol wax, and used Sasol wax from our previous two phase project. Surface tension measurements were conducted at temperatures of 200 and 260 °C with two measurements at each temperature. There was not a significant effect of aging on the surface tension of FT–300. However, the surface tension of used Sasol wax was approximately 18 % lower than the surface tension of fresh Sasol wax. The surface tension of the FT–200 wax was approximately the same as that of the

FT–300 wax.

We were unable to have the large iron oxide and silica particles sized by an outside company within our budgeted costs. An 18 inch diameter Sweco sieve has been loaned to us for the purpose of sieving large particles. We purchased 325 and 400 mesh sieves. We plan to use 37 – 44 μm particles as opposed to 45 – 53 μm particles to reduce the amount of wear on the pump. Some initial experiments have been made to determine the percent of raw material in the desired range, as well as the rate at which the material can be processed. Based on our initial tests, approximately 8 – 10 wt % of the raw material is in the desired range.

Work on Task 4 was continued during the past quarter. All items needed to construct the movable assembly mechanism have been ordered. The majority of these items have been received. Fabrication of the support structure for the axial movement system has been completed. An application for a license permitting the use of radioactive material has been submitted. The high energy (Cesium-137) source holder was modified so that it can be mounted perpendicular to the column. The source holders for the high energy Cesium-137 and low energy Americium-241 sources have been approved for use by the Radiological Safety Office. A new data acquisition system has been ordered from MetraByte. The new data acquisition system will be used in conjunction with a Zenith 286 computer for recording data from the two nuclear density gauges and the high temperature Sensotron pressure transducer.

IV. Detailed Description of Technical Progress

A. Task 3 – Measurement of Hydrodynamic Parameters by Conventional Techniques

During the past quarter, we have finalized the operating procedure and data reduction procedure. These procedures are given in Appendix A.

A.1. Physical Property Measurements

The Fischer Model 215 Autotensiomat (Surface Tension Analyzer) was modified for high temperature applications as suggested by the manufacturer. Surface tension measurements were made using FT–300, Sasol, and FT-200 wax at temperatures of 200 and 260 °C. In order to check for reproducibility of our measurements, the surface tension was measured twice at each condition. The difference in surface tension values between the two measurements was less than 0.001 N/m for all cases.

In order to determine if the age of the wax had a significant effect on the surface tension, measurements were also made with used FT–300 and used Sasol wax. Table 1 contains our results for the surface tension values of the various waxes we analyzed, as well as, Mobil's (Kuo et al., 1985) result for FT–200 at 260 °C and Deckwer's (Deckwer et al., 1980) results for paraffin wax at 220 and 260 °C.

Three samples of used FT–300 wax were analyzed, two from our current project (Runs F160-FES10-2S and F260-FES30-2S) and one from our previous two–phase project (Run 2-4; Contract DEAC–22–84PC70027). The samples analyzed from our current project were from the last of the six experiments with each batch of wax. Both of these samples had iron oxide particles present (10 and 30 wt.% respectively). The surface tension results compared favorably with our results for fresh FT–300 wax. However, the surface tension of used FT–300 from our previous project (Run 2–4) was lower than the surface tension of fresh FT–300 wax. At 200 °C, the surface tension of used FT–300 was 14 % lower than the surface tension of fresh FT–300, and at 260 °C, the surface tension was 8.3 % lower. The wax sample from Run 2-4 was heated for approximately 340 hours; whereas, the samples from Runs F160-FES10-2S and F260-FES30-2S were heated for approximately 144 hours. Thus, the age of the wax was significantly different, and this might be the reason for the difference in surface tension values for the two waxes.

A similar trend was observed with used Sasol wax (i.e. the surface tension was lower for used Sasol wax (Run 8–3)). The evaporation rate for Sasol is significantly higher than that for FT–300. We believe that with time on stream, the lighter components (having relatively high surface tensions) evaporate leaving the heavier components (having relatively low surface tensions) in the wax. This could explain the lower surface tensions observed for used Sasol wax.

The surface tension of FT–200 was approximately the same as FT–300 at both 200 and 260 °C. However, our result at 260 °C was considerably lower than the value reported by Mobil (i.e. 0.0185 N/m as compared to 0.024 N/m). At the present time, we have no explanation for the difference in the surface tension values. The method used by Mobil to determine the surface tension of FT–200 was not specified in their report (Kuo et al., 1985).

A.2. Overview of Bubble Column Operations

During the past quarter, modifications of the small stainless steel column circuit, initiated towards the end of the quarter before, were completed. The new (larger) slurry storage tank for the small (2" diameter) columns was installed, and replacement of half inch lines in the slurry feed system with one inch lines was completed. The old storage tank was modified and relocated for use as a solvent tank for cleaning the column. The calibration tank, to be used for slurry flow rate measurements, was installed in the slurry return line from the top of the column. The replacement parts for the slurry pump were received and the pump was rebuilt and mounted vertically, directly below the new storage tank.

During our earlier experiments in the small stainless steel column, we had experienced some problems with the plugging of the DP cells with solid particles. We therefore decided to test different options that would improve the system.

The first option was to install a sintered metal plate filter at the end of a 0.5" diameter tube. The tube was then attached to the column wall, with the filter element in contact with the slurry. A nitrogen purge was used to prevent any backflow of liquid into the tube. The other end of the 0.5" diameter tube was connected to the DP cell. We then tested the system with a slurry of 0-5 μm silica (20 wt.%) in water. The system appeared to function fairly well initially, however, with time on stream, the DP readings increased significantly, and the DP system continued to indicate positive pressures even after the column was emptied. The readings went back to zero after about ten minutes following the emptying of the column. This indicated that the filter plate, in contact with the slurry, trapped some of the solid particles which in turn increased the pressure drop for the purge flowing through the sintered plate. Following the removal of the slurry from the column, the gas purge cleared the trapped solids from the plate and restored the readings back to zero.

We then designed a purgeless system to measure differential pressures. This consisted of a 0.25" diameter tube attached to the column wall. The other end of the tube was attached to a chamber (approximately 20 cc in volume) and the DP cell was attached to this chamber (see Figure 1). When the column is filled with a slurry, the nitrogen trapped in the chamber acts as a buffer between the hot slurry in the column and the low-temperature DP cell. The chamber also serves as a trap for any slurry that flows into the 0.25" tube. A drain located at the bottom of the chamber is used to clean the trap between runs. A ball valve located in the 0.25" line serves to isolate the system from the column, in case the trap has to be emptied during a run. The 0.25" line (including the ball valve) and the chamber are heat traced and insulated to prevent solidification of slurry in this section. Following a successful test of this system with a slurry of 20 wt.% of 0-5 μm silica in water, four additional units were fabricated and installed on the small stainless steel column. The system functioned very well during four cold-flow and thirteen hot-flow experiments conducted in the small column. When the traps were drained between runs, the total slurry removed from all five traps was between 15-20 cc.

The high-temperature Sensotron transducer was received during the past quarter and was mounted on the small stainless steel column. The transducer was encased in a heating block to maintain it at the system temperature. During the first six experiments with the hot wax slurry, differential pressures at five different locations were obtained using the low-temperature Validyne pressure transducers (purgeless system). During subsequent runs, one of the Validyne transducers was replaced with the high-temperature Sensotron transducer. The transducer performed fairly well and raw pressure data from it were very consistent with no unexpected trends.

The slurry pump was brought into service for the first time, during the hot-flow studies, following the replacement of the worn out parts. During eight runs conducted in the continuous mode of operation with 0-5 μm iron oxide (10, 20 and 30 wt.%), and one run with 45-53 μm iron oxide (10 wt.%), no mechanical problems were encountered. However, there were some problems in adjusting the pump speed to obtain the desired slurry flow rate, particularly in runs conducted using a superficial liquid velocity of 0.005 m/s. The slurry flow rates required in the small column are at the lower end of the pump's capacity, making flow control difficult. In addition to this, the flow rate also varied with the pressure head in the column, requiring pump speed adjustments following every change in gas velocity (pressure head in the column is a function of gas hold-up in the column which in turn depends on the gas flow rate). The control was better when the pump was set at a higher speed and some of the discharge was circulated back to the storage tank via the bypass line. Slurry flow control was much easier for experiments conducted using a superficial liquid velocity of 0.02 m/s. Solids did not appear to settle in the pump during runs conducted using the 0-5 μm iron oxide at all three solids concentrations employed. However, during the run made with 10 wt.% of 45-53 μm iron oxide we experienced problems with the settling of solids in the storage tank. Some minor modifications of the storage tank have since been initiated to improve the suspension of solids. These include the addition of nitrogen purge lines at the tank bottom. This would agitate the slurry in this region and suspend any solids that are settled.

During the second run with 45-53 μm iron oxide ($u_\ell = 0.02$ m/s), the pump malfunctioned. The pump seal, located between the chamber containing the hot process stream and the chamber containing the cooling water, broke and cooling water flooded the system. The run had to be aborted and the pump was dismantled for inspection. Some of the large iron oxide particles were wedged in the seal and had caused the seal to open up, however, there was no permanent damage to the seal unit itself. The pump was reassembled and reinstalled for further experiments. We decided to conduct experiments with 0-5 μm silica first instead of using the 45-53 μm iron oxide. The pump appeared to function normally when the first experiment was initiated, however, after about 4 hours on stream, it became increasingly difficult to obtain the desired liquid flow rate even though the pump setting was increased. We decided to abort the run and reinspect the pump. The idle gear (made of graphite) had chipped and needed to be replaced. We believe that the problem originated during the trial runs with water (conducted during the previous quarter), and the problem worsened with each run and lead to the present malfunction. We have ordered the required parts but have not received them yet.

The slurry withdrawal system worked smoothly after some initial problems with plugging of the withdrawal lines. We increased the temperature of the withdrawal lines and cleaned the lines manually before each sample withdrawal. This greatly simplified the withdrawal procedure. The slurry samples were allowed to cool in the sampling cylinders and the solid slurry plugs were then removed and analyzed for solids fraction. During the first run with solids (10 wt.% of 0-5 μm iron oxide), we initiated the filtration procedure to determine the weight fraction of solids, however, we had to abandon the procedure after the first set of samples were analyzed. This is because the filtration procedure at high temperatures was tedious and took too long for analyzing each set of samples (there are five sets of samples withdrawn per run). We subsequently developed a much simpler, faster and accurate method to determine the weight fraction of solids in the slurry samples. The new procedure took between five to ten minutes per set of samples compared to about 1.5 to 2 hours for the filtration procedure.

This procedure involves the calculation of the solids weight fraction in slurry from an estimated value of the density of a solidified slurry sample. An accurate estimate of the sample density is obtained using the sample weight, measured on a precision balance, and the sample volume, measured using the Archimedean Principle (the apparent loss in weight of a solid body, when completely immersed in a liquid, equals the weight of the displaced fluid). The procedure used is outlined below.

The slurry withdrawn into the sampling cylinder is allowed to cool and solidify. The solid slurry plug is then removed for solids fraction determination. The sample is first weighed on a precision balance ($m_{s\ell}$). It is then suspended with a thin wire from a support structure placed on the balance and the combined weight of the support structure and sample recorded (m_1). The sample, while still suspended from the support structure, is then completely immersed in a beaker of acetone and then balance reading recorded (m_2). The three measured quantities along with the known densities of solidified wax (ρ_w), solids (ρ_s), and acetone (ρ_{acet}), are used to determine the weight fraction solids (W_s) in the slurry sample, by

$$W_s = \frac{\rho_s - \left(\frac{\rho_w \rho_s}{\rho_{acet}}\right)\left(\frac{m_1 - m_2}{m_{s\ell}}\right)}{\rho_s - \rho_w} \tag{1}$$

The density of solidified wax was determined experimentally, and was found to be 0.95 g/cc. Acetone was selected as the liquid medium for this procedure since it is lighter than wax (ρ_{acet}=0.792 g/cc), it evaporates quickly from the sample surface, and the solubility of wax in acetone is negligible. The procedure was first tested with samples containing known quantities of solids. Samples containing 3, 7, 10, 15, 20, 25, 30 and 35 wt.% solids in wax were prepared with 0-5 μm iron oxide, 45-53 μm iron oxide, and 0-5 μm silica. The mean error between the actual and estimated weight fraction solids for these samples was less than 1 % (relative). The error was highest for samples containing 3 wt.% solids (3% relative) and was as low as 0.2% relative at 35 wt.% solids. A density of 5.1 g/cc for iron oxide and 2.65 g/cc for silica was used in the calculations. Sensitivity analysis of eqn (1) revealed that the results were very sensitive to small variations in the density of solidified wax. A variation of only 1% in the density of wax caused a 12% change in the value of W_s, whereas a 5% change in the density of solids caused only a 2% change in W_s for slurries containing iron oxide, and a 4% change for slurries containing silica. Because of the high sensitivity to wax density, we determined the density of fresh wax and of used wax (which had been on stream for over 100 hours) and found the two values to be the same. We therefore believe that the accuracy of this procedure will not decrease when solids fractions are determined for slurries that have been on stream for extended periods of time.

During the past quarter we obtained all of the 0-5 μm iron oxide that will be required during the course of the project. Samples of large iron oxide and silica were sent to Vortec Corp. of Long Beach, CA, and the cost and time estimates for grinding/classification of the powders were obtained. They were unable to classify solids into the exact size range of 45 – 53 μm; approximately 75 – 80 % would be in the desired range. The estimates for obtaining this cut were $8000 for each type of solids; whereas, we have only $2000 budgeted for this purpose. We therefore decided to do the classification ourselves. An 18" diameter Sweco vibrating classification unit has been loaned to us for the purpose of classifying large particles. With this unit we can process larger quantities of solids per batch. Because of our problems with the pump during the run with the 45-53 μm iron oxide, we have decided to use particles in the size range 37-44 μm instead of the 45-53 μm particles. We have purchased the necessary sieves (325 and 400 mesh) to obtain the desired size range. Some initial experiments have been made with large iron oxide to determine the percent of raw material available in the desired size range, and to determine the rate at which the material can be processed. These tests indicate that the raw material contains approximately 8-10 % of solids in the desired size range.

A.3. Experimental Results

During the past quarter, a total of 17 runs were conducted in the small stainless steel column. Four of these were conducted using water as the liquid medium, and the remaining 13 were conducted using FT-300 wax as the liquid medium. The results from these experiments are discussed in this section

A.3a. Cold-flow studies:

The four cold-flow experiments were conducted in the batch mode. The primary objective of these experiments was to test the new purgeless DP system in the presence of solids. Tap water was used as the liquid medium in all four runs. One experiment was performed for each combination of solids type and solids size. All experiments were conducted using 20 wt % solids. The column was initially charged with the desired amount of water, and solids were added from the top of the column. Slurry samples were withdrawn from four locations along the column height to obtain solids concentration profiles. The weight fraction of solids was determined by evaporating the water from the sample.

Average gas hold-ups were determined and compared to results obtained in the 0.051 m ID glass column under similar operating conditions. Results obtained in the SS column for small silica (0-5 μm) particles and large iron oxide particles (45-53 μm) compare favorably with the results obtained in the glass column. For experiments conducted with small silica, relatively low values of gas hold-up were obtained (approximately 22 % at a gas velocity of 0.12 m/s). However, the results from experiments with large iron oxide indicated the presence of a substantial amount of foam (ϵ_g was approximately 48 % at a gas velocity of 0.12 m/s).

The gas hold-ups obtained from the experiment with small iron oxide particles in the SS column were lower than those obtained in the glass column under similar operating conditions for $u_g > 0.04$ m/s (see Figure 2). The results from the stainless steel column agreed with the results from the glass column at gas velocities of 0.02 and 0.04 m/s; however, at higher gas velocities the hold-ups in the stainless steel column appeared to be lower than those in the glass column. This is probably due to the approximations involved in estimating the gas hold-up in the upper most section of the dispersion. The hold–up in the upper most section is estimated by extrapolating the profile for the sections below. For cases where foam is present at the top of the dispersion (i.e. in the upper most section), extrapolation of the profile from the sections below (where foam is absent and hold-ups are relatively low), would lead to the underestimation of gas hold-up for the upper most section. Whereas, in the glass column, hold-ups are estimated from visual observations of the expanded height (including the actual amount of foam present). Therefore, for cases where foam is present in both columns and expanded heights are equal, estimates of hold-up for the stainless steel column would be lower than those for the glass column.

Figures 3 and 4 show the effect of particle size on average gas hold-up for experiments conducted in the SS column. In general, the average gas hold-up increased with an increase in particle size. The increase in hold-up due to large iron oxide particles was substantial (see Figure 3); whereas, the increase in hold-up associated with the large silica particles was not as significant (see Figure 4). At the present time we do not know why iron oxide particles induce foaming.

A.3b. Hot-flow studies:

Thirteen successful experiments were conducted using FT-300 wax at 265°C. In addition to these two more runs were initiated, but had to be aborted because of operational problems. Results from the first twelve runs are discussed here. Four of the experiments were conducted in the batch mode of operation, and the remaining experiments were conducted in the continuous mode of operation using the rebuilt slurry pump for recycling the slurry. Two batches were used for the first twelve experiments, with six runs made with each batch of wax as was proposed in the matrix of experiments (see Quarterly Technical Progress Report for the period 1 October, 1987 - 31 December, 1987). The first three experiments were conducted using FT-300 wax with no solids. One of these was conducted in the batch mode, and the other two in the continuous mode of operation using superficial liquid velocities of 0.005 and 0.02 m/s respectively. The same format was repeated with slurries containing 10, 20 and 30 wt.% of 0-5 μm iron oxide in FT-300 wax. The thirteenth experiment was conducted in the continuous mode using a 10 wt.% slurry of 45-53 μm iron oxide in FT-300 wax and a superficial liquid velocity of 0.005 m/s.

No major operational problems were encountered in the first twelve runs. The temperatures of the various system components remained fairly stable during the experiments. The axial hold-up profiles for the three phases were determined for all experiments (two phases for runs with pure FT-300 wax). The gas and liquid phases account for most of the dispersion volume and only a small volume is occupied for by the solids phase. Therefore, the axial gas hold-up profiles are complementary to the axial liquid hold-up profiles. The axial solids hold-up profiles are mainly determined by the axial weight fraction solids profiles. Therefore, only the axial gas hold-up profiles and the axial weight fraction solids distributions are presented for each of the twelve runs. These are included in APPENDIX B. The average gas hold-ups were determined for all runs using the procedure outlined in the data reduction section (APPENDIX A). Results from the different runs are categorized in terms of the various effects and are discussed here. Wax and solids inventories were also made for runs conducted with the second batch of wax. The discrepancies in these balances and the possible causes are discussed briefly.

Effect of liquid velocity on average gas hold – up :

Figure 5 shows the results from experiments conducted with pure FT-300 wax. Foam was produced for the batch case ($u_\ell=0$) with gas hold-up values as high as 28% at a superficial gas velocity of 0.04 m/s. However, gas hold-up values decreased significantly for superficial gas velocities in the range 0.04-0.09 m/s when the experiment was conducted in the continuous mode of operation using a superficial liquid velocity of 0.005 m/s. A further decrease was observed when the superficial liquid velocity was increased to 0.02 m/s, however the change was less significant. Average gas hold-up values at $u_g = 0.02$ m/s from all three runs are very similar. At this gas velocity the flow regime is expected to be similar for all three cases (predominantly homogeneous bubbly flow). At $u_g = 0.12$ m/s hold-ups from all three runs once again approach the same value. The foam produced with FT-300 wax, while operating in the batch mode, appears to have dissipated at this gas flow rate, and hold-ups have converged to values similar to those for the continuous mode of operation. Our earlier experiments in the glass column (Bukur et al., 1987) have shown that slug flow dominates for gas velocities in this range, and hold-ups are determined by the size of these slugs.

Figure 6 compares results obtained for pure FT–300 wax for experiments conducted under similar conditions in the batch mode of operation. Results from Runs 1-7 and 6-1 (from our previous contract, DEAC–22–84–PC70027), Run FT-2 (Quarterly Technical Progress Report for the period January 1, 1988 - March 31, 1988), and Run F110-00000-2S are included in this figure. The first two runs were conducted in the 0.051 m ID glass column and the last two runs were conducted in the 0.051 m ID stainless steel column. Hold-up values obtained from Run 6-1 show a transition from the foamy to the slug flow regime at a gas velocity of 0.05 m/s, whereas, hold-up values from the other three runs show no such behavior. There is excellent agreement in results from all experiments for gas velocities less than 0.05 m/s. At gas velocities greater than 0.05 m/s the gas hold–ups obtained in the various runs are significantly different. For Run 1-6, where foam broke, hold-ups are the lowest. The poor reproducibility of results from experiments conducted using FT-300 wax in the batch mode of operation, is primarily due to differences in the amounts of foam produced during each run. In general, hold-up values obtained from experiments in the 0.051 m ID stainless steel column are within the range of values obtained during our previous project from runs conducted in the 0.051 m ID glass column.

Figure 7 shows average gas hold-up values from the three runs made with a slurry of 10 wt.% 0-5 μm iron oxide in FT-300 wax. The trends are qualitatively similar to those for pure FT-300 wax, i.e. the highest gas hold-ups are observed with no liquid circulation and the lowest values were obtained at a superficial liquid velocity of 0.02 m/s. Once again the largest difference was observed when the liquid velocity was increased from 0 to 0.005 m/s. Results for 20 and 30 wt.% 0-5 μm iron oxide (Figures 8 and 9) show the same trends in gas hold-ups with changes in liquid velocity.

The above results indicate that liquid circulation tends to lower gas hold-ups, however, further increases in liquid flow rates do not have a significant effect on the average gas hold-up values. The significant difference in hold-ups between the batch mode of operation and continuous mode of operation is because of the changes in the foaming characteristics of the medium. For the batch case, the foam accumulates at the top and increases the gas hold-up. However, as slug flow sets in, the foam tends to disperse and the hold-up profiles tend to flatten out. In the continuous mode of operation, any foam that tends to accumulate at the top is carried back down by the circulating slurry and dispersed. With an increase in the liquid circulation, this rate of dispersion increases, decreasing the average gas hold-up.

Effect of solids concentration on average gas hold – up :

Figures 10, 11 and 12 illustrate the effect of solids concentration on the average gas hold-up for runs made with the 0-5 μm iron oxide, for different superficial liquid velocities.

Figure 10 shows the average gas hold-up values for experiments conducted in the batch mode of operation (u_ℓ=0). Our earlier experiments with iron oxide and water (Quarterly Technical Progress Report for the period Januar 1, 1988 - March 31, 1988) have shown that in the batch mode, hold-ups with solids were higher than values obtained in the absence of solids. The same trend continues to be seen for experiments conducted using FT-300 wax as the liquid medium. The data included in Figure 11 are for slurries containing 0, 10, 20 and 30 wt.% solids. Hold-ups increase as solids are increased from 0 to 10 wt.% (except at u_g=0.04 m/s). A further increase in gas hold-up is observed when solids concentration is increased to 20 wt.%. At 30 wt.% solids, gas hold-ups decrease, however, they are still higher than those at 0 wt.% solids. The lower gas hold-up at u_g=0.04 m/s at 10 wt.% solids could be due to the inability of the DP cells to detect the presence of foam in the top most section of the column.

Figures 11 and 12 show the effect of solids concentration on gas hold-up for superficial liquid velocities of 0.005 and 0.02 m/s, respectively. In Figure 11 the differences in hold-up values for the various slurries are not as significant as differences observed in experiments conducted in the batch mode of operation (Figure 10). However, in general, gas hold-up at a given gas velocity decreases as solids concentration increases. The highest hold-up values are observed with pure FT-300 wax and the lowest are obtained with a slurry containing 30 wt.% solids. This trend is the reverse of that observed in the batch mode experiments (Figure 10). The effect of solids concentration is even less significant at a superficial liquid velocity of 0.02 m/s (Figure 12), however the trends are qualitatively similar to those observed at u_ℓ=0.005 m/s (Figure 11).

The above results show some interesting trends. For the batch mode of operation, the addition of solids increases the amount of foam and produces higher hold-ups, however, for the experiment in the continuous mode, addition of solids decreases gas hold-ups. This implies that the foaming tendency of the liquid medium is greatly reduced when a small upward liquid flow is used. This small liquid flow is sufficient to disperse the fine bubbles that are responsible for foam, and only the hydrodynamics of the actual dispersion (i.e. without the foam) determines the gas hold-up.

Axial gas hold – up profiles :

The axial gas hold-up profiles at gas velocities of 0.02, 0.04 and 0.12 m/s are shown in Figures B-1 through B-12. The three profiles for each run clearly illustrate the effect of superficial gas velocity on the axial gas hold-up. For all runs, the axial hold-ups increased with increases in gas velocity, which is expected, since the overall average gas hold-up increases with superficial gas velocity. The results also show that gas hold-up increases with an increase in height above the distributor for all cases. The increases are more significant at higher superficial gas velocities than the increases at lower gas velocities. For experiments conducted in the batch mode of operation (u_ℓ=0), the foam at the top of the dispersion is responsible for the significantly higher hold-ups in the upper section, compared to the values obtained in the continuous mode of operation. In general, the axial gas hold-up profiles are affected by the superficial liquid velocity. At u_ℓ=0 (batch case), the profiles are non-uniform (low hold-ups at the bottom of the column and higher values at the top). However, as u_ℓ is increased to 0.005 m/s, the profiles are less steep and become fairly uniform at u_ℓ=0.02 m/s. The trends are qualitatively similar for each slurry (i.e. 0, 10, 20 and 30 wt.% solids).

Axial solids distribution profiles :

The axial solids distribution profiles for runs conducted using the 0-5 μm iron oxide are shown in Figures B-4 to B-12. The profiles are uniform for all runs made in the continuous mode of operation and no significant effect of gas velocity is observed. For experiments conducted in the batch mode of operation, higher solid concentrations are present at the lower end of the column than at the top end. These results once again illustrate the effect of liquid velocity on the solids distribution profiles. When no liquid circulation is present, the dispersion of solids is poor, however, even a small liquid flow rate is sufficient to suspend the 0-5 μm iron oxide particles uniformly.

Wax and solid inventories :

For experiments conducted with the second batch of wax (20 and 30 wt.% 0-5 μm iron oxide), it was possible to make wax and solids balances for individual runs. The hold-up profiles in the column, together with the weight fraction and volume information for the slurry in the storage tank, were used to make the balances. The solids inventory showed that approximately 25% of the solids were unaccounted for after the very first run with this batch of wax. However, the absolute weight of solids unaccounted for remained the same for all six runs conducted with this batch of wax, even though the weight fraction of solids was increased to 30 wt.% following the third run with this batch of wax. This could be caused by settling of solids in the storage tank as soon as they were added to the molten wax before the first run. These solids could have been transported to the column when the slurry was first introduced and deposited on top of the distributor. No further settling took place after this point in time and the remaining solids were accounted for during the next five runs. In order to test this hypothesis, the column was not cleaned after the aborted run with 10 wt.% 0-5 μm silica, and the distributor was removed. The plug of solidified wax slurry in the three inch section between the distributor and the slurry feed line was removed and analyzed for solids concentration. The sample had a solids concentration of around 40-50 wt.%, indicating settling of solids in this zone. We have since initiated some minor modifications in the storage tank and to the bottom of the column to reduce the extent of settling.

Wax inventories for the various runs were as expected. With time on stream, the quantity of wax unaccounted for increased. This is because the wax evaporates during the experiments and is carried out with the exit gas. At the end of the sixth run with this batch of wax approximately 8% of the wax was unaccounted. We had to add wax to the storage tank following the third run with each batch to make up for the lost wax.

A.3c. Comparisons between cold-flow and Hot-flow studies:

The effect of liquid medium and superficial liquid velocity on average gas hold-ups is shown in Figures 13 and 14. Figure 13 compares the results obtained with pure FT–300 wax at liquid velocities of 0.0, 0.005, and 0.02 m/s to those obtained with distilled water from an experiment conducted in the batch mode of operation. The effect of liquid velocity on gas hold–up for pure water is negligible. An increase in liquid velocity for experiments conducted with wax causes a decrease in gas hold-up. As the liquid velocity is increased, the average gas hold-ups in experiments with wax tend to approach the gas hold-up values associated with distilled water. The same trend was observed for experiments conducted with 20 wt % iron oxide (see Figure 14). Foam was observed in all runs conducted in the presence of solids leading to higher hold-ups.

B. Task 4 – Application of a Gamma Radiation Density Gauge for Determining Hydrodynamic Parameters

During the past quarter, we ordered the remaining material needed to construct the movable assembly mechanism which will be used to transport the nuclear density gauges along the column. This includes the ball screws which will be used to move the density gauges axially and radially along the column, and the ball nuts which will be used with the radial movement system. We have received all previously ordered parts and have completed the construction of the axial support system. The axial support system consists of four 2" diameter by 125" long support shafts, four support platforms (two located at the top of the column and two secured to the floor), and two platforms which will be used to support the radioactive sources and detectors.

A control panel has been made which contains the controllers for the two motors which will be used to move the nuclear density gauges axially and radially along the column, and the nuclear electronics. The wiring for the controllers and motors has been completed.

The high energy source holder we received from Catalytic, Inc. was modified so that it can be mounted perpendicular to the column. Once the source holder was modified, both the low energy source holder and high energy source holder were brought to the radiological safety office for examination. The sources were placed in the holders and radiation leak tests were performed on all sides of the source holders. The results from the tests indicated that no additional shielding of the source holders would be required.

During this quarter, we submitted our application for a license permitting the use of radioactive material. We are currently waiting for approval of the license.

The data acquisition system we received from Catalytic, Inc. is not adequate for our needs. Neither the Fortran compiler nor operating system are present. We have ordered a new data acquisition system from MetraByte, which will be used in conjunction with a Zenith 286 personal computer for obtaining data from both nuclear density gauges, and the Sensotron pressure transducer.

V. NOMENCLATURE

a	slope of the DP cell calibration curve
b	intercept of the DP cell calibration curve
d_c	column diameter (m)
h	height above the distributor (m)
m	weight (Kg)
P	pressure (m water)
u_g	superficial gas velocity (m/s)
u_ℓ	superficial liquid velocity (m/s)
W_s	weight fraction solids in slurry

Greek letters

ϵ	hold-up
ρ	density (Kg/m^3)

Subscripts

acet	acetone
g	gas
ℓ	liquid
s	solids
$s\ell$	slurry
t	column top
w	wax

VI. REFERENCES

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Deckwer, W.D., Louisi, Y., Zaidi, A. and Ralek, M., "Hydrodynamic properties of the Fischer-Tropsch slurry process," Ind. Eng. Chem. Proc. Des. Dev., 19, 699 (1980).

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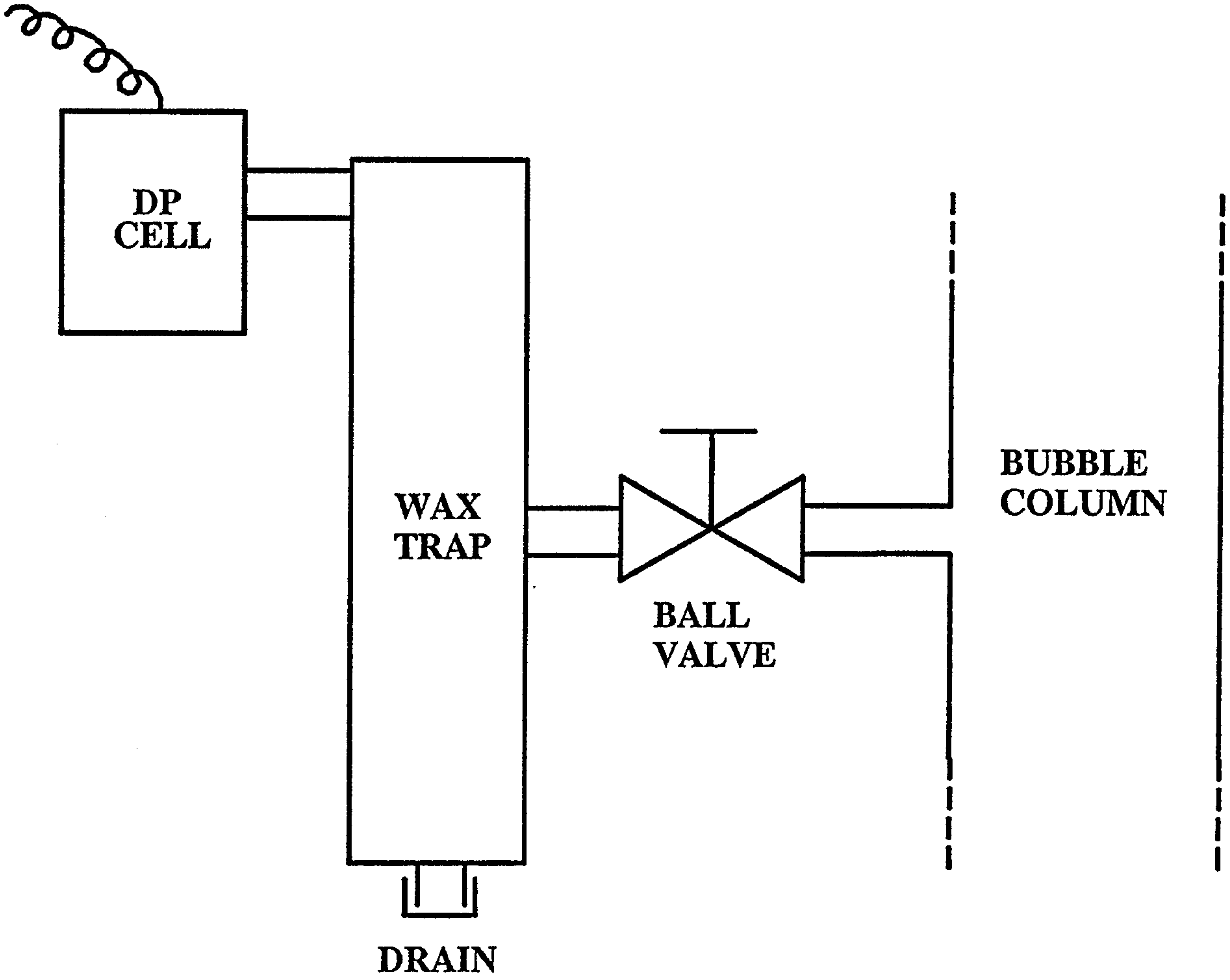
Table 1. Surface tension of waxes

LIQUID	RUN NUMBER	200 °C	260 °C
FT–300 (Fresh)	N/A	21.5	18.5
FT–300 (Used)	F160–FES10–2S	21.5	18.0
FT–300 (Used)	F260–FES30–2S	21.5	18.5
FT–300 (Used)	2–4	18.5	17.0
SASOL (Fresh)	N/A	18.5	17.0
SASOL (Used)	8–3	17.0	14.0
FT–200 (Fresh)	N/A	21.0	18.0
FT–200 (Mobil)	N/A		24.0
Paraffin (Deckwer)	N/A	24.0 (220 °C)	21.5

N/A: Not Applicable

Figure 1. Purgeless DP system



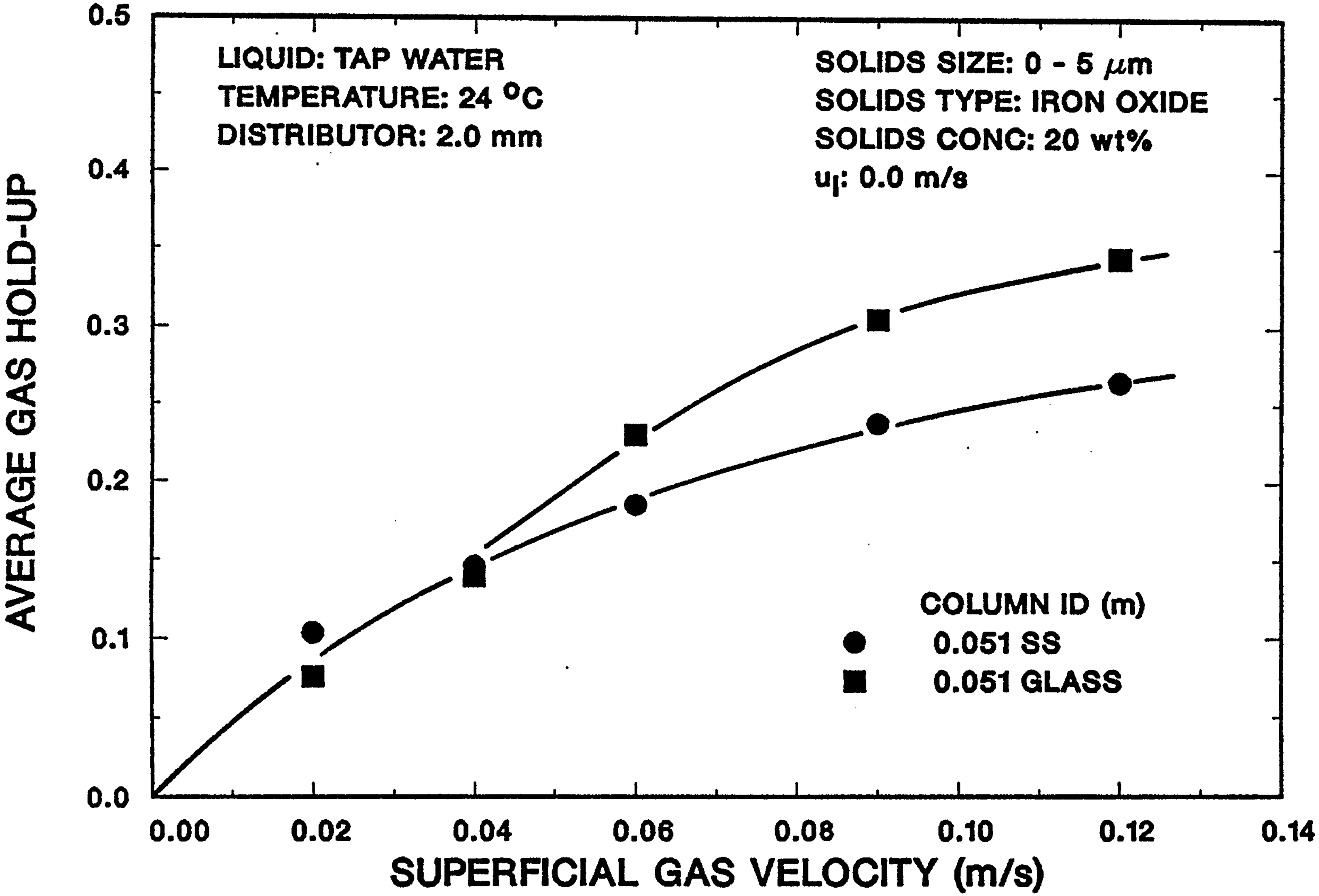


Figure 2. Effect of superficial gas velocity on average gas hold-up.

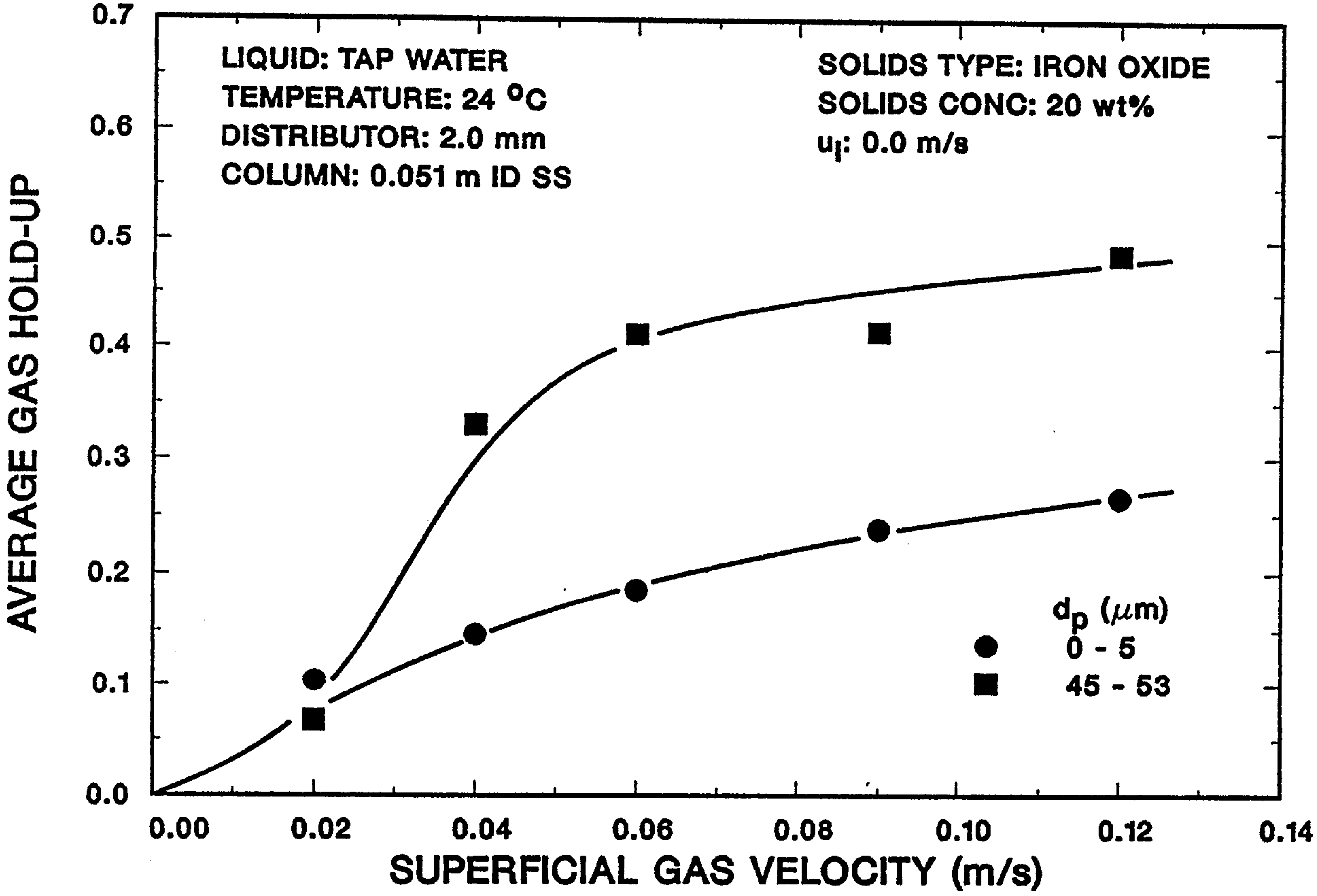


Figure 3. Effect of particle size on average gas hold-up.

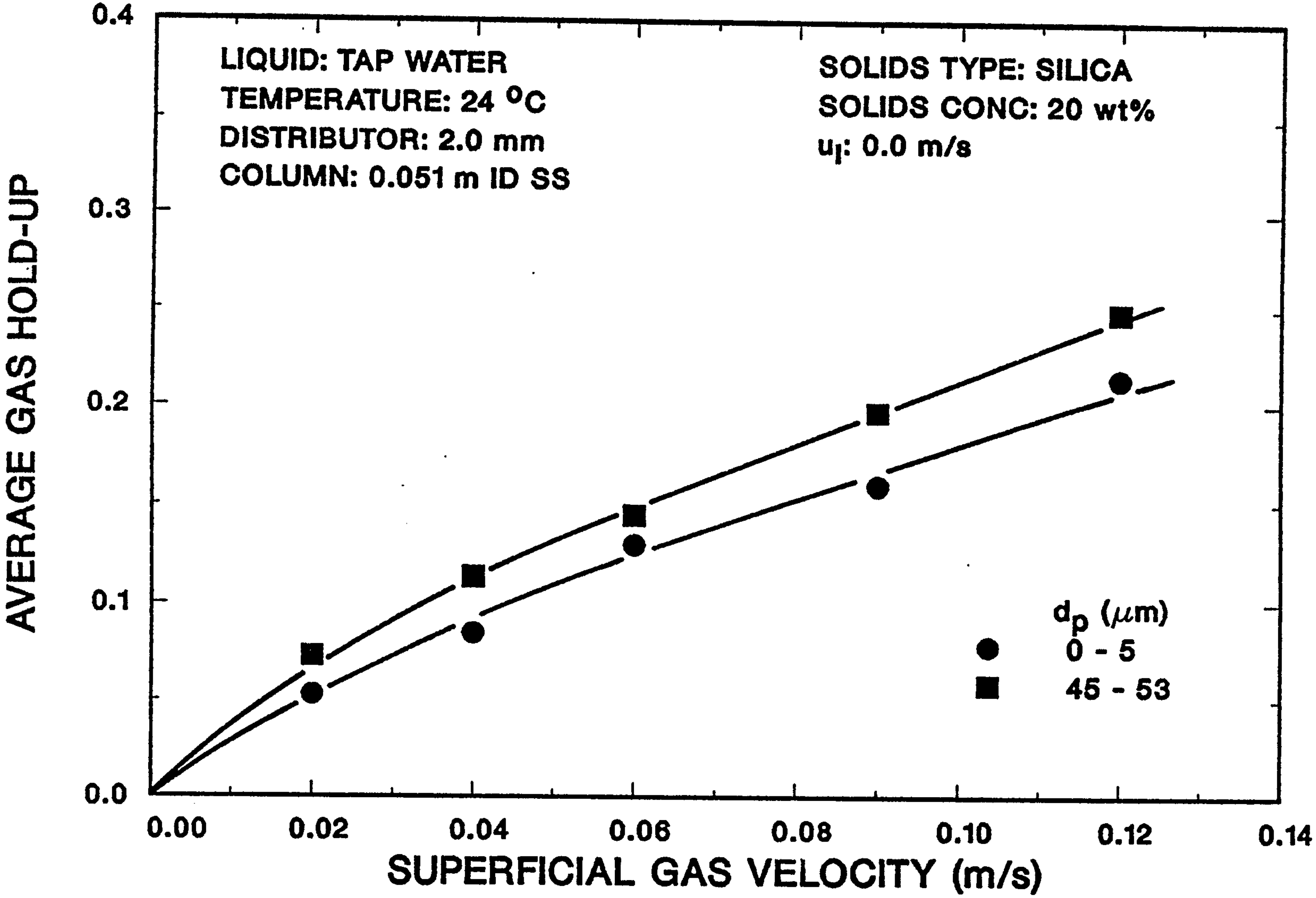


Figure 4. Effect of particle size on average gas hold-up.

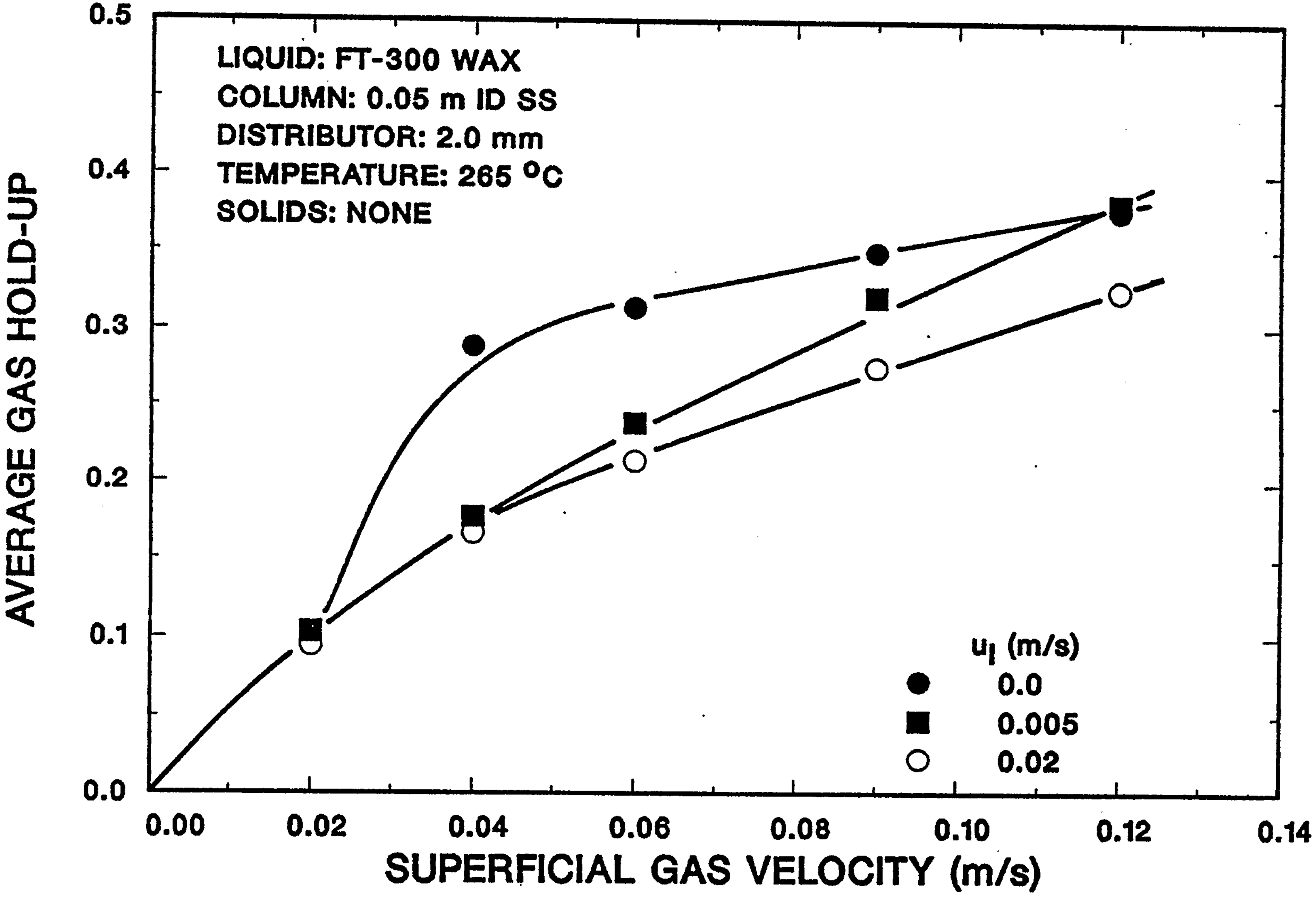


Figure 5. Effect of liquid velocity on average gas hold-up.

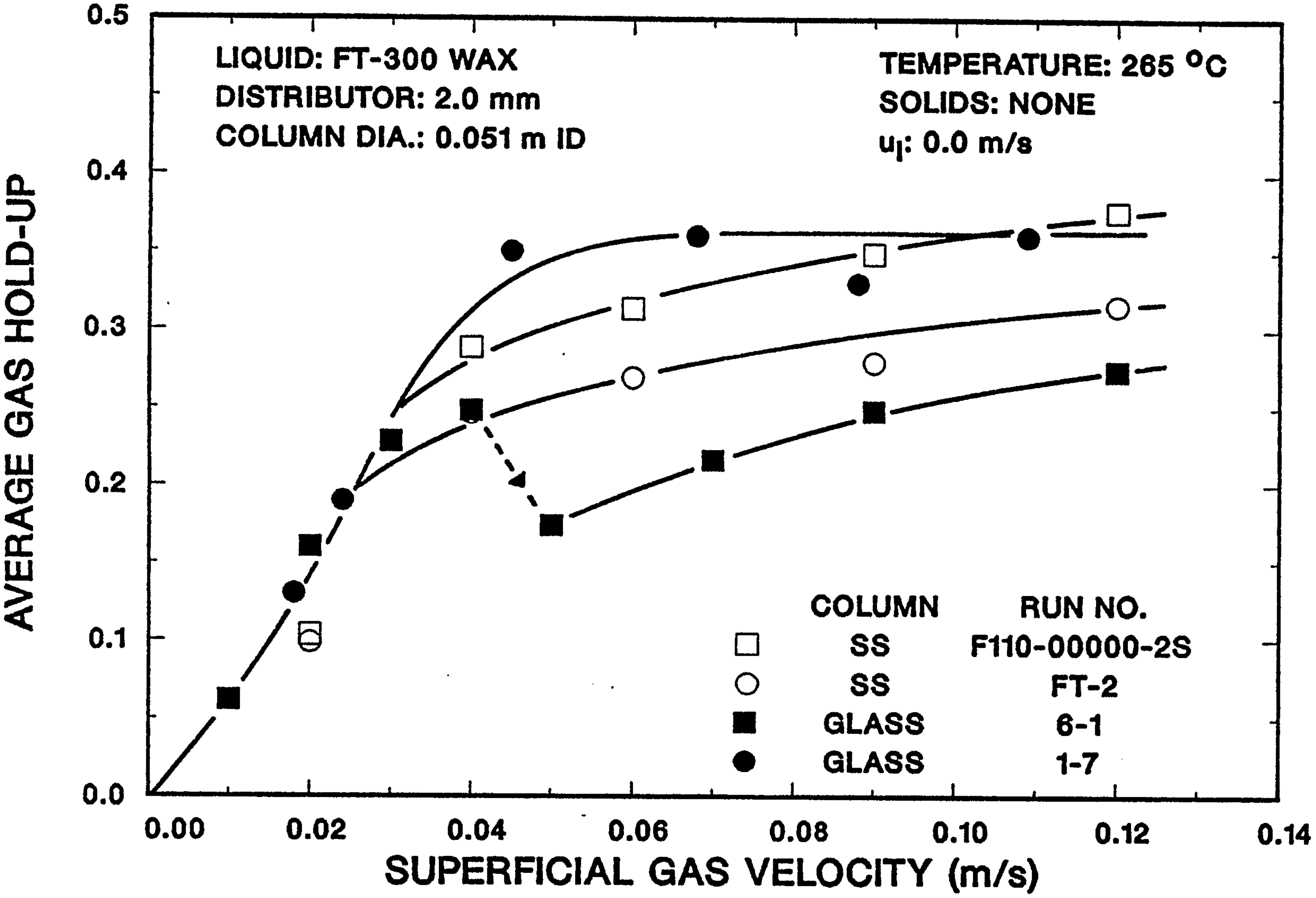


Figure 6. Reproducibility of results for average gas hold-up.

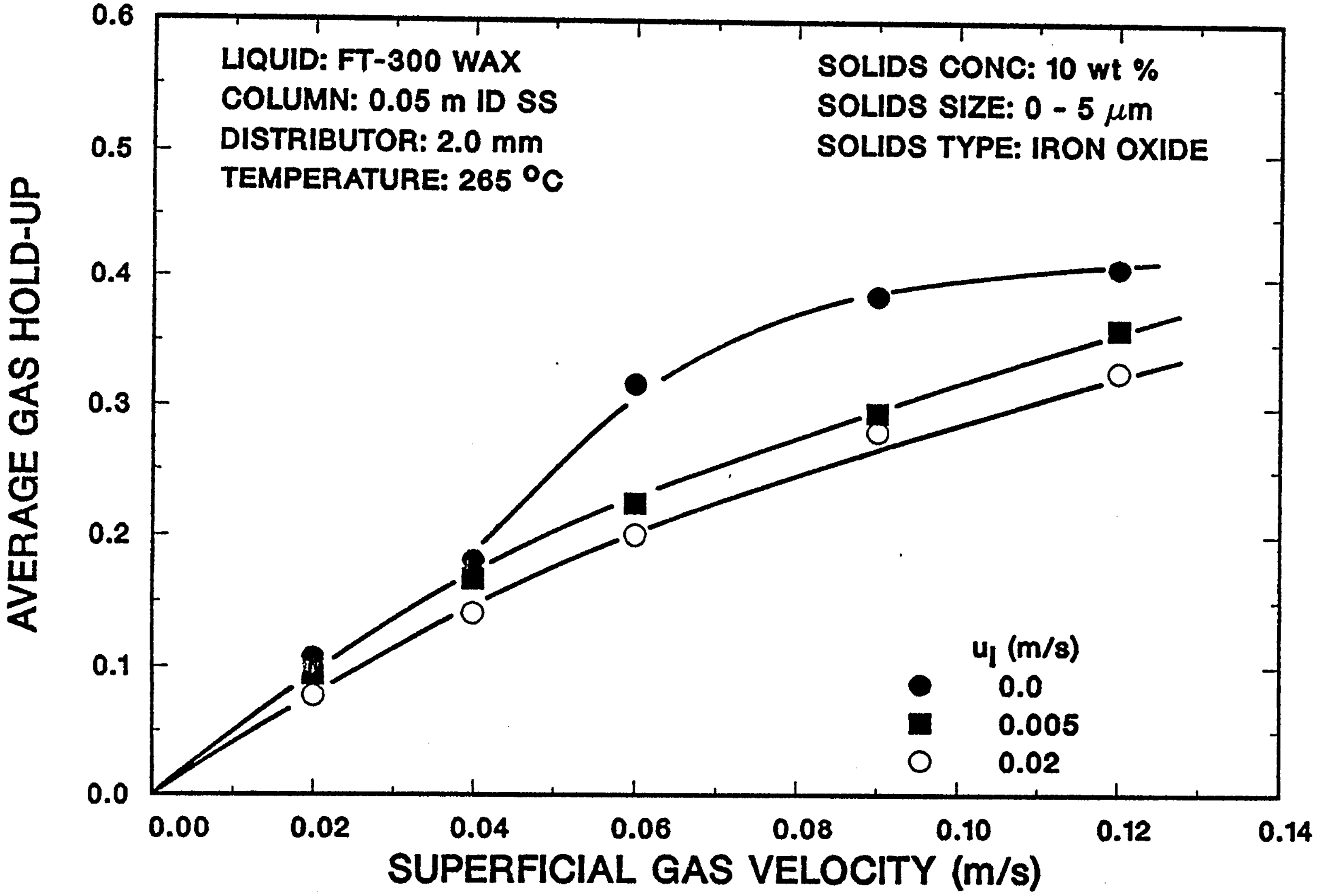


Figure 7. Effect of liquid velocity on average gas hold-up.

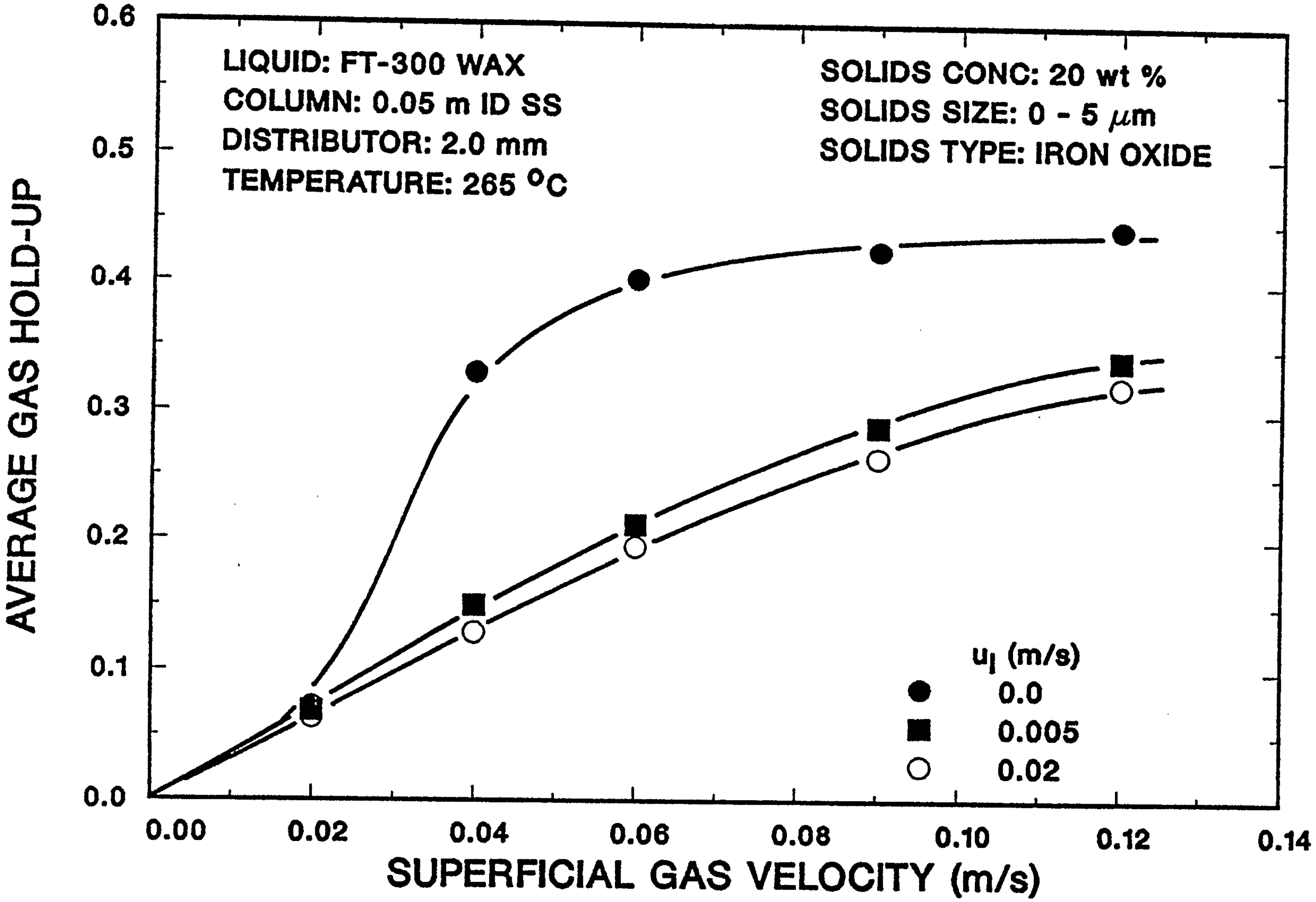


Figure 8. Effect of liquid velocity on average gas hold-up.

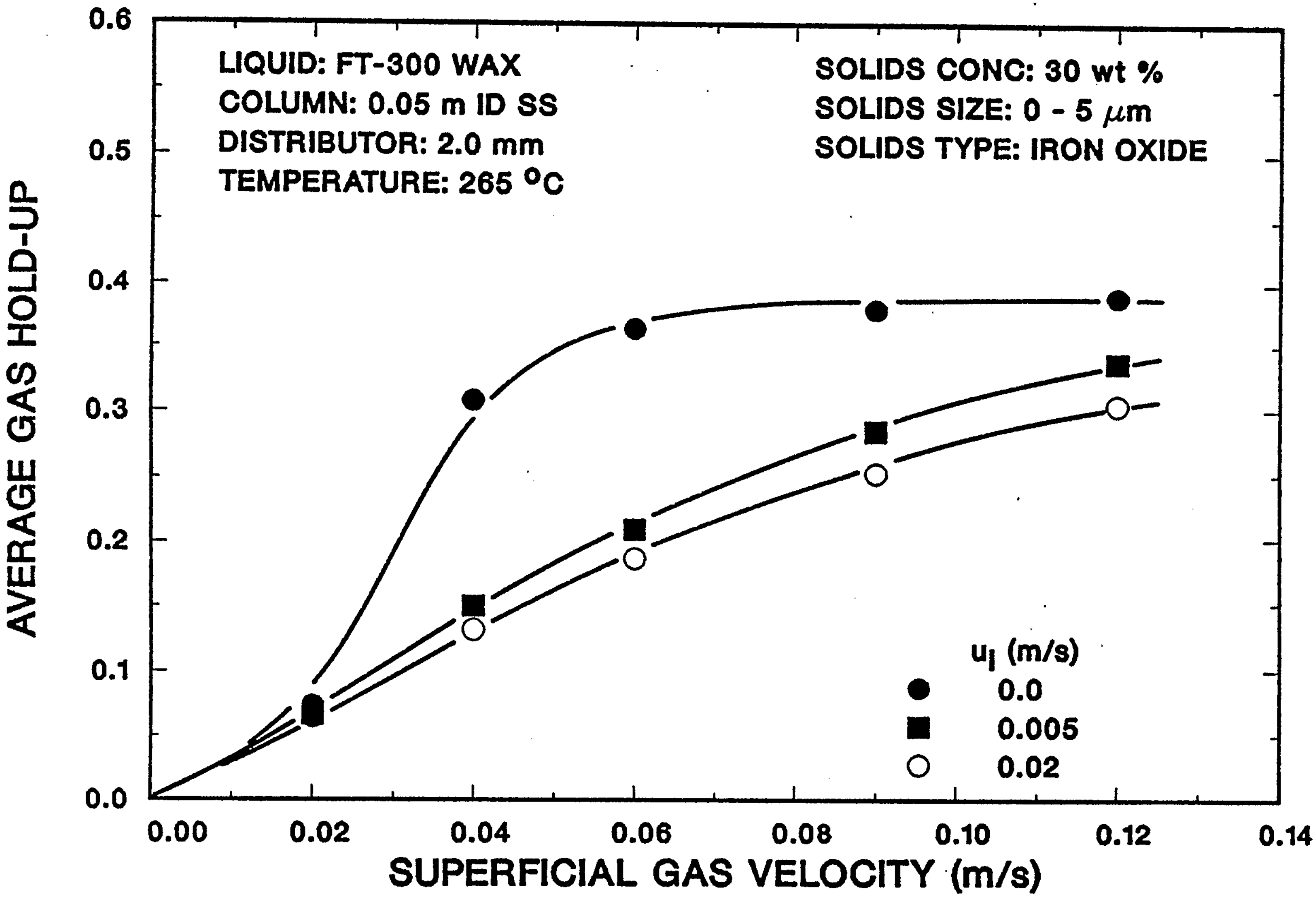


Figure 9. Effect of liquid velocity on average gas hold-up.

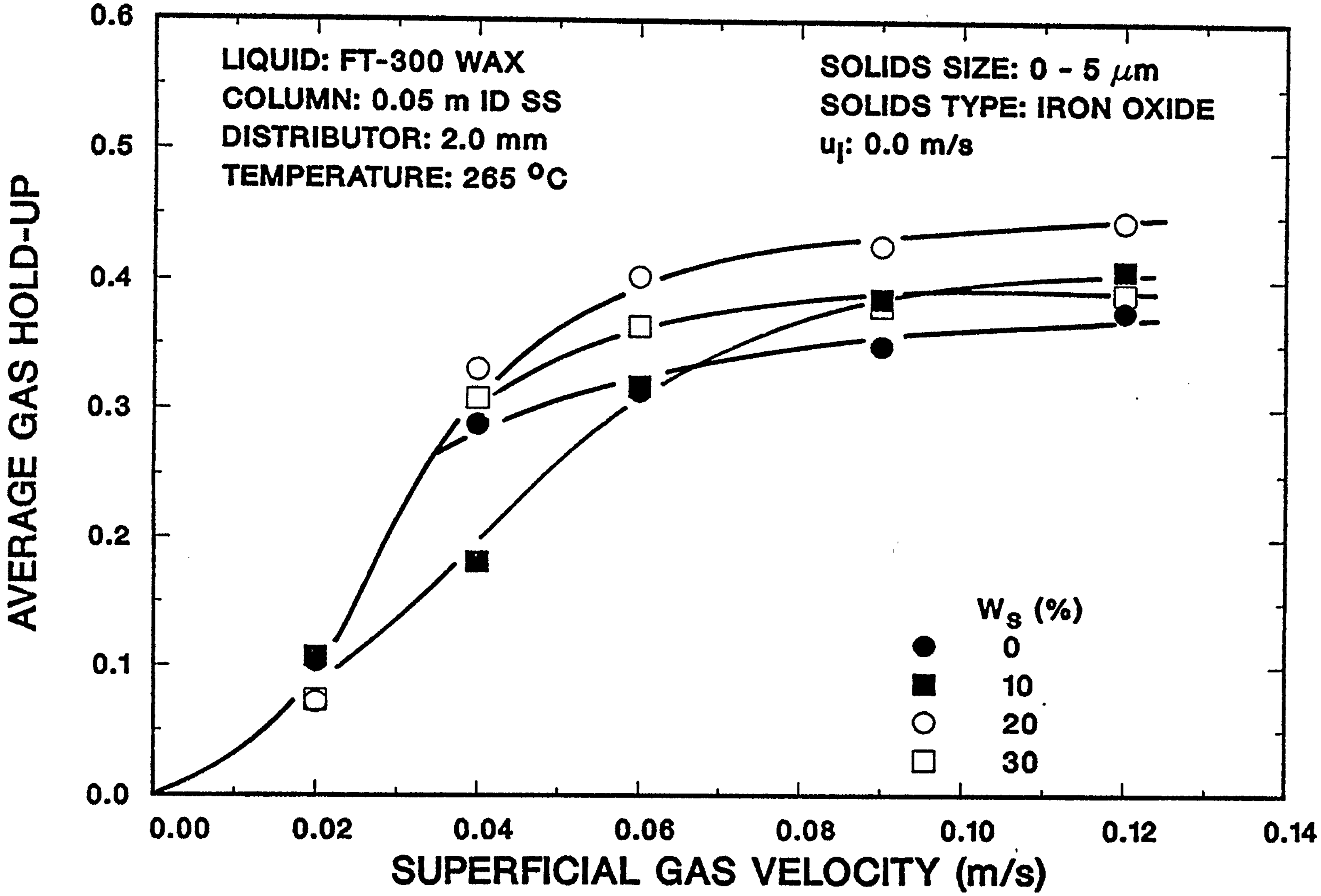


Figure 10. Effect of solids concentration on average gas hold-up.

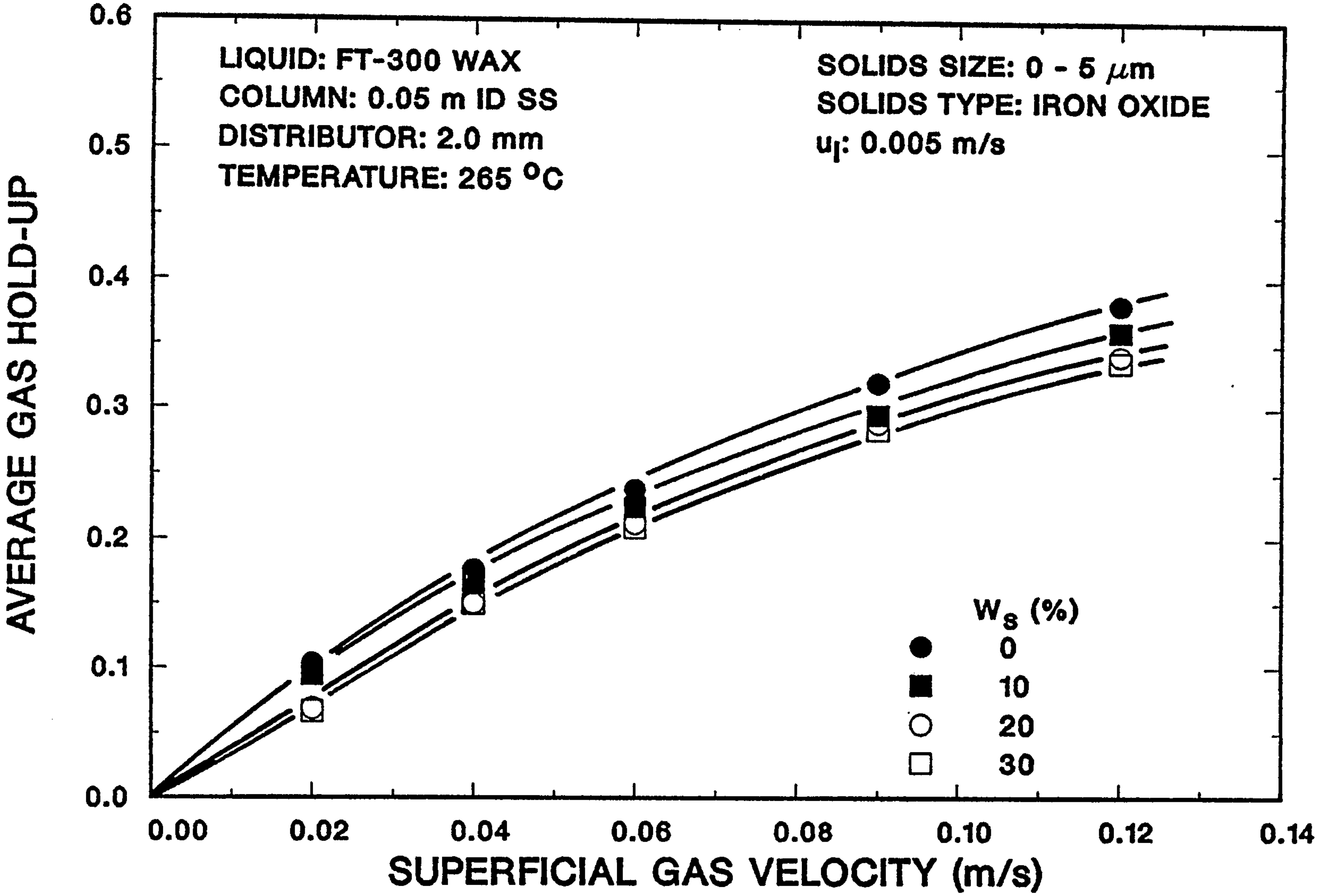


Figure 11. Effect of solids concentration on average gas hold-up.

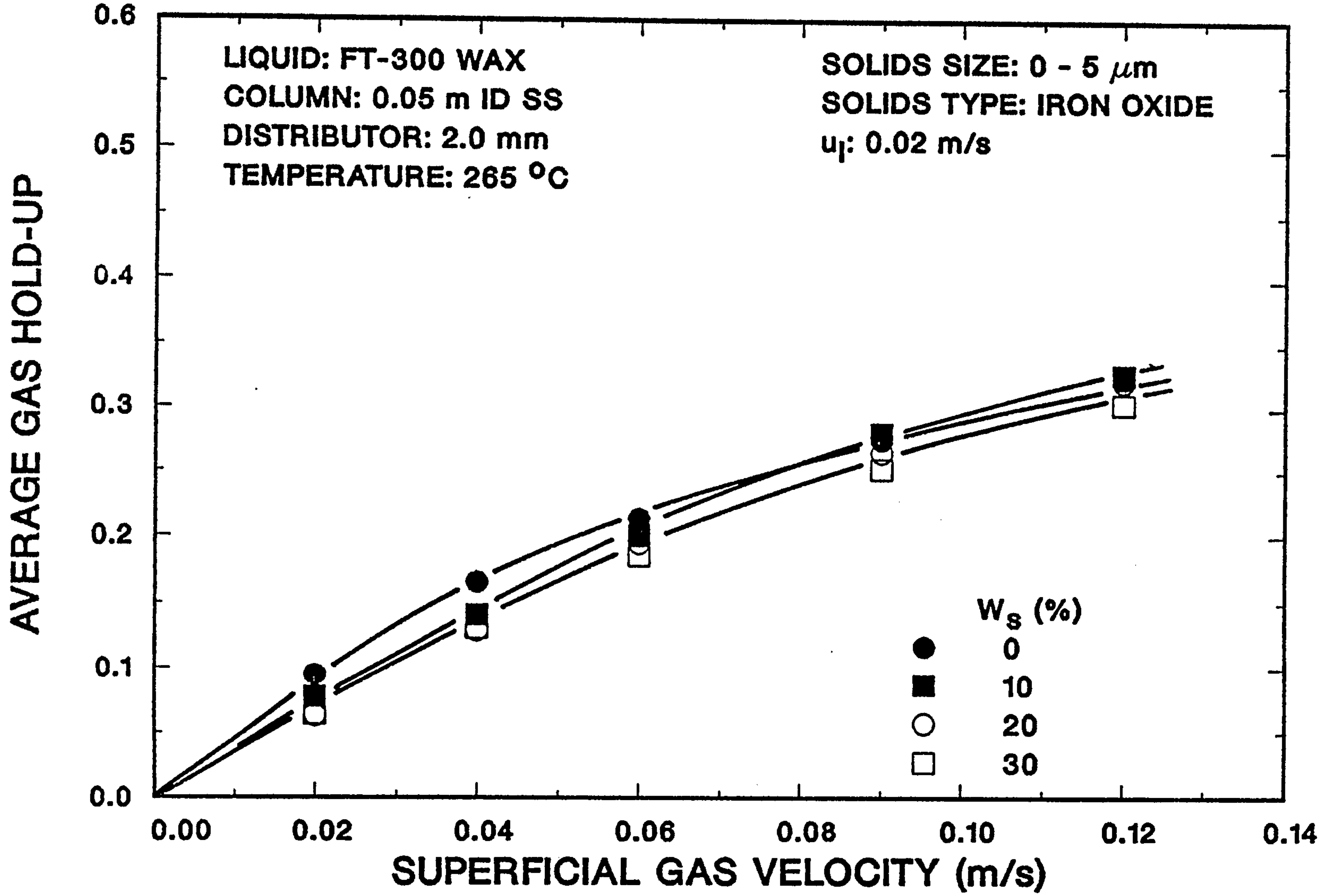


Figure 12. Effect of solids concentration on average gas hold-up.

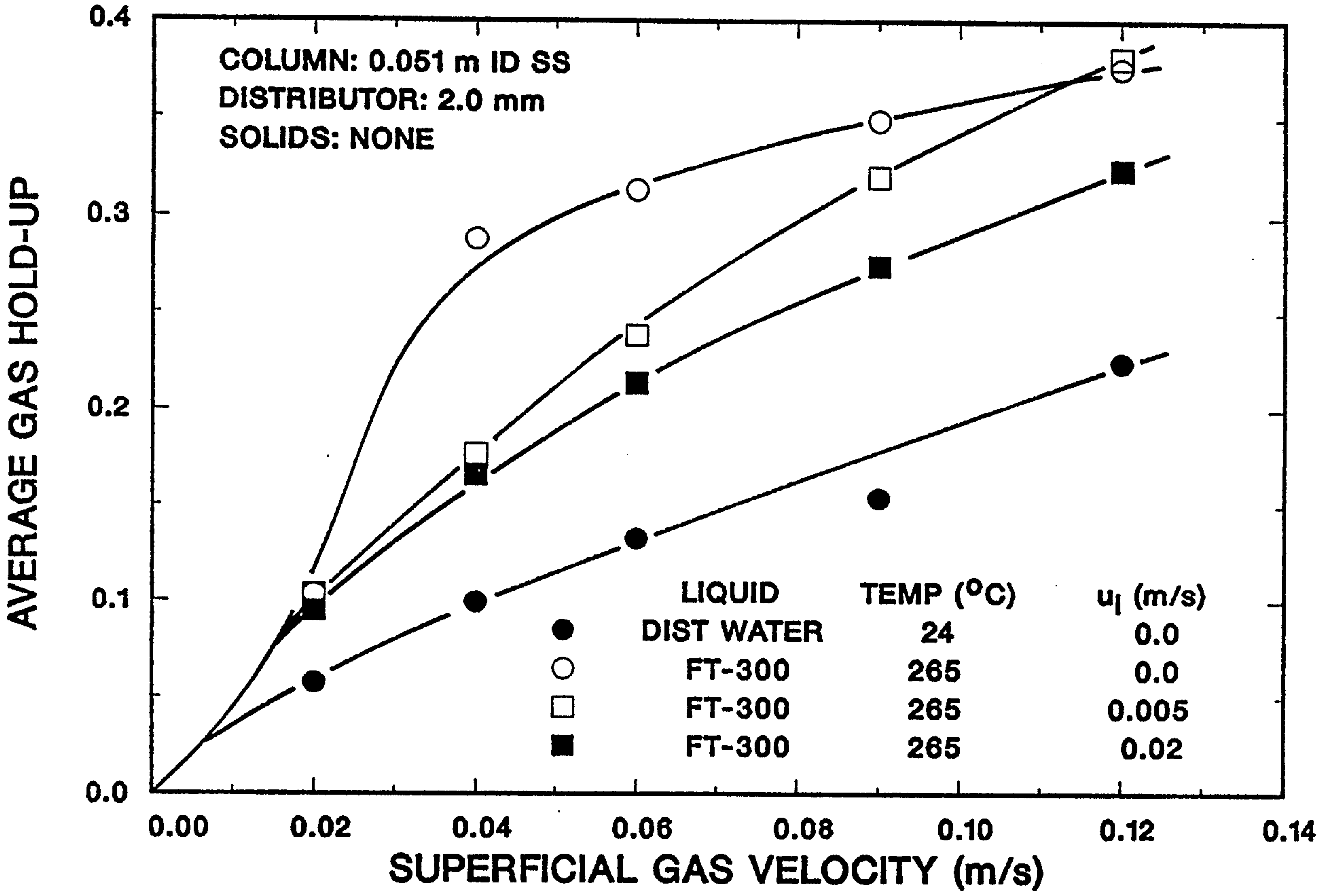


Figure 13. Effect of liquid medium and liquid velocity on average gas hold-up.

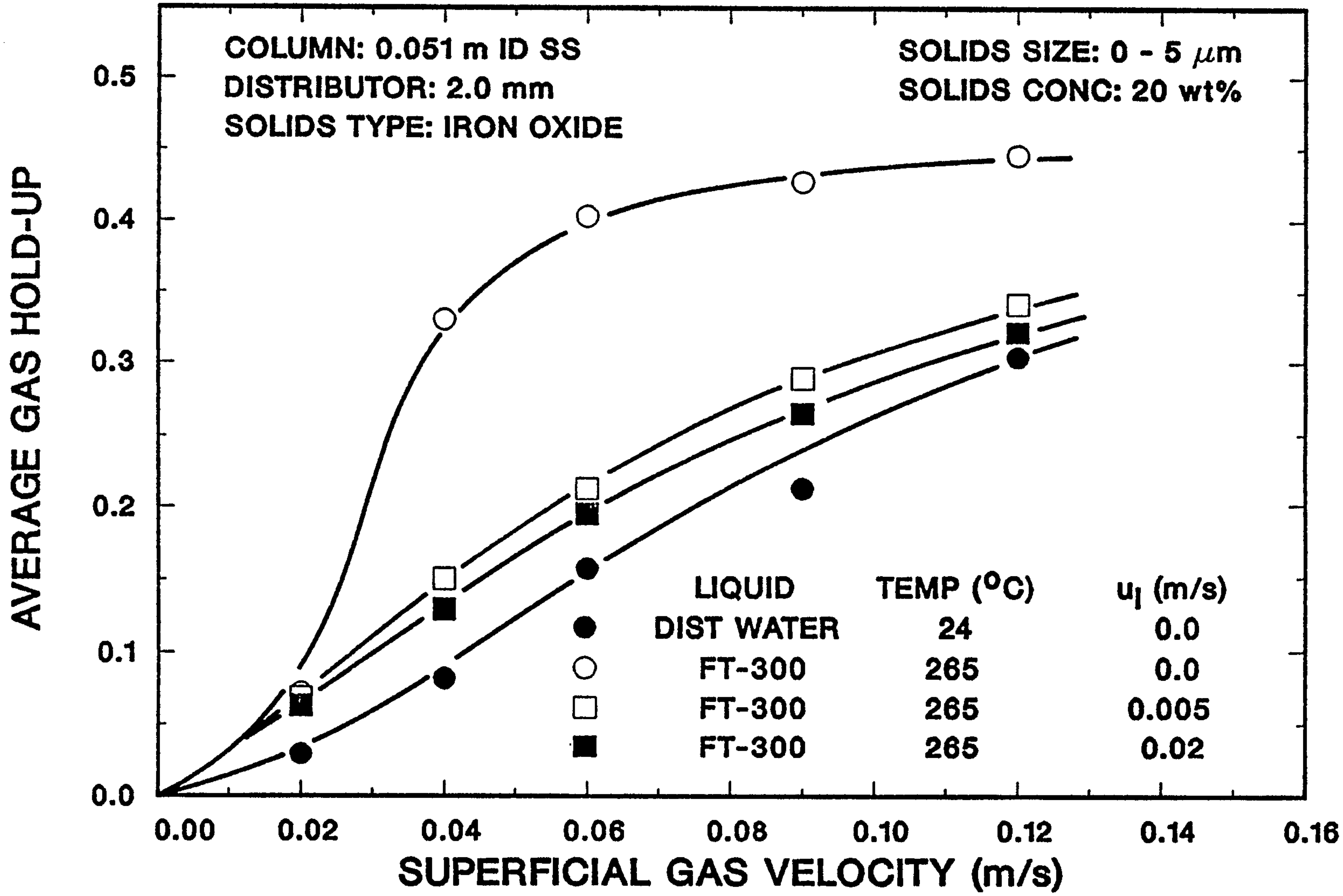


Figure 14. Effect of liquid medium and liquid velocity on average gas hold-up.

APPENDIX A – OPERATING AND DATA REDUCTION PROCEDURES

APPENDIX A

Operating Procedure

A brief outline of the procedure employed to operate the small stainless steel column for hot flow runs is outlined here. Figure A.1 shows the run numbering scheme used to identify the individual runs.

Prior to experiments with a given batch of wax, the mass flow meter and the pressure transducers are calibrated over the entire operating range. The mass flow meter is calibrated with a wet test meter hooked up in series with it. The pressure transducers are calibrated, while they are at their respective locations on the column wall, by filling up the column to different heights with distilled water and recording the transducer meter readings.

The wax granules are charged in the storage tank and brought up to temperature by heating the tank. Once the solid wax melts, the stirrer is switched on to improve the heating process. For runs conducted with solids, the necessary quantity of solid particles are added to the molten wax. The slurry is heated to a temperature of 220-240°C before it is transported to the column. Once the storage tank reaches the desired temperature, slurry samples are withdrawn and the slurry height in the tank measured. The column is heated up to the desired operating temperature (265°C) before the wax is introduced. For all experiments, batch and continuous, the wax is transported to the column using a slight overpressure of nitrogen in the storage tank. For the continuous mode of operation, the pump is switched on only after the column is at least half filled with wax. This is done to prevent any solids, settled in the storage tank, from clogging the pump. Throughout the preheating period and during transportation of wax to the column, a low flow rate of nitrogen is maintained through the distributor. Once the wax is in the column, the temperatures of the various units are allowed to stabilized before the actual run is started. For experiments conducted in the batch mode, only the column is maintained at the operating temperature using two temperature controllers, one for each half of the column (top and bottom). The exit lines and expansion unit are maintained at around 200°C. The scrubber, which is filled with a mineral spirit to recover components of wax that evaporate from the column, is maintained at 100°C. The gas inlet line and the distributor section are maintained between 220-240°C. The lines connected to the pressure transducers and the slurry withdrawal lines were maintained at 200°C. For continuous mode of operation, all lines and vessels carrying the slurry, including the column, expansion unit, overflow line, calibration chamber, storage tank, slurry feed line, and the slurry pump are maintained at the operating temperature. The remaining temperatures are the same as those used for the batch case. All temperatures are monitored regularly, every half hour during the preheating period (i.e. while wax is in the column and being heated up) and every hour once temperatures have stabilized.

All experiments are conducted using pre-purified nitrogen as the gas. Superficial gas velocities of 0.02, 0.04, 0.06, 0.09 and 0.12 m/s were employed in all runs. A duration of one and a half hours is used for each gas velocity. Pressure measurements are made three times for every gas velocity (i.e. every half hour), with the first measurement made half an hour after the gas velocity is changed. After the last pressure measurement, slurry samples are withdrawn from the column at five different locations. In addition to this, two samples are also withdrawn from the storage tank (top and bottom ports), and the height of slurry in the tank is measured. The gas flow rate is then changed to the next setting. For experiments conducted in the continuous mode of operation, the superficial liquid velocity is monitored using the calibration chamber. The desired liquid flow rate is set by varying the pump speed. Liquid flow rate checks are made three times at each gas velocity. Once the slurry samples are withdrawn into the sampling cylinders, they are allowed to cool and solidify before being removed. The cylinders are then cleaned with a solvent and reinstalled. The solids fractions of the slurry samples are determined using the procedure outlined previously.

Following the completion of a run, the slurry is withdrawn into the storage tank by using a slight vacuum in the tank (the pump is switched off for runs conducted in the continuous mode). Slurry samples are withdrawn from the storage tank and the slurry height measured once all of the slurry is back in the tank. If a new run is conducted with the same batch of wax the next day, all temperatures are lowered to around 200°C, nitrogen is allowed to flow through the distributor, and the traps in the DP cell lines are emptied to remove any accumulated slurry. If the run is the last one for the current batch of wax, the procedure is extended to include the cleaning of the system. The wax in the storage tank is drained into a pan, the drain below the distributor is opened to drain the distributor section and the region just above the distributor, and all temperatures are lowered to about 100°C. Once the system is at this temperature, solvent (toluene) is introduced into the column through a nozzle just above the distributor and the gas flow rate is increased to agitate the solvent. Sufficient amount of solvent is added so that some of it is carried into the expansion unit, overflow lines and the calibration chamber. The spent solvent is pulled into the slurry storage tank after approximately 45 minutes and drained. The process is repeated if deemed necessary. Following the cleaning process, the system is kept at 130-150°C overnight with nitrogen flowing through the distributor to evaporate any remaining solvent. The system is now ready for the next batch of experiments.

Data Reduction Procedure

The procedure used to estimate the various hydrodynamic parameters using measured quantities, for experiments conducted in the stainless steel columns, is outlined here. Figure A.2 is a schematic showing the pressure ports and sample ports on the small stainless steel column. Only the final forms of the pertinent equations, in terms of system constants and measured quantities, are presented here. The system constants used include the densities of the solids (ρ_s), liquid (ρ_ℓ), solidified wax (ρ_w) and acetone (ρ_{acet}), heights of the five pressure ports above the distributor (h_1 to h_5), and distance between the distributor and the top of the column (h_t). The measured quantities include the meter readings for the five pressure transducers (MR_1 to MR_5), the weights of solidified slurry samples ($m_{s\ell ij}$), the combined weight of the support structure and slurry sample (m_{1ij} - with the sample in air), and the combined weight of the support structure and slurry sample (m_{2ij} - with the sample immersed in acetone). For simplicity, the measured quantities have been replaced with the primary derived quantities, i.e. the differential pressure across the section i-j (ΔP_{ij}), and the average weight fraction of solids for the section i-j ($\langle W_s \rangle_{ij}$). The differential pressure is defined by

$$\Delta P_{ij} = P_i - P_j = (a_i MR_i + b_i) - (a_j MR_j + b_j) \qquad i = 1 \text{ to } 5, \;\; j = i+1 \qquad (2)$$

where a_i and a_j are the slopes of the calibration curves relating the meter readings to pressure (in equivalent height of water) for transducers at ports i and j, respectively, and b_i and b_j are the intercepts of the two curves. The average weight fraction of solids for the section between pressure ports i and j is given by

$$\langle W_s \rangle_{ij} = \frac{\rho_s - \left(\frac{\rho_w \rho_s}{\rho_{acet}}\right)\left(\frac{m_{1ij} - m_{2ij}}{m_{s\ell ij}}\right)}{\rho_s - \rho_w} \qquad i = 1 \text{ to } 5, \;\; j = i+1 \qquad (3)$$

The gas hold-up in the section i-j is given by

$$\langle \epsilon_g \rangle_{ij} = 1 - \left(\frac{\Delta P_{ij}}{\Delta h_{ij}}\right)\left[\frac{\langle W_s \rangle_{ij}}{\rho_s} + \frac{1 - \langle W_s \rangle_{ij}}{\rho_\ell}\right] \qquad i = 1 \text{ to } 5, \;\; j = i+1 \qquad (4)$$

where $\Delta h_{ij} = h_j - h_i$. The liquid hold-up in the section i-j is given by

$$\langle \epsilon_\ell \rangle_{ij} = \left(\frac{\Delta P_{ij}}{\Delta h_{ij}}\right)\left(\frac{1 - \langle W_s \rangle_{ij}}{\rho_\ell}\right) \qquad i = 1 \text{ to } 5, \;\; j = i+1 \qquad (5)$$

and the solids hold-up in this section is given by

$$\langle \epsilon_s \rangle_{ij} = \left(\frac{\Delta P_{ij}}{\Delta h_{ij}} \right) \left(\frac{\langle W_s \rangle_{ij}}{\rho_s} \right) \qquad i = 1 \text{ to } 5,\ \ j = i + 1 \tag{6}$$

The above equations can be used to estimate hold-ups of the three phases in the section between any two adjacent pressure ports, however, for the equations to be valid the entire section must be filled with the dispersion. For runs conducted in the continuous mode of operation, all five sections are always full, since the dispersion fills the entire column. For runs conducted in the batch mode of operation, eqns (4), (5) and (6) can be used for those sections that are full, i.e. all sections below the top most non-zero pressure port. The section just above this pressure port (say section n) is only partially full, therefore the height of the dispersion in this section (Δh_n) is not known. However, the differential pressure for this section (ΔP_n) is known. If a slurry sample was not withdrawn from this section, $\langle W_s \rangle_n$ would also be an unknown quantity. For such cases, the gas hold-up and if necessary, the weight fraction of solids in this section are estimated by extrapolation of the $\langle \epsilon_g \rangle$ and $\langle W_s \rangle$ profiles. The height of the dispersion in this section is then calculated using

$$\Delta h_n = \left(\frac{\Delta P_n}{1 - \langle \epsilon_g \rangle_n} \right) \left[\frac{\langle W_s \rangle_n}{\rho_s} + \frac{1 - \langle W_s \rangle_n}{\rho_\ell} \right] \tag{7}$$

Eqns (5) and (6) can then be used to estimate the liquid and solids hold-ups for this section. For all runs, no measurements are made for the 3" section (section 0-1) between the distributor and pressure port 1 (see Figure A.2). It is assumed that the volume fractions for the three phases in this section are the same as those in the section above (i.e. section 1-2).

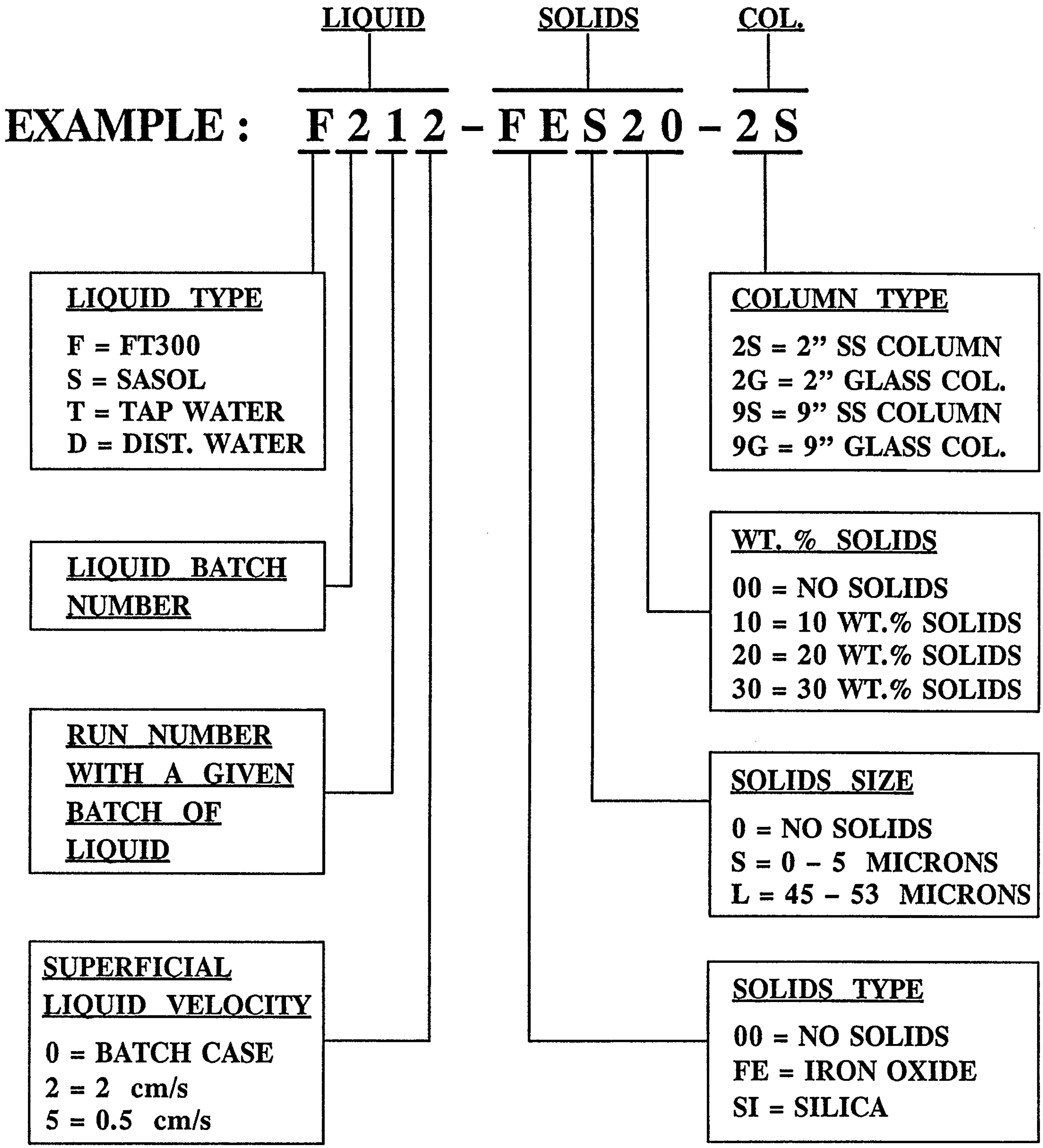
The average gas hold-up for the entire dispersion can be obtained using a weighted average of the gas hold-ups in the individual sections and is given as

$$\langle \epsilon_g \rangle = 1 - \frac{\sum_{i=0}^{n} \left(\Delta P_{ij}\right) \left[\frac{\langle W_s \rangle_{ij}}{\rho_s} + \frac{1 - \langle W_s \rangle_{ij}}{\rho_\ell} \right]}{\sum_{i=0}^{n} \Delta h_{ij}} \qquad j = i + 1 \qquad (8)$$

where $\Delta P_{01} = \Delta P_{12} \Delta h_{01} / \Delta h_{12}$, and $\langle W_s \rangle_{01} = \langle W_s \rangle_{12}$. n=5 for the continuous mode of operation, wheareas, it is dependent of the location of the top of the dispersion for the batch mode of operation.

FIGURE A.1

RUN NUMBERING SCHEME



LIQUID TYPE

F = FT300
S = SASOL
T = TAP WATER
D = DIST. WATER

LIQUID BATCH NUMBER

RUN NUMBER WITH A GIVEN BATCH OF LIQUID

SUPERFICIAL LIQUID VELOCITY

0 = BATCH CASE
2 = 2 cm/s
5 = 0.5 cm/s

COLUMN TYPE

2S = 2" SS COLUMN
2G = 2" GLASS COL.
9S = 9" SS COLUMN
9G = 9" GLASS COL.

WT. % SOLIDS

00 = NO SOLIDS
10 = 10 WT.% SOLIDS
20 = 20 WT.% SOLIDS
30 = 30 WT.% SOLIDS

SOLIDS SIZE

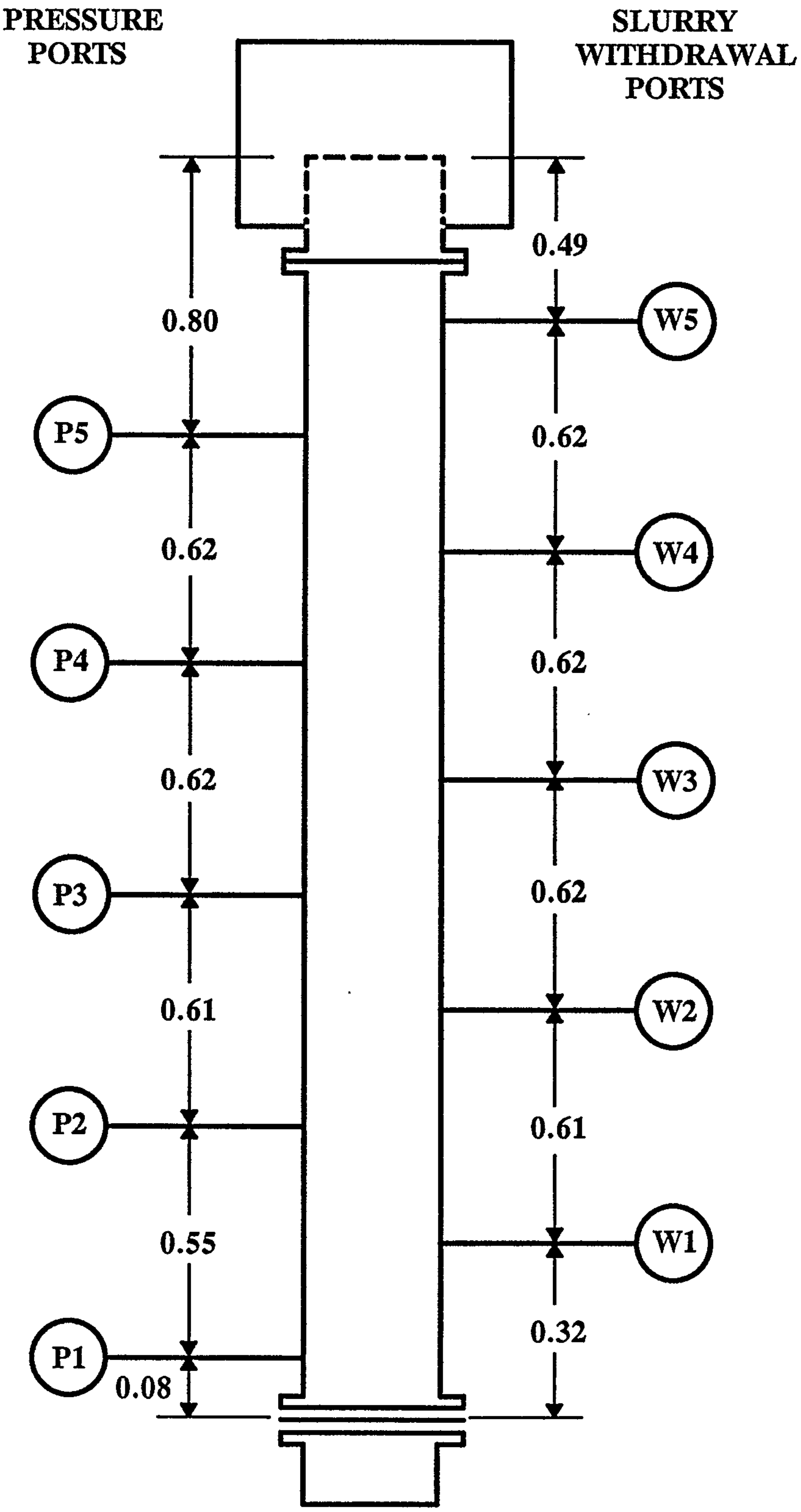
0 = NO SOLIDS
S = 0 – 5 MICRONS
L = 45 – 53 MICRONS

SOLIDS TYPE

00 = NO SOLIDS
FE = IRON OXIDE
SI = SILICA

Figure A.2. Schematic of 0.051 m ID SS column

(all dimensions are in meters)



APPENDIX B – AXIAL GAS HOLD–UP AND SOLIDS DISTRIBUTION PROFILES

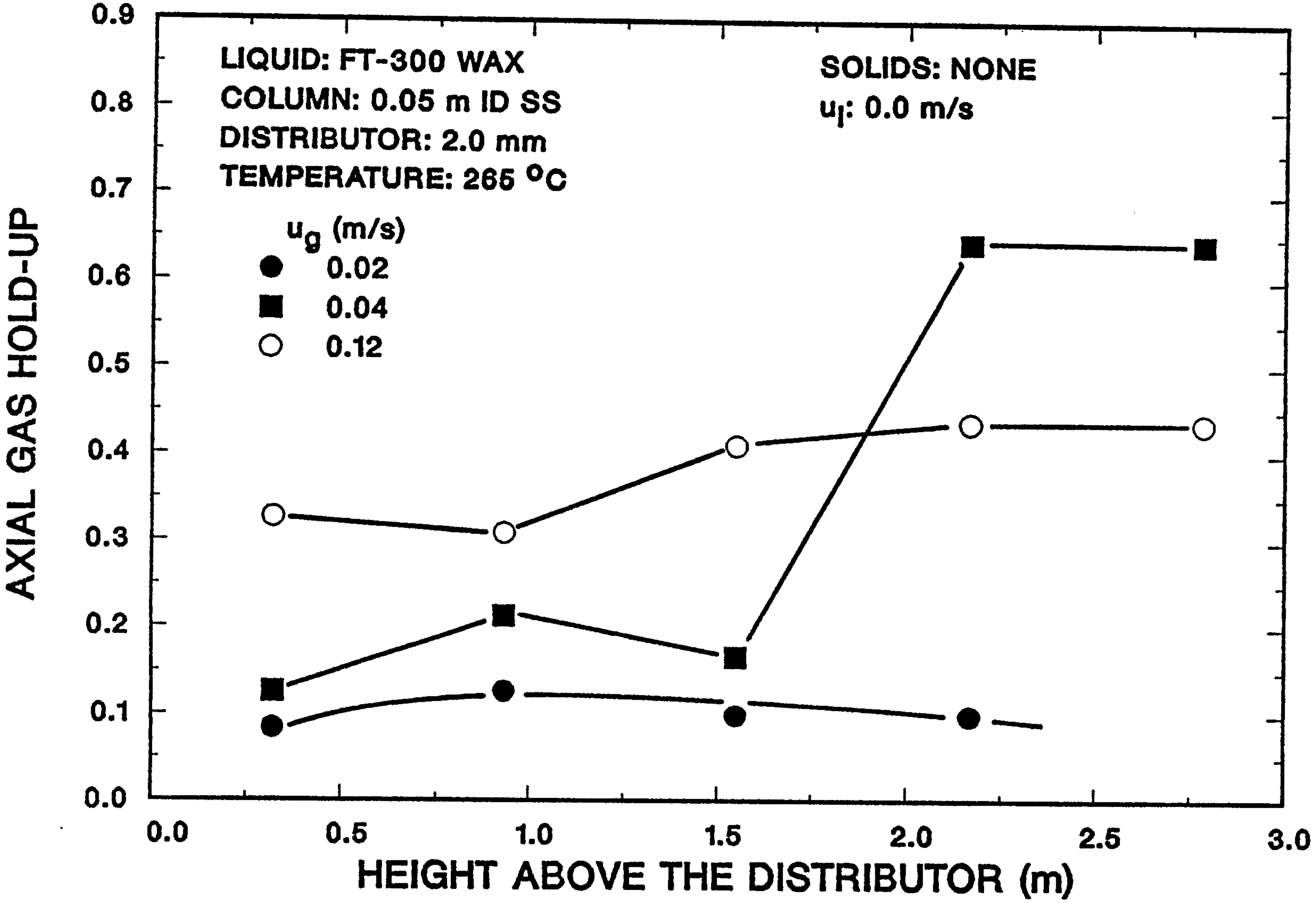


Figure B-1. Effect of superficial gas velocity on axial gas hold-up.

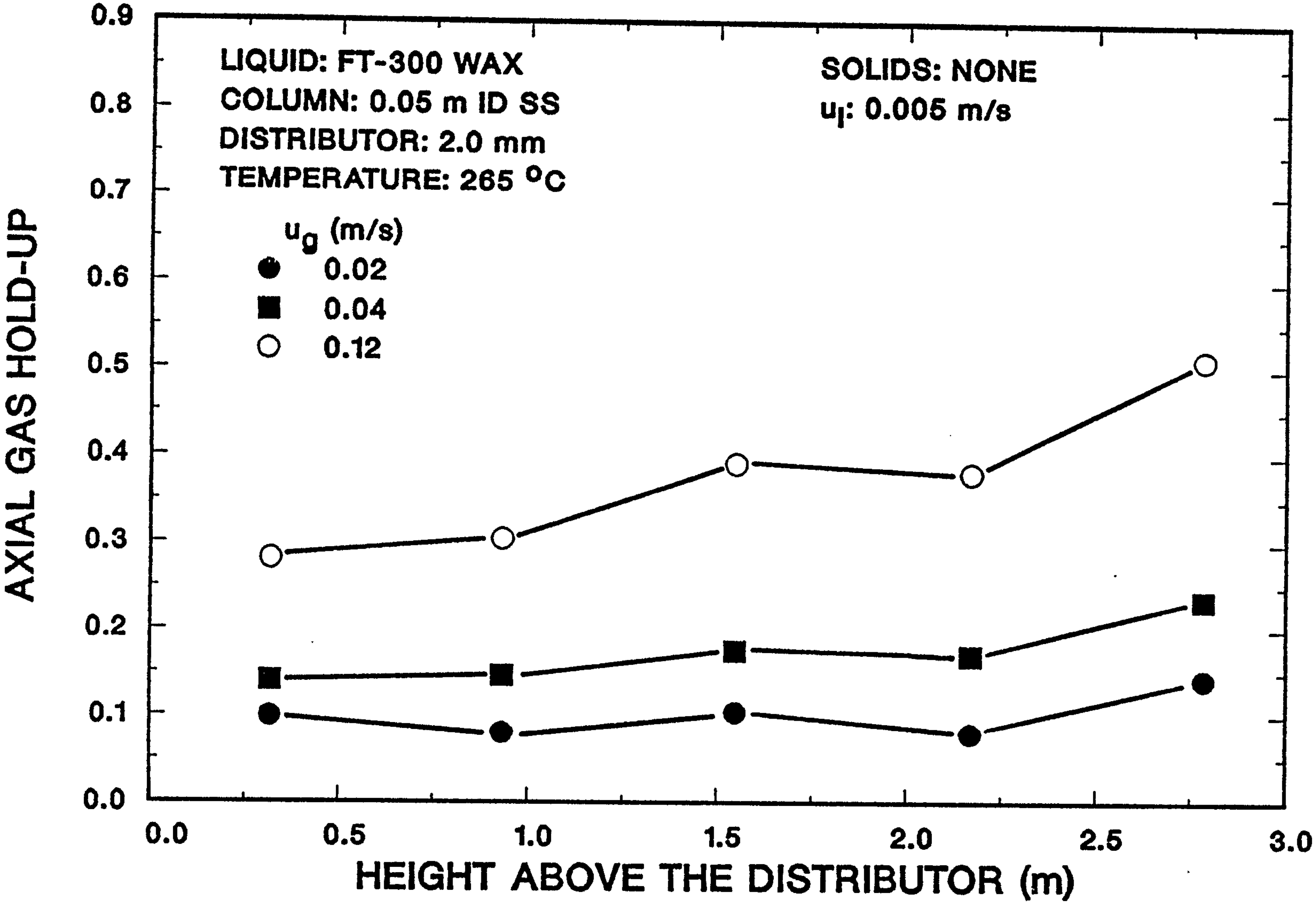


Figure B-2. Effect of superficial gas velocity on axial gas hold-up.

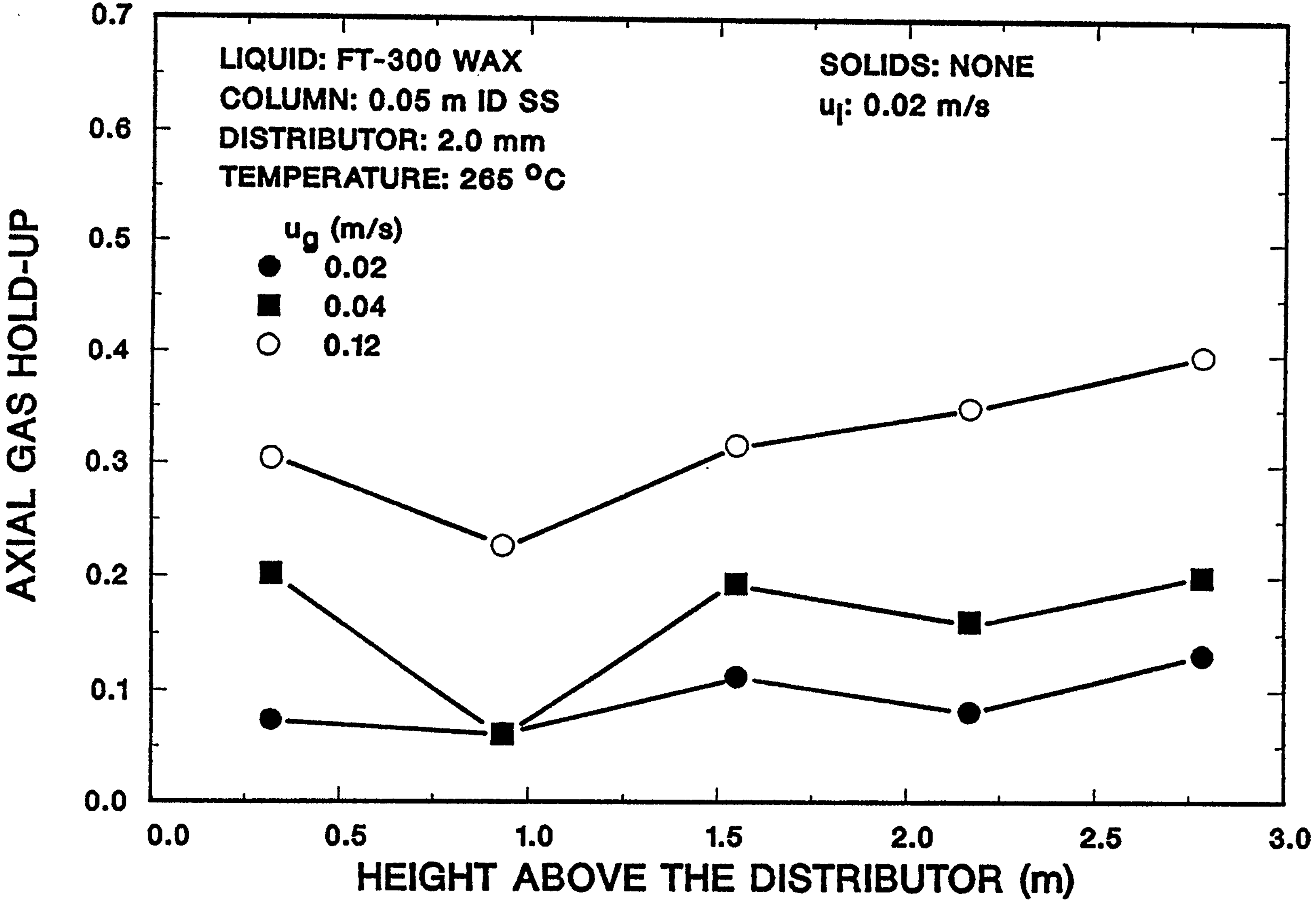


Figure B-3. Effect of superficial gas velocity on axial gas hold-up.

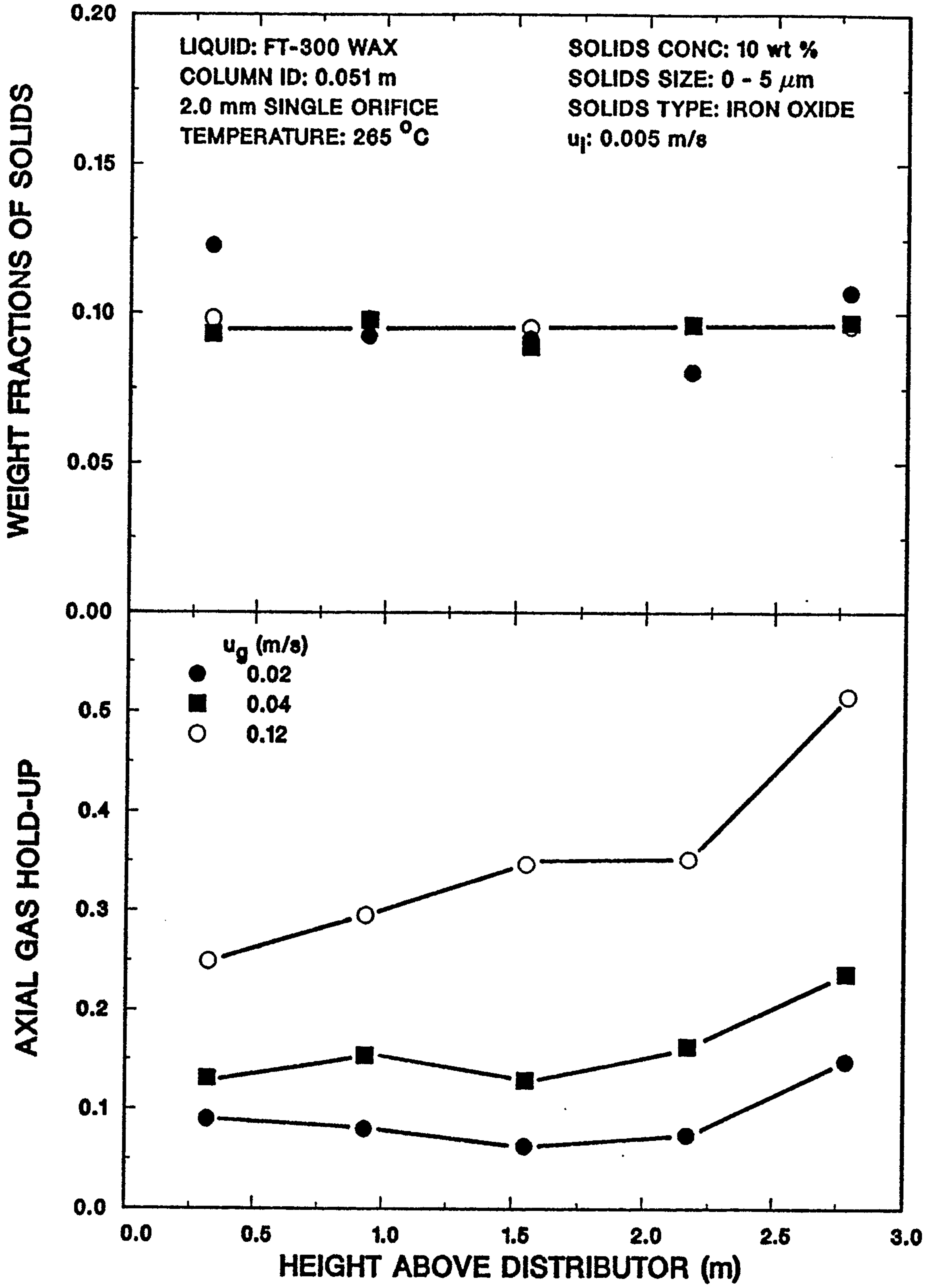


Figure B-4. Effect of superficial gas velocity on axial solids distribution (Top) and on axial gas hold-up (Bottom).

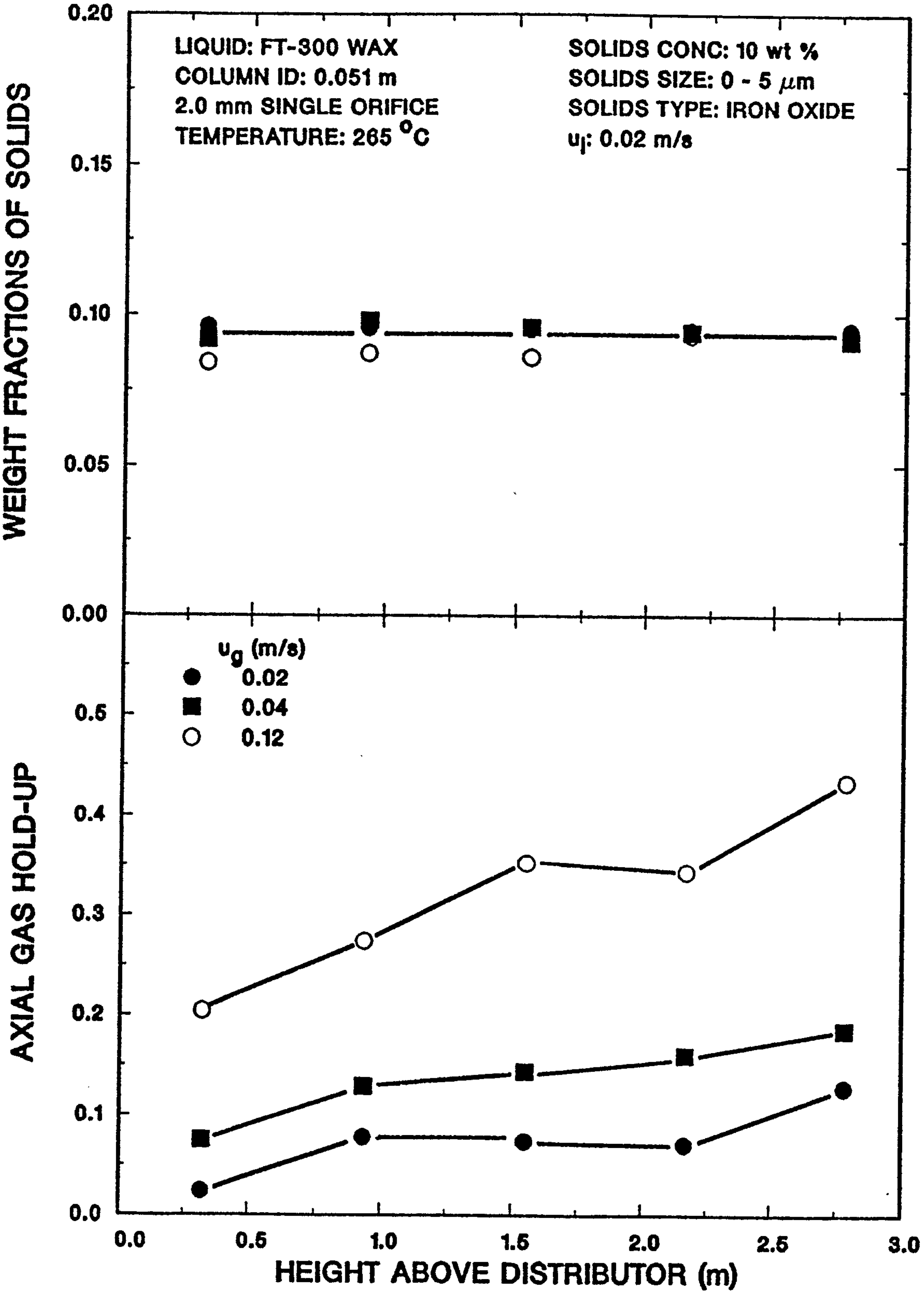


Figure B-5. Effect of superficial gas velocity on axial solids distribution (Top) and on axial gas hold-up (Bottom).

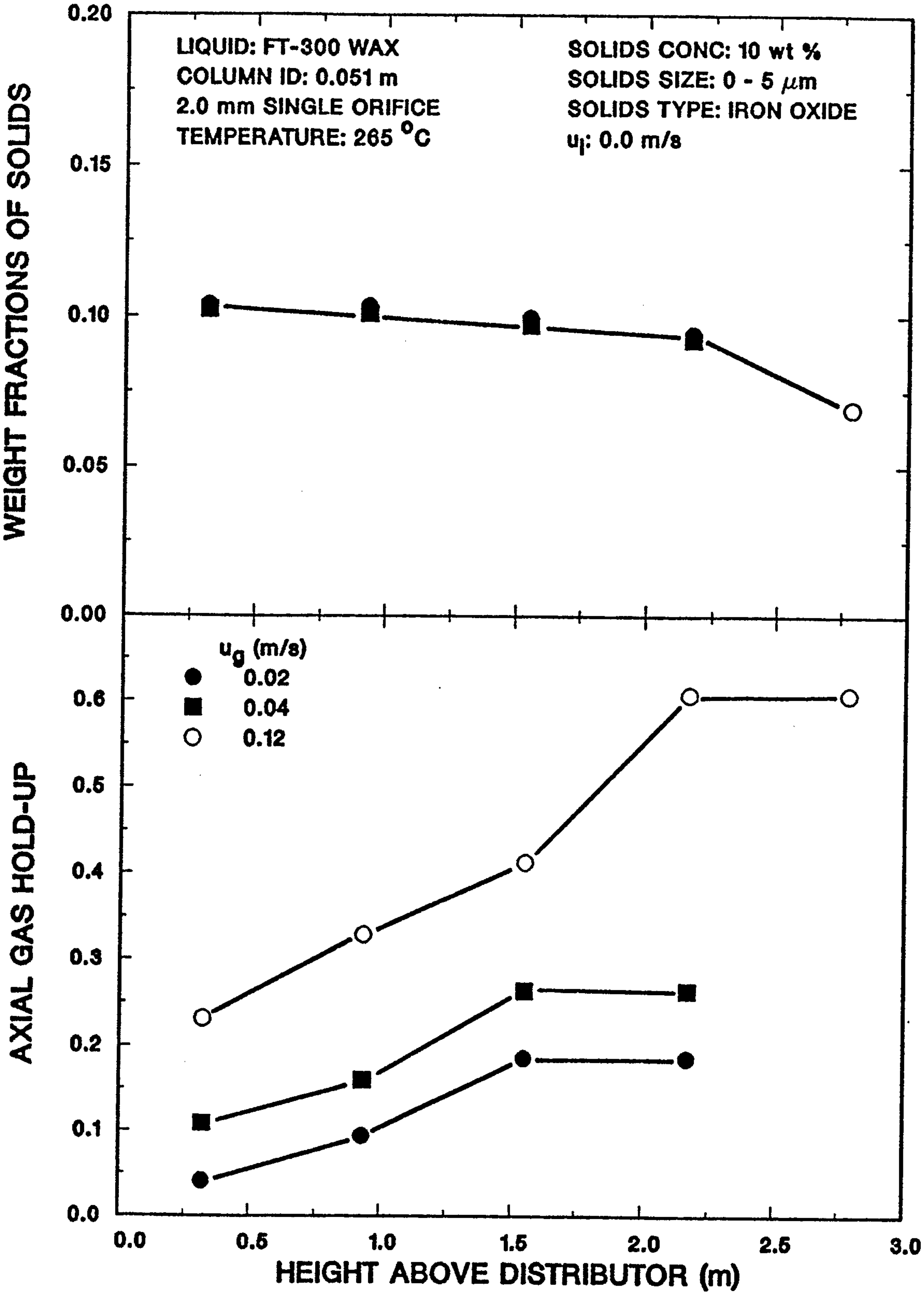


Figure B-6. Effect of superficial gas velocity on axial solids distribution (Top) and on axial gas hold-up (Bottom).

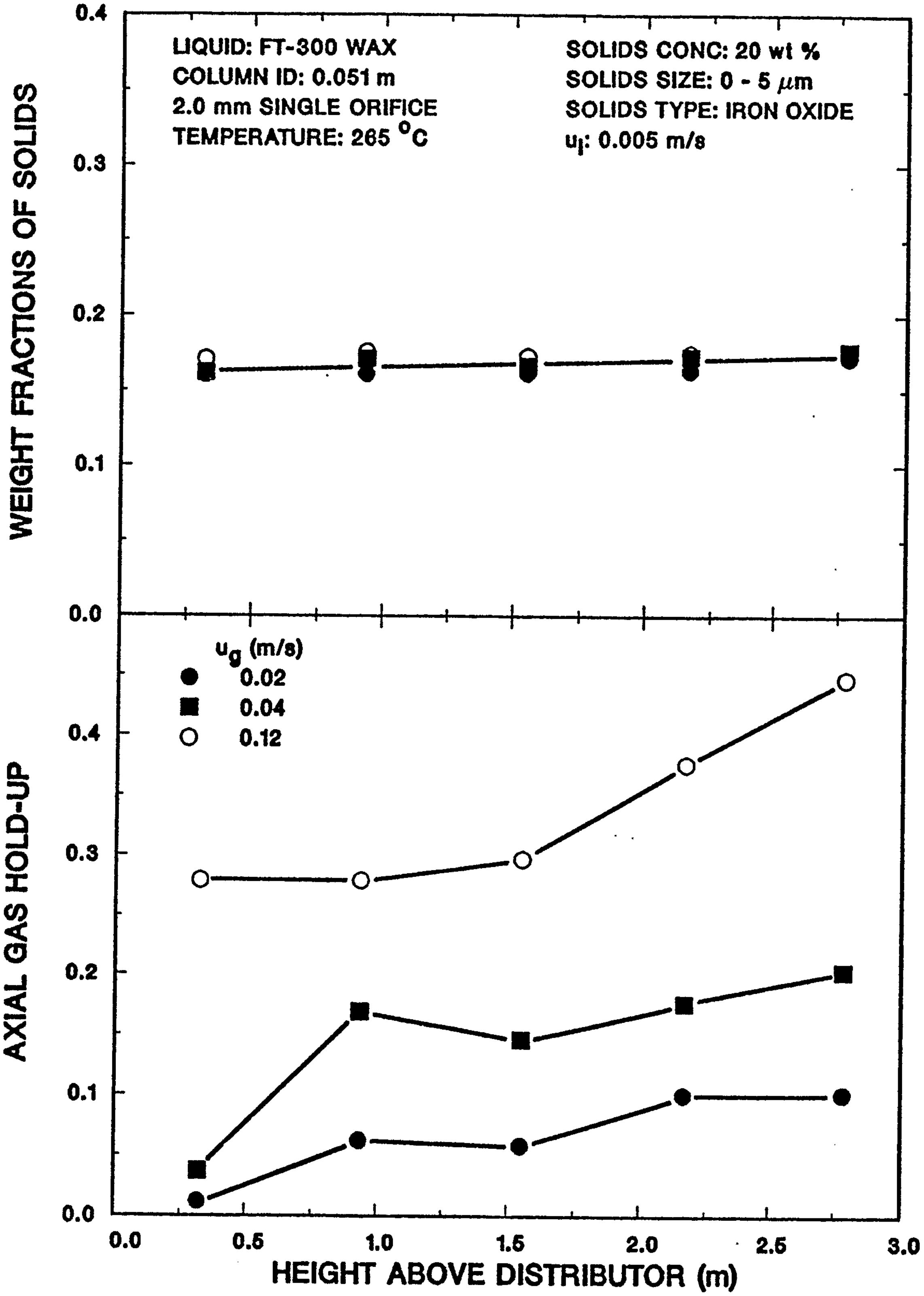


Figure B-7. Effect of superficial gas velocity on axial solids distribution (Top) and on axial gas hold-up (Bottom).

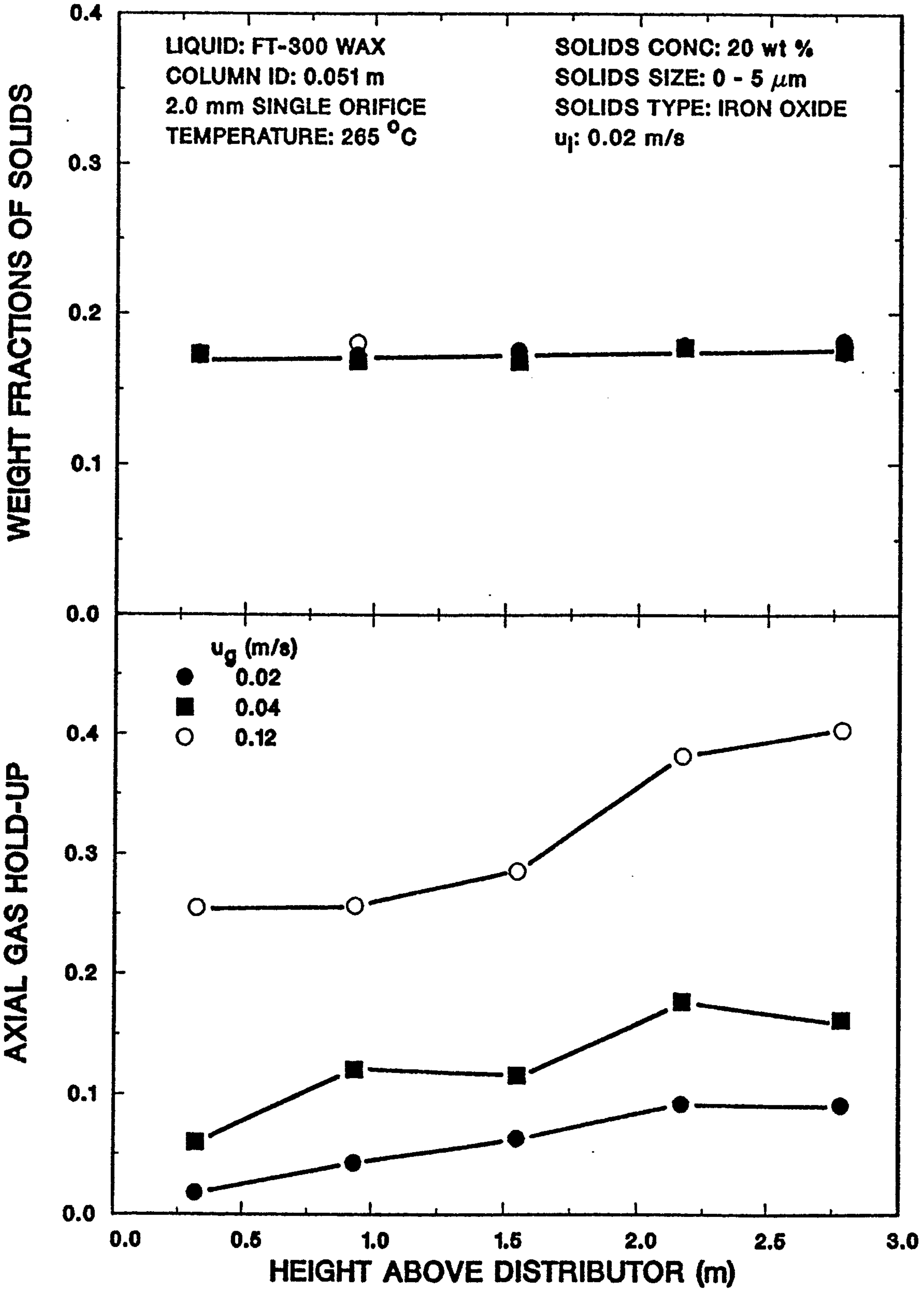


Figure B-8. Effect of superficial gas velocity on axial solids distribution (Top) and on axial gas hold-up (Bottom).

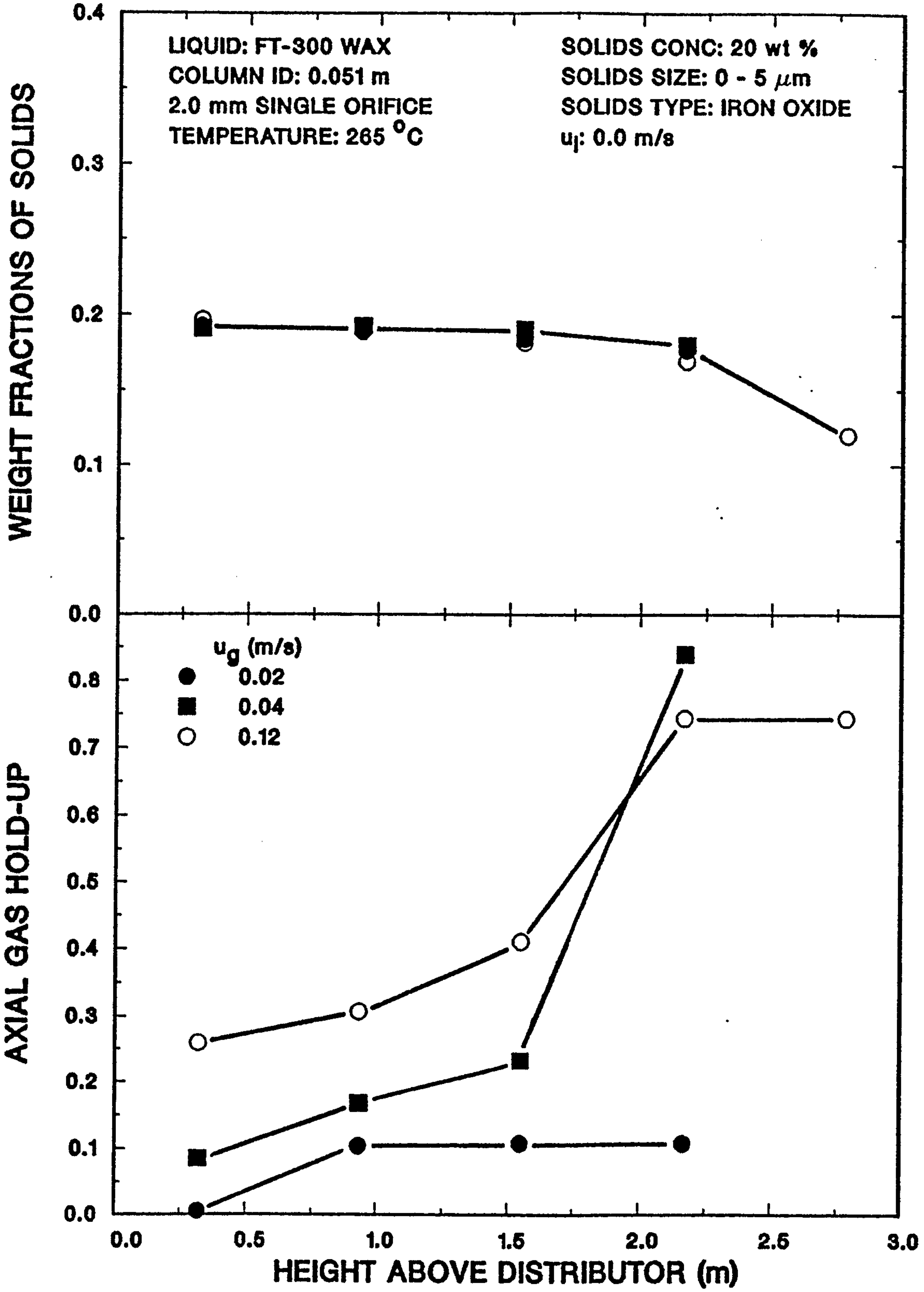


Figure B-9. Effect of superficial gas velocity on axial solids distribution (Top) and on axial gas hold-up (Bottom).

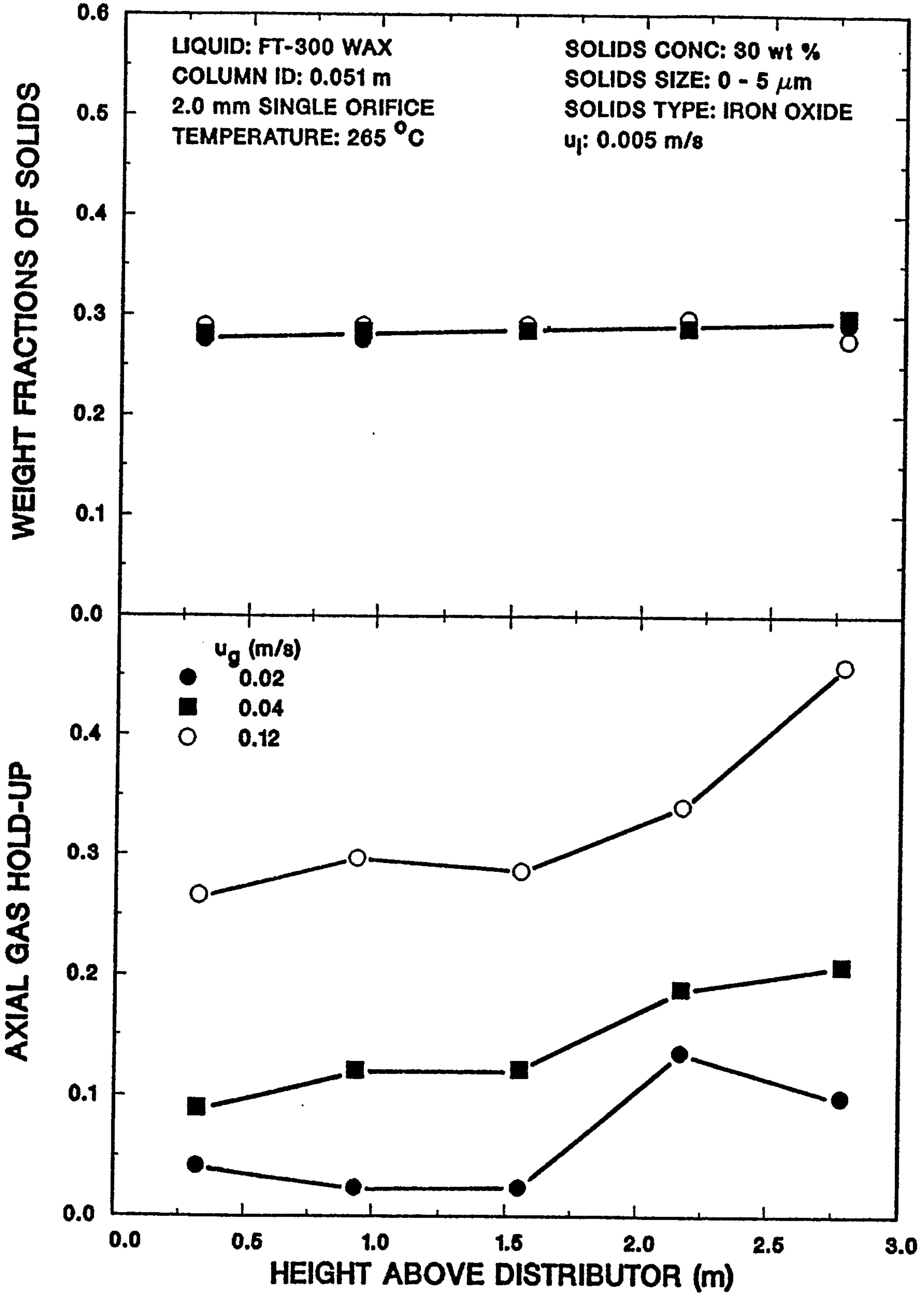


Figure B-10. Effect of superficial gas velocity on axial solids distribution (Top) and on axial gas hold-up (Bottom).

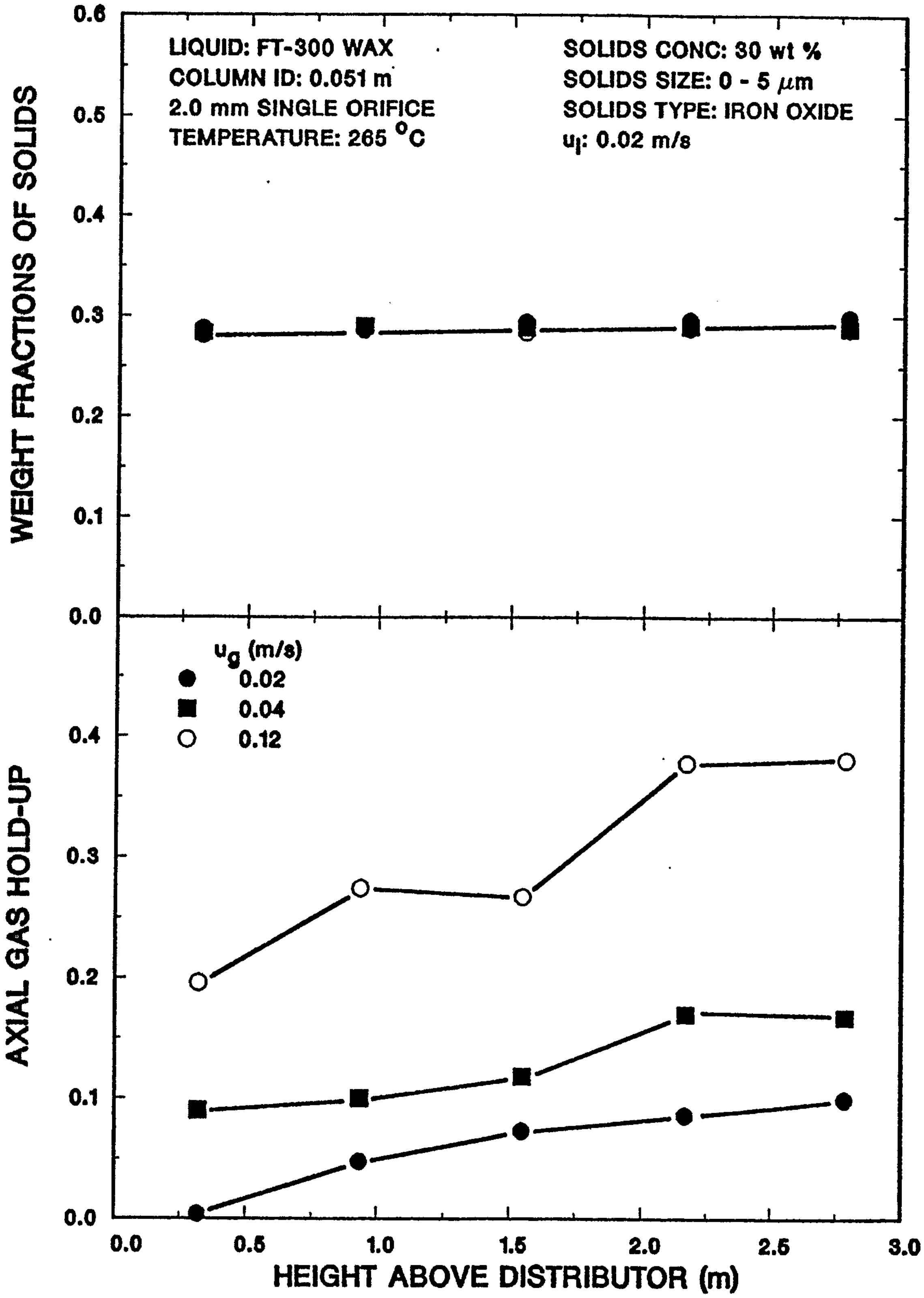


Figure B-11. Effect of superficial gas velocity on axial solids distribution (Top) and on axial gas hold-up (Bottom.)

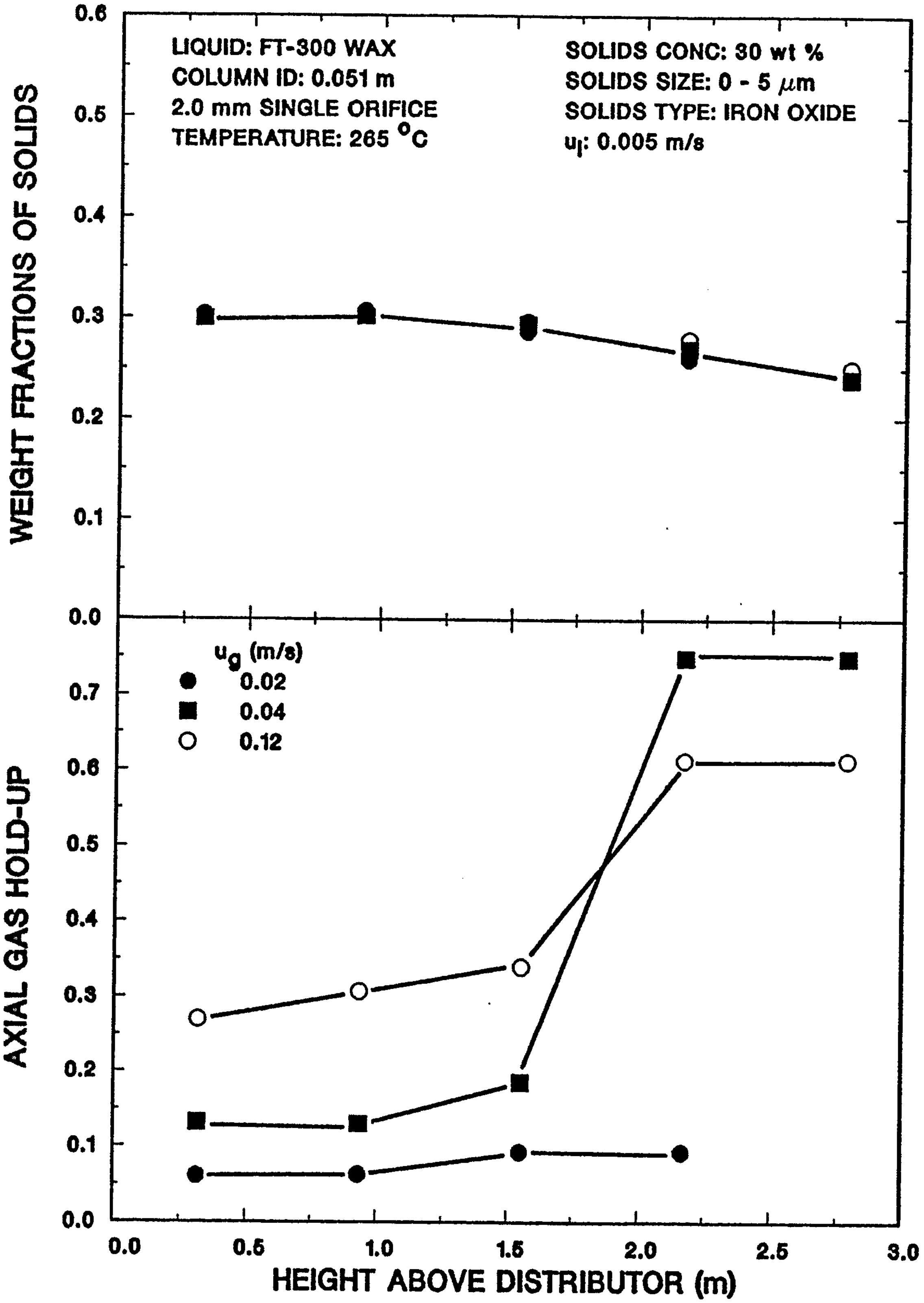


Figure B-12. Effect of superficial gas velocity on axial solids distribution (Top) and on axial gas hold-up (Bottom.)

DOE/PC/90012-3

Hydrodynamics of Three-Phase Slurry Fischer-Tropsch Bubble Column Reactors

Quarterly Technical Progress Report

for the Period 1 January 1988 - 31 March 1988

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April 30, 1988

Prepared for the Pittsburgh Energy Technology Center,
the United States Department of Energy Under Contract No. DE-AC22-86PC90012
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NOTICE

PATENT STATUS

TECHNICAL STATUS

TABLE OF CONTENTS

I. Abstract

During the past quarter, the remaining work on the small (2 inch diameter) stainless steel and glass columns was completed and the system made operational for both cold and hot flow studies in batch (no liquid flow) and continuous modes of operation. We also completed the installation and calibration of the process instrumentation. A sampling system for collecting slurry samples, and filtration units for determining solids concentration in these samples, were constructed and tested.

Work on Task 3 (Measurement of Hydrodynamic Properties by Conventional Techniques) was initiated during this quarter. We have conducted a total of 21 trial runs in the two small columns. These served to shakedown the system as well as to provide some preliminary information on the behavior of the nitrogen-water system with and without solids, using both batch and continuous modes of operation. Majority of the runs were conducted under ambient conditions using distilled water or tap water as the liquid medium. Three runs were conducted with FT-300 wax as the liquid medium at 265°C in the absence of solids. Preliminary analysis of the data from these runs indicate that the foaming tendency of FT-300 wax is reduced in the presence of liquid circulation. Experiments with water as the liquid medium suggest that solids concentration does not appear to affect the average gas hold-up significantly, however solids type appears to have an effect on hold-up, with higher values obtained with iron oxide. This is because iron oxide tends to foam when mixed with water. Our results also indicate that in the batch runs dispersion of solids is poor when iron oxide is used, compared to the dispersion with silica. Experiments with liquid circulation suggest that a small change in the liquid velocity has a stronger effect on solids dispersion than does a large change in the gas velocity.

The trial runs with solids revealed problems due to the settling of solids in slurry lines and in the pump. As a result, we have decided to modify the slurry feed system

and relocate the pump to minimize settling. The differential pressure (DP) system used in the small stainless steel column also had some problems. An effort to improve this system is under way.

During the past quarter we have placed orders for parts and material needed to construct the movable assembly mechanism for the nuclear density gauges. Some of these parts have already been received. We have subsequently requested quotes for machining of some of the parts.

II. Objective and Scope of Work

The overall objective of this contract is to determine the effects of bubble column diameter, solids loading and particle size, and operating conditions (temperature, gas and liquid flow rates) on hydrodynamics of slurry bubble columns for Fischer–Tropsch synthesis, using a molten wax as the liquid medium. To accomplish these objectives, the following specific tasks will be undertaken.

Task 1 – Project Work Plan

The objective of this task is to establish a detailed project work plan covering the entire period of performance of the contract, including a detailed program schedule, analytical procedures, and estimated costs and manhours expended by month for each task.

Task 2 – Design and Construction of the Experimental Apparatus

The existing glass and stainless steel columns (0.051 m and 0.229 m in diameter, 3 m tall) that were constructed under our previous DOE contract (DE–AC22–84PC70027), will be modified and additions made in order to study the effect of continuous upward liquid flow. After the procurement of equipment and instrumentation, and construction of the unit is completed, a shakedown of test facilities will be made to verify achievement of planned operating conditions.

Task 3 – Measurement of Hydrodynamic Parameters by Conventional Techniques

In this task, the effects of operating conditions (liquid and gas superficial velocities, temperature), gas distributor, column diameter, and solids concentrations and particle size on hydrodynamic parameters in the glass and stainless steel columns will be determined. All experiments will be conducted using nitrogen at atmospheric pressure.

The hydrodynamic parameters that will be determined as a function of the independent variables mentioned above are: average gas hold–up, axial solids distribution, axial gas hold–up, flow regime characterization, and qualitative information on bubble size distribution.

Task 4 – Application of a Gamma Radiation Density Gauge for Determining Hydrodynamic Parameters

The objective of this task is to determine hydrodynamic parameters for the three–phase system using a nuclear density gauge apparatus. A movable assembly mechanism and positioning racks for the two nuclear density gauges and detectors will be designed and constructed. Following the interfacing of the apparatus with an on–line microprocessor, the gauges will be calibrated using pure components (liquid wax and solid particles), and with known proportions of liquid and solid. After calibration, the following parameters will be obtained from experiments in the large stainless steel column: axial gas hold–up, axial concentration of solids, and qualitative information on flow regimes and bubble size distributions.

III. Summary of Progress

During the past quarter, all piping for the small (2 inch diameter) bubble columns was completed and the system made operational for both batch (no liquid flow) and continuous modes of operation. All vessels and pipelines, and the two small bubble columns were heat traced and insulated. We also installed and calibrated all related process instrumentation. The differential pressure (DP) system using nitrogen purge was installed in the small stainless steel column and calibrated. We have not received the two high temperature pressure transducers from Sensotron, therefore, five low temperature transducers (Validyne) were used to obtain differential pressure measurements. The slurry sampling system, which will be used to collect samples of the slurry at various heights along the small stainless column, was installed and tested. The slurry filtration system using paper filters did not prove to be efficient, therefore, we designed and successfully tested a filtration unit which used a 2 μm sintered metal plate. Five such units have been fabricated and are now operational.

Work on Task 3, Measurement of Hydrodynamic Parameters by Conventional Techniques, was initiated during the past quarter. A total of 21 trial runs were conducted in the two small columns. Three of these runs were conducted with FT-300 wax as the liquid medium, whereas tap/distilled water was the liquid medium in the remaining runs. The runs with FT-300 wax were conducted without solids at 265°C. Superficial gas velocities in the range 0.02-0.12 m/s were employed in all runs. Superficial liquid velocities in the range 0.005-0.020 m/s were employed in runs conducted in the continuous mode of operation. The 2 mm orifice plate distributor was used for all runs in both columns. Results from these runs indicate that the medium foams in the batch mode of operation, resulting in higher gas hold-ups, whereas, in the continuous mode of operation (with a superficial liquid velocity of 0.005 m/s) hold-ups were significantly lower. Experiments with water were conducted in both, continuous and batch modes of operation. Both iron oxide and silica (0-5 μm and 45-53 μm) were used in these runs. Runs with iron oxide in water gave significantly higher gas hold-ups compared to hold-ups from runs with silica in water, primarily because the former tends to produce foam. Solids concentration did not have a significant effect on the average gas hold-up. With iron oxide, the hold-up was significantly greater when the larger 45-53 μm size particles were used, compared to values obtained with 0-5μm particles. The most interesting observation from the runs with water and solids was the effect of liquid velocity on the axial dispersion of solids. For 0-5 μm silica in water, a small increase in liquid velocity resulted in a significantly greater improvement in solids dispersion than did a large increase in the superficial gas velocity.

The trial runs revealed several operational problems in the system, particularly in the presence of solids. In runs with 20 wt.% silica (0-5 μm) in distilled water, we experienced problems with solids settling in the line between the slurry storage tank and the column inlet. The settling of solids also lead to the excessive wear of the graphite seals and bearings in the slurry pump, limiting the number of runs that could be conducted in the continuous mode of operation. Some problems were also encountered with the DP system installed on the small steel column. As a result of our experiences with the system during the trial runs, several modifications were initiated during the later part of the past quarter. We decided to redesign the slurry feed system (replace 1/2" tubes with 1" tubes, and reduce the total length and avoid horizontal lines). The configuration of the pump will be changed from horizontal flow to vertical flow with suction at the top and outlet at the bottom. The orifice meter for measurement of the slurry flow rates will be replaced with a calibration chamber located in the return line from the top of the column to the storage tank. We are also examining different options for the DP system to improve its performance. We expect to alleviate most of the operational problems with these modifications.

During the past quarter we located the sources and placed orders for parts needed to build the movable assembly mechanism for the nuclear density gauges. We have ordered all parts except for the ball screws which will be used to move the density gauge axially and radially along the column. Several of the parts have already been received. Schematics for parts that require machining have been prepared and sent out for obtaining quotes.

IV. Detailed Description of Technical Progress

A. Task 1 – Project Work Plan

The Project Work Plan was submitted to the DOE in July, 1987 with the exception of the matrix of proposed experiments. This section of the Project Work Plan was submitted in January, 1988 as an Appendix of the Quarterly Technical Progress Report for the period October 1 - December 31, 1987.

B. Task 2 – Design and Construction of the Experimental Equipment

During the past quarter, the remaining piping for the small (2 inch diameter) bubble columns was completed and the system made operational for both batch (no liquid flow) and continuous modes of operation. All pipelines and process vessels associated with the two small columns were heat traced with beaded heaters/heating tapes and insulated. Thermocouples and temperature controllers were installed and tested. All rotameters (used to control the flow of nitrogen purge to the DP system), mass flow meters, and pressure transducers were calibrated. The bottom of the slurry storage tank for the large (9" diameter) columns has been changed to a conical shape to prevent the accumulation of solids in the storage tank.

The sampling system, which will be used to withdraw slurry samples at various heights along the small stainless steel column, was installed and tested. The system consists of five 0.25" air actuated high temperature ball valves attached to the column at five different heights. A sampling chamber is then attached to each of these valves. The sampling chamber consists of a piston located inside a cylindrical tube. Air and vacuum lines attached to each of the sampling cylinders, control the movement of the piston and allow for the simultaneous withdrawal of samples from all five locations. All parts, except for the sampling vessels, are heat traced and insulated. During normal operation, the sampling vessels have the piston pushed all the way in. To initiate sample withdrawal, the five air actuated valves are first opened simultaneously using an air switch (four-way ball valve). The vacuum lines are then activated to pull the piston out of the sampling chambers and withdraw slurry samples. The air actuated valves are then closed. Each of the sampling chambers is then disconnected manually and the sample removed. The clean chambers are then reinstalled on to the sampling system. This system has been successfully tested with cold slurries (water and iron oxide/silica). Tests have not yet been made with a hot slurry (molten wax and solids). With hot slurries, we plan to cool the sampling chambers once they are disconnected from the sampling system. This would solidify the wax into a solid plug, which can easily be removed by pushing it out using the piston.

Filtration of the slurry samples required excessive time when filter papers were employed and also resulted in loss of particles through the filter. We subsequently designed and constructed a filtration unit using a 1.5" diameter galvanized iron pipe, approximately 15" long (schematic of the unit is shown in Figure 1). A 2 μm sintered metal plate was used as the filtering element in this unit. The unit was successfully tested with a 30 wt.% slurry of water/silica using particles in the 0-5 μm size range. Four additional units were then built so that slurry samples from the five sampling ports could be filtered simultaneously. The units are heat traced and insulated. Before a wax slurry sample is analyzed, the filtration unit is heated to a temperature of 100°C. The solidified slurry sample, removed from the sampling chamber, is weighed and put into the filtration unit. After the slurry in the unit has melted, which takes around ten minutes, hot varsol (4 times the volume of the slurry) is introduced into the filtration unit. The unit is then closed and pressurized to 2 psig. The pressure is increased by 2 psi every 10 minutes (not to exceed 20 psig) until filtration is complete. Following this the pressure in the unit is reduced to atmospheric pressure and the lid removed. Approximately 50 cc of additional hot varsol is introduced into the unit and the filtration procedure repeated. This serves to rinse the solids of any remaining wax. The sintered metal plate filter element is then removed and all solids collected on a filter paper. The filter element, and the filter paper with solids, are dried in an oven maintained at 150°C. The solids are then weighed and the weight fraction of solids calculated. The filter elements are soaked in a beaker of hot varsol for 10-15 minutes before they are ready to be used again. The whole procedure takes about two hours.

A total of 38 filtration experiments were conducted using a slurry of wax with known weights of solids. Eighteen of these experiments were conducted using solids (iron oxide and silica) in the 45-53 μm size range, and the remainder were conducted with solids in the 0-5 μm size range. Majority of the experiments were conducted using 20 wt.% of solids, and a limited number were conducted using 10 wt.% and 30 wt.% of solids. We used 2 μm sintered metal plate filters for experiments with 0-5 μm particles, and 10 μm filters for experiments with 45-53 μm particles. The filtration units performed very well with these samples, and solids recovery was within ±3 % in most cases. We have decided to make some minor modifications of the units to reduce the total time required for filtration.

The trial runs in the two small columns revealed several operational problems in the system, particularly in the presence of solids. In runs with 20 wt.% of 0-5 μm silica particles in distilled water we experienced problems with solids settling in the lines between the slurry storage tank and the column inlet. This occurred because the slurry flow rates employed were not sufficient to keep the solids suspended. We also experienced difficulties in measuring slurry flow rates with the orifice plate at low flow rates (superficial liquid velocities of 0.005 m/s in the small column). Settling also caused the accumulation of solids in the slurry pump, resulting in excessive wear of the pump graphite seals and bearings. Experiments with water and solids also were detrimental to the pump's performance. The viscosity and lubricity of the water/solids slurry were inadequate, and the slurry pump became inoperable following these runs. Replacement parts for the pump have been ordered and will be installed once we receive them. Some problems were also encountered with the DP system. The orifice at the pressure sensing point, which is in contact with the slurry, plugged during some runs when solid particles got lodged in the hole. We decided to test different options to improve upon the present system. One of these options involved the use of an external filter in conjunction with a nitrogen purge. This allowed us to use a larger hole at the pressure sensing point. The use of such a device at one of the DP cell locations proved to function satisfactorily in experiments with a slurry of 45-53 μm silica particles in water. Other options are currently being examined. The high temperature pressure transducers (from Sensotron) have not yet arrived. We will therefore continue to use the low temperature Validyne transducers instead.

As a result of our experiences with the system during the trial runs, several modifications were initiated during the later part of the past quarter. We decided to redesign the slurry feed system and to eliminate the orifice plate for measurement of the slurry flow rate during the continuous mode of operation. A schematic of the modified slurry feed system is shown in Figure 2. All 1/2" lines carrying the slurry will be replaced with 1" tubes. The total length of these lines will be reduced and horizontal sections eliminated. The larger lines will help decrease pressure drops across the lines, and the absence of horizontal lines will prevent the settling of solids in the lines. The gas inlet line has been moved from the bottom of the plenum chamber (chamber below the distributor) to the side of the chamber. This should prevent the plugging of this line with slurry that might weep through the distributor during an experiment. A new stand has been fabricated for the pump, which will enable us to change the orientation of the slurry pump so that instead of having horizontal flow through the pump, we can now have vertical flow through the pump. The pump will be located directly below the storage tank, and feed will enter at the top of the pump and will leave at the bottom. We expect to prevent any settling of solids in the pump using this configuration.

The slurry flow rate will be measured using a calibration chamber located in the return line from the top of the column. A schematic of the circuit containing the calibration chamber is shown in Figure 3. We have designed and fabricated a 9" diameter cylindrical tank for this purpose with an internal volume of approximately 4 liters. A drain valve is located at every liter, giving 4 calibration points for slurry flow rate measurements. The calibration chamber will also have a bypass line, through which the slurry will flow once the flow rate has been determined. During flow measurements, the slurry will be allowed to accumulate in the chamber and the time taken to fill the chamber up to each of the four drain valves will be noted and the flow rate determined. Once these measurements are complete, the slurry in the chamber will be drained into the slurry storage tank, located directly below the calibration chamber. The chamber will then be isolated using ball valves located above and below it and slurry flow will be redirected through the bypass line. The desired slurry flow rate is obtained by controlling the flow rate through the pump and the flow through the bypass line connecting the pump outlet back to the storage tank (see Figure 2). The flow rate through the pump can be varied by changing the pump speed, while a valve in the bypass line controls flow through that line. Our experience from previous experiments with liquid flow through the column has shown that fairly good flow control is achieved by maintaining a relatively high flow through the pump and then fine tuning the feed flow rate by controlling the amount of slurry that is cycled back to the storage tank through the bypass line. The calibration tank will be used to monitor the feed flow rate during this period.

The introduction of the calibration chamber and the installation of larger diameter feed lines in the slurry flow circuit will increase the slurry hold-up in the system. To accommodate this additional volume, a larger slurry storage tank was designed with a volume that is approximately one and a half times the volume of the storage tank currently in use. The new tank will also have a conical bottom to prevent the accumulation of solids. The old storage tank will be modified and used as a solvent tank for cleaning the columns.

We expect to complete the modifications/improvements of the two small columns and related equipment, and have the system operational early in the next quarter. Hot flow experiments will then be resumed.

At the present time we plan to classify solids into the 45-53 μm range ourselves for experiments in the small columns. However, for experiments in the large (9" diameter) columns, we will have to use an outside source for grinding and sieving. Vortex Corp. of Long Beach, California, can grind and sift silica, however, they can only sift iron oxide. Since the price of processing solids is based on the weight of original raw material processed, we are trying to locate alternative sources for iron oxide and silica so as to minimize the cost involved in obtaining a cut in the 45-53 μm size range.

C. Task 3 – Measurement of Hydrodynamic Parameters by Conventional Techniques

A total of 21 runs were conducted in the two small (2" diameter) columns during the past quarter. The 2 mm orifice plate distributor was used in all experiments. The first few experiments were conducted using superficial gas velocities of 0.01-0.05, 0.07, 0.09 and 0.12 m/s, thereafter all experiments were conducted using superficial gas velocities of 0.02, 0.04, 0.06, 0.09 and 0.12 m/s. Majority of the experiments were conducted with distilled or tap water under ambient conditions. The cold flow experiments served to shakedown the system and to establish the behavior of water in our system in the presence and absence of solids. Three experiments were conducted at 265°C using FT-300 wax. Both solid types (iron oxide and silica) in the two size ranges (0-5 μm and 45-53 μm), and three different weight percents (10, 20 and 30), in addition to 0 wt.% solids, were used in the various experiments. Gas hold-ups in the glass column were determined using visual measurements of the expanded and static heights of the slurry. In the stainless steel column, differential pressure measurements were used to obtain average gas hold-ups. Slurry samples withdrawn at a maximum of five locations (depending on the expanded height of the slurry) along the height of the stainless steel column, were used to obtain the solids concentration profiles. Only a limited number of experiments were conducted with liquid circulation because the pump became inoperative after only seven runs.

For runs with water, the duration of the experiment was 0.5 to 1 hr per gas velocity employed, whereas with wax the duration was 1.5 hours per velocity. This gave sufficient time for the dispersion in the column to become stable. Three readings of the expanded height were taken for each velocity in the glass column, and of the differential pressure gauges for runs in the stainless steel column. One set of slurry samples were withdrawn at the end of the period with a given gas velocity, when solids were used in the run. After each velocity in the glass column, the gas was shut off and the bubbles in the dispersion allowed to disengage. The height of the ungassed slurry was then measured to obtain the static height of the slurry. The average gas hold-ups in the stainless steel column were estimated using the DP readings and the solids concentration profiles obtained from slurry samples (for cases where solids were used). We are in the process of streamlining the calculation procedure for estimating the various parameters, and the details will be included in the next quarterly progress report. Results from some of these trial runs are presented here and are categorized in terms of different effects.

C.1. Effect of liquid velocity:

Experiments were conducted with FT-300 wax at 265°C without solids in the 2" stainless steel column to study the effect of liquid velocity on the average gas hold-up. In Figure 4, results from the run with no liquid velocity (batch mode of operation) are compared with results from an experiment where a superficial liquid velocity of 0.005 m/s was employed. Hydrodynamic studies with FT-300 wax conducted in our previous DOE contract (DE-AC22-84PC70027; Bukur, et al., 1987) showed that FT-300 wax has a tendency to foam when operating in the batch mode using increasing order of gas velocities. Results shown in Figure 4 for the batch case are consistent with those findings. During this run, hold-up increased sharply as gas velocity was increased from 0.02 m/s to 0.04 m/s, indicating the formation of foam. However, as gas velocity was further increased, the increase in gas hold-up was gradual, and it reached 31.6 % at a superficial gas velocity of 0.12 m/s. For the run with a small superficial liquid velocity (0.005 m/s), results were significantly different at gas velocities of 0.04 and 0.06 m/s. At 0.04 m/s the gas hold-up was only 14.5 % compared to 24.6 % for the batch case. Similarly at 0.06 m/s hold-up in the continuous mode of operation was 19.9 % compared to 26.9 % for the batch case. Hold-up for the continuous case increased gradually as gas velocity was increased, reaching values similar to those for the batch case at a superficial gas velocity of 0.12 m/s. In a batch run conducted in the small glass column with FT-300 wax at 265°C, a significant amount of foam was produced at velocities greater than 0.02 m/s, and gas hold-up approached 47% as the superficial gas velocity was increased to 0.06 m/s. A further increase in gas velocity to 0.12 m/s did not show any appreciable changes in the gas hold-up values. These results indicate that even a small liquid velocity might be sufficient to inhibit the foaming tendency of the FT-300 wax. However, this conclusion, though logical, might be premature and more runs are necessary before any definite conclusions can be made.

C.2. Effect of solids type, size and concentration:

All runs with solids were conducted using distilled or tap water as the liquid medium. Figures 5 and 6 show the effect of solids type on the average gas hold-up. Average gas hold-ups from runs made in the batch mode in the glass column with 0-5 μm iron oxide and silica (20 wt.%) are shown in Figure 5. Hold-up values for water alone are shown for comparison. These results indicate that the addition of silica to water decreases the average gas hold-up, although the effect is not very significant. However, the addition of iron oxide to water seems to increase the gas hold-up significantly. Visual observations of the dispersion showed the presence of some foam on top of the dispersion at most gas velocities, which might be responsible for the relatively high hold-ups with this system. Figure 6 shows hold-up values for 45-53 μm silica and iron oxide in tap water. Results from runs made in both the glass and stainless steel columns with iron oxide are shown. Once again hold-ups with the iron oxide are significantly higher than those for the silica slurry. In both columns, the iron oxide slurry gave high gas hold-ups and the values from the two runs are very similar, indicating fairly good reproducibility of results. Visual observations of the run with iron oxide in the glass column clearly showed the presence of foam at all gas velocities employed. The foam consisted of bubbles between 5 and 10 mm in diameter. At gas velocities of 0.04 m/s and higher, slugs could clearly be seen rising through the dispersion.

Figure 7 shows the effect of solids concentration on the average gas hold-up for runs made in the glass column with different sizes of iron oxide (0-5 μm and 45-53 μm). A 20 wt.% slurry in water was used in each case. The hold-up with the larger particles is significantly higher than hold-ups for the 0-5 μm iron oxide slurry. Foam was present in both cases, with higher amounts produced with the slurry containing larger particles. The reasons for these differences are not clear at this point, although it is possible that the differences between distilled water (used as the liquid medium with the 0-5 μm iron oxide) and tap water (used with the 45-53 μm particles) could have influenced the hold-up values to a certain extent.

The effect of solids concentration on average gas hold-up is illustrated in Figure 8 for experiments conducted with a slurry of water and 0-5 μm silica. Results for slurries containing 0, 10, 20 and 30 wt.% solids are shown. These results clearly indicate that solids concentration does not have an appreciable effect on the average gas hold-up, although the addition of solids (silica) gives slightly lower hold-ups compared to values for the run with no solids.

C.3. Solids concentration profiles:

The solids content of slurry samples withdrawn at different heights along the length of the stainless column, gave the solids concentration profile. These profiles provide an indication of how well the solids are axially distributed in the system.

Figure 9 shows concentration profiles from a batch run conducted using 20 wt.% of 45-53 μm silica in water. The effect of superficial gas velocity on the concentration profile is evident from these results. At a gas velocity of 0.02 m/s, the dispersion of solids is very poor as indicated by the open circles. The lowest section of the column contains 13 wt.% solids while only 7-8 wt.% solids are present in sections above. Since the original slurry contained 20 wt.% solids, it is clear that a large portion of solids are not accounted for in this concentration profile. This is probably because these unaccounted solids have accumulated on the distributor plate. The lowest sampling port is located at approximately 15" above the distributor plate, therefore any solids present below this port would not show up in the concentration profile. When the gas velocity is increased to 0.04 m/s, the solids concentration becomes more uniform and approaches 20 wt.%. Further increases in gas velocities cause only a minor change in the concentration profile, with the most even distribution obtained at 0.12 m/s. The slight upward shift in the concentration profile with an increase in gas velocity is because more and more solids are carried from the dead zone (between the distributor and the lowest sampling port) to the section above the lowest sampling port.

The concentration profiles from a batch run conducted with a slurry containing 20 wt.% of 45-53 μm iron oxide in water are shown in Figure 10. The dispersion of solids is very poor at all gas velocities for this run, with most of the solids remaining in the lower section of the column. Almost no solids are carried up to the upper three sections. The figure shows that at 0.04 m/s the profile is higher than at other gas velocities. The reasons for this are not apparent since concentrations at 0.12 m/s are expected to be higher than concentrations at 0.04 m/s. This could be attributed to experimental error. The slurry samples are withdrawn in a very short time at a given velocity (it takes less than 5 seconds to withdraw the samples). Therefore, if the concentration profile is actually the same at all velocities, the differences in the profiles could be simply due to the random fluctuations that take place in the dispersion at all times.

Figures 11 and 12 show solids concentration profiles obtained with 20 wt.% slurries of 0-5 μm silica in water. For the run in Figure 11, a superficial liquid velocity of 0.0065 m/s was employed, while a liquid velocity of 0.019 m/s was used for the results presented in Figure 12. Several interesting observations can be made from the results shown in these figures. It is obvious that the small silica particles can be easily dispersed in water, even at very low gas velocities. As indicated in both figures, no improvement in the dispersion takes place when gas velocity is increased from 0.02 m/s to 0.12 m/s. However, when we compare the profiles in Figure 11 with those in Figure 12, we can see that the distribution at the higher liquid velocity (0.019 m/s) is more uniform (Figure 12) than the distribution at a liquid velocity of 0.0065 m/s (Figure 11). This appears to suggest that even a relatively small increase in liquid velocity might be sufficient to cause an appreciable improvement in the axial distribution of solids, whereas, large increases in the gas velocity may not help at all. The two figures also illustrate the problems we encountered with settling of solids in the slurry feed lines and in the pump (discussed in Section B, above). In both cases, the solids concentrations are 3-7 wt.% (absolute) lower than the expected 20 wt.% despite nearly uniform concentration profiles. The uniform profiles also imply that the solids concentration in the dead zone between the distributor and the lowest sampling port must be similar to the values just above the port. Therefore the unaccounted solids must have settled in the slurry lines and in the pump, as was the case. The results presented in Figure 11 also show that the concentration profile shifts downwards with an increase in gas velocity, which is unexpected. However, if solids are settling in the lines, the loss of solids from the slurry would increase as time progresses, i.e. as we increased the superficial gas velocity. This continuous loss of solids is reflected by the lowering of the concentration profile with an increase in gas velocity, though there may not be any relation between the two. This is not evident from the profiles in Figure 12, probably because a higher liquid velocity was employed in that run. The results from these two figures show that the concentration profiles could be used to indicate operational problems with the system.

D. Task 4 – Application of a Gamma Radiation Density Gauge for Determining Hydrodynamic Parameters

During the past quarter, we located all manufacturers of the parts needed to build the movable assembly mechanism for the density gauges, and began placing orders for these parts. We have ordered all parts except for the ball screws which will be used to move the density gauges axially and radially along the column. The parts ordered include the drive mechanism and the support assembly. The drive mechanism consists of the motor, controller (which controls the motor speed and direction of motion - forward or reverse) and the reducing gears. A separate drive mechanism has been ordered for axial and radial motion of the gauges. The support assembly for axial motion of the density gauges includes two 2" diameter by 125" long support shafts and four pillow blocks. The pillow blocks will enable the support platform, which holds the density gauges, to slide along the support shafts. The support assembly for radial motion includes four 3/4" diameter by 24" long support shafts and eight pillow blocks. We have already received the drive mechanism and the support system for radial motion, and the support system for axial movement.

The schematics for the machining of the ball screws have been prepared and sent to the manufacturers. We are awaiting prices on these parts before we can place an order. We plan to order these items and material for the support platforms early in the next quarter. Once all the items are received, we will commence with the fabrication/assembly of the movable assembly mechanism.

V. NOMENCLATURE

H_s	static bed height (m)
u_g	superficial gas velocity (m/s)
u_l	superficial liquid velocity (m/s)
W_s	weight percent solids in slurry

VI. REFERENCES

Bukur, D.B., Daly, J.G., Patel, S.A., Raphael, M.L. and Tatterson, G.B., Hydrodynamics of Fischer-Tropsch synthesis in slurry bubble column reactors, Final Report to the Department of Energy for Contract No. DE-AC22-84PC70027 (1987).

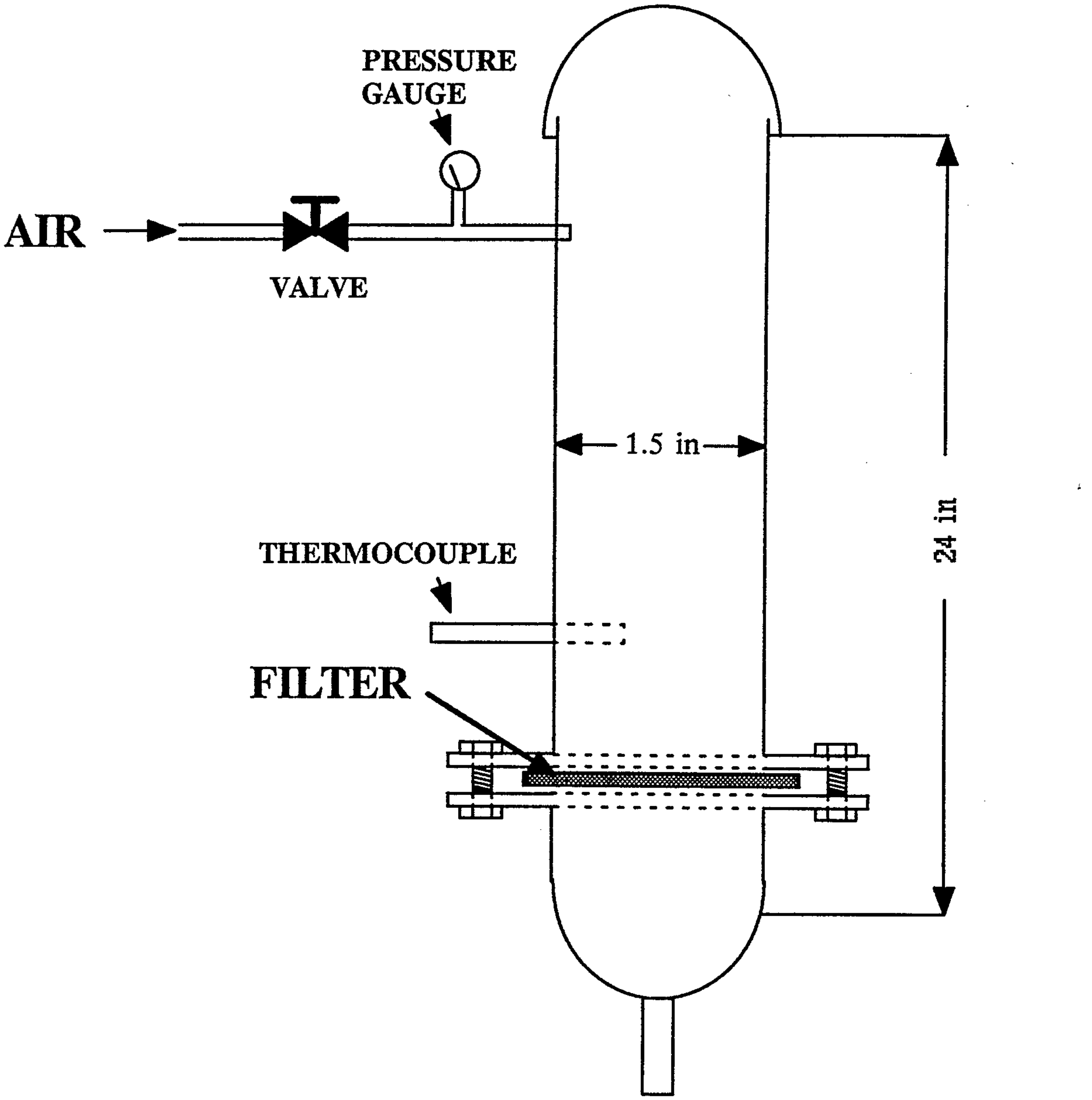

Figure 1. Schematic of the filtration unit.

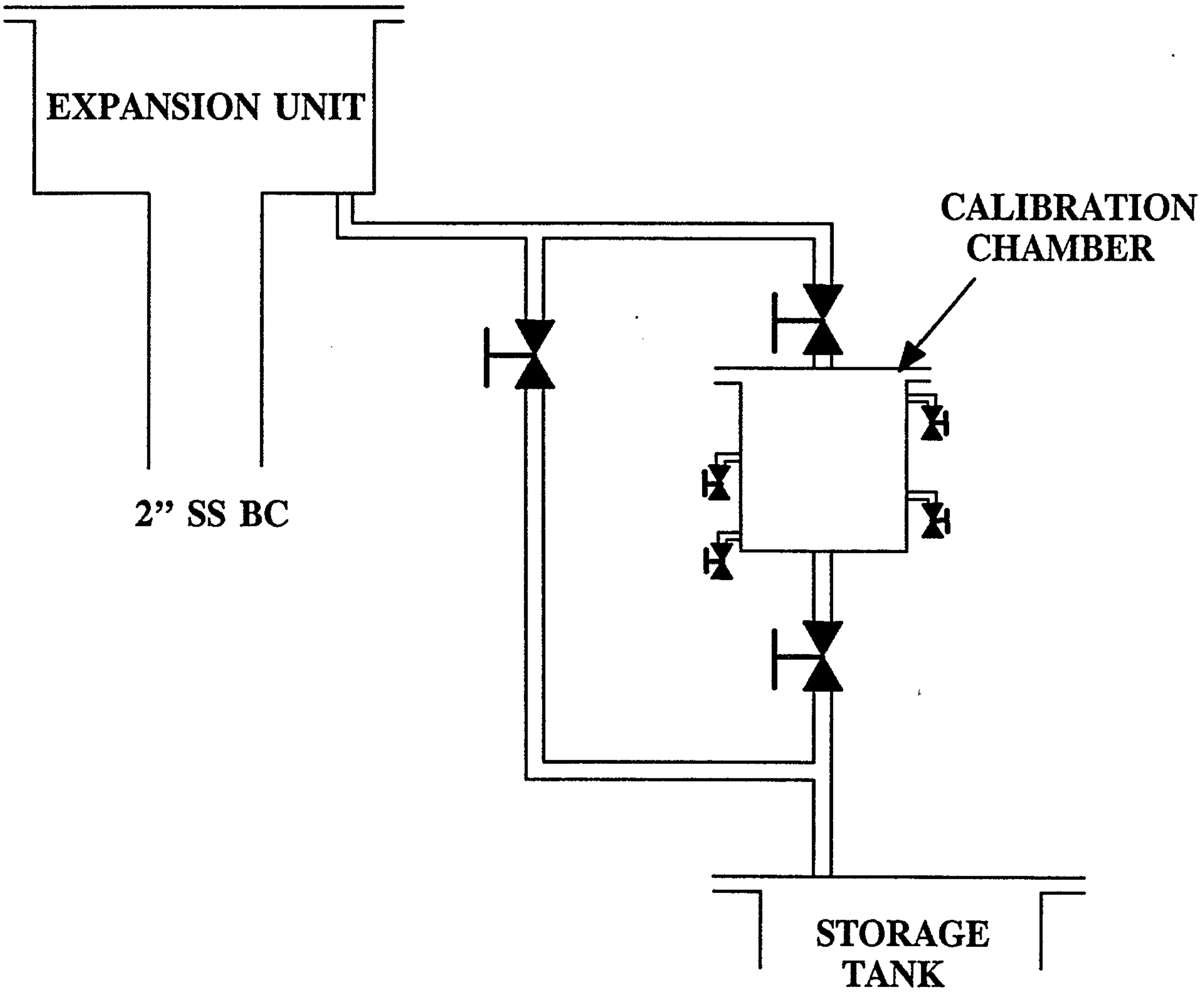

Figure 2. Schematic of recirculation system.

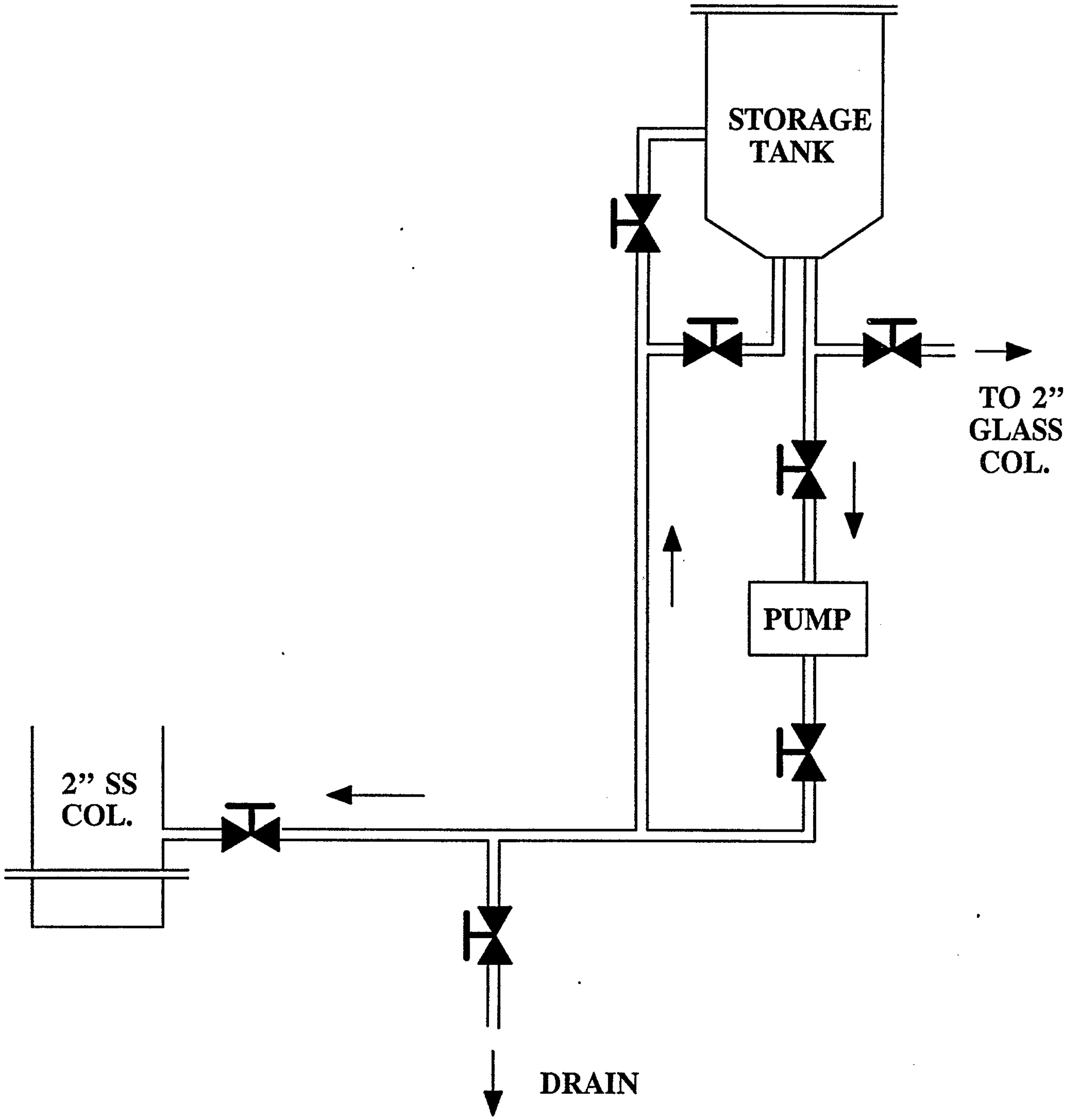

Figure 3. Schematic of slurry feed system

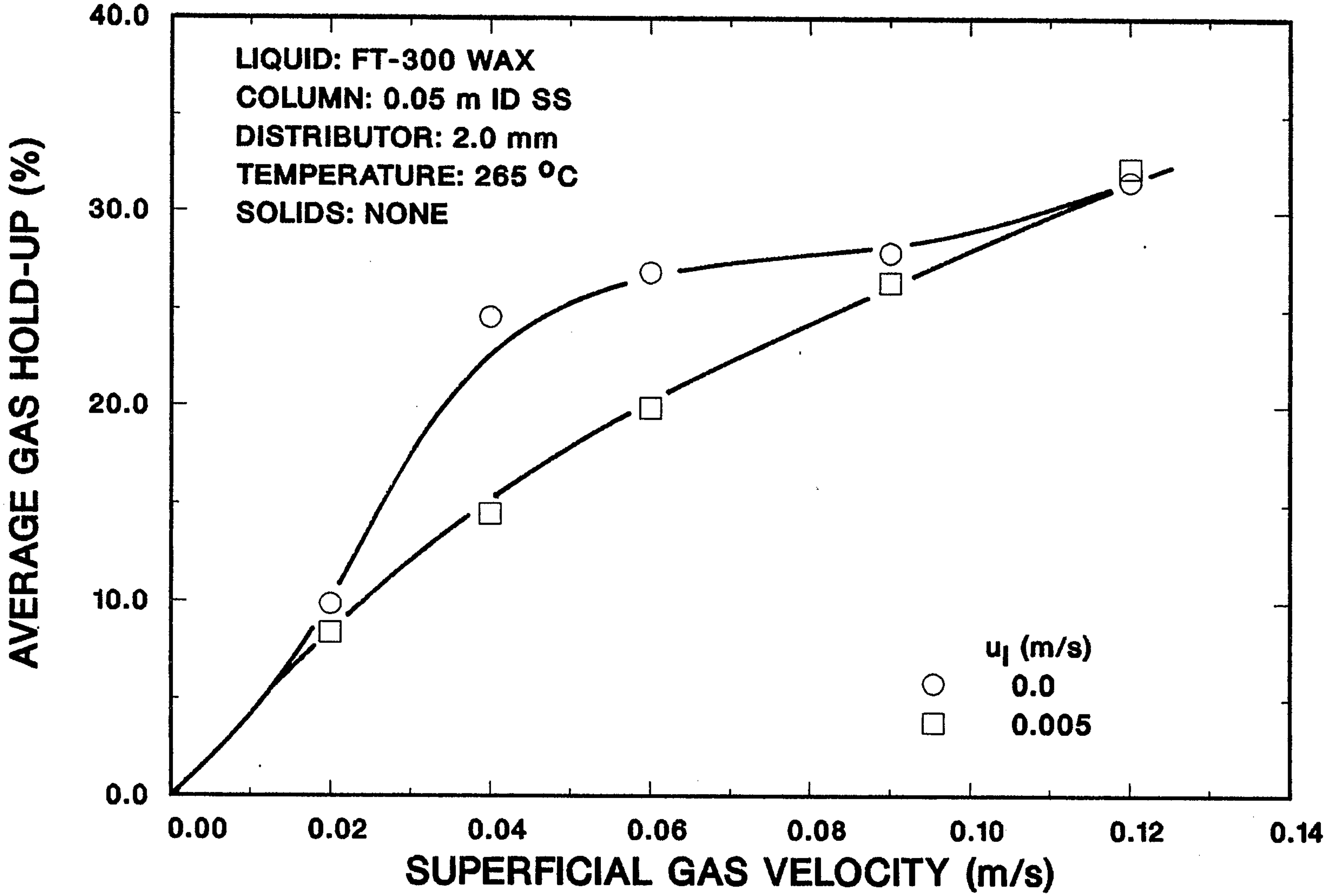

Figure 4. Effect of liquid velocity on average gas hold-up.

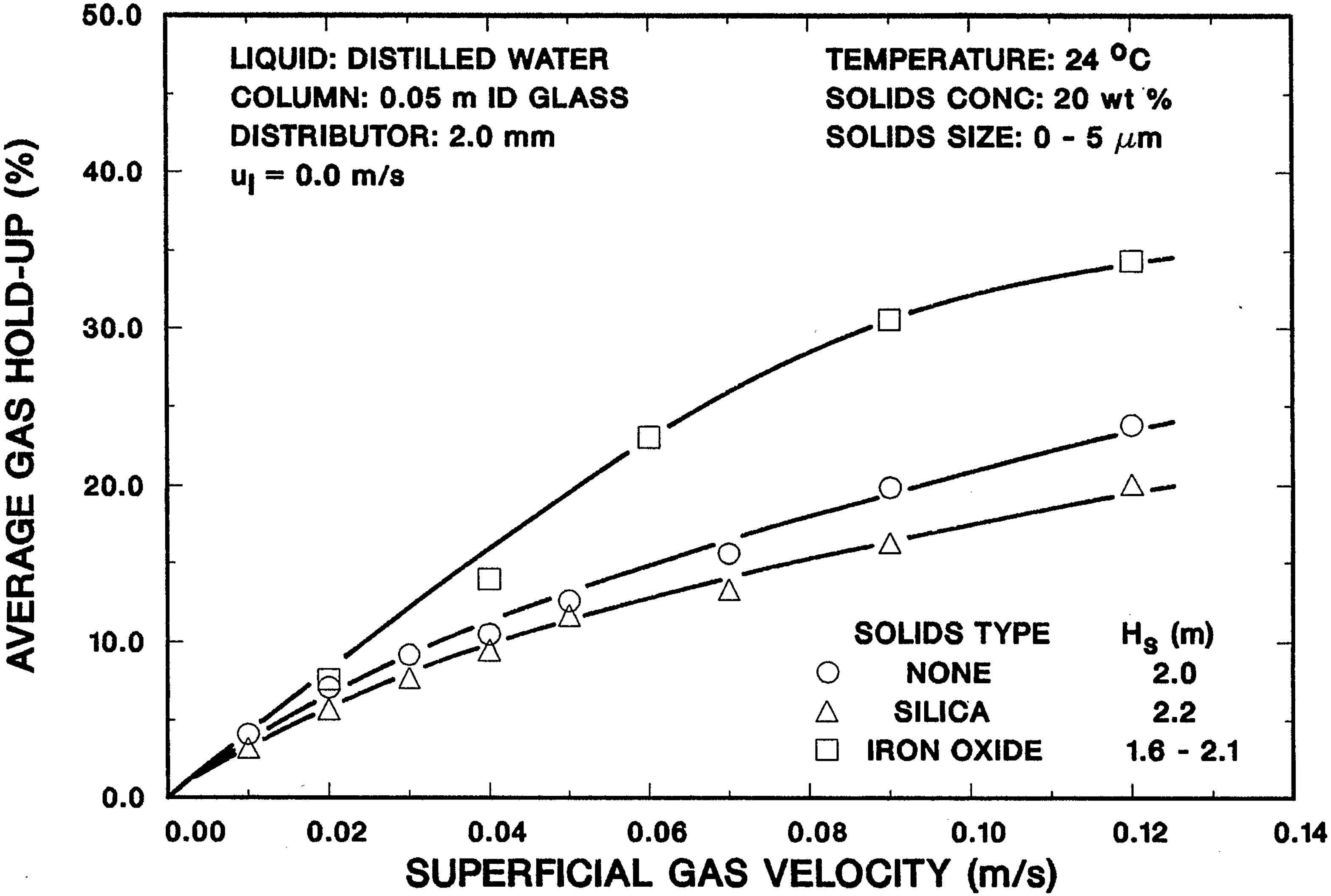

Figure 5. Effect of solids type on average gas hold-up.

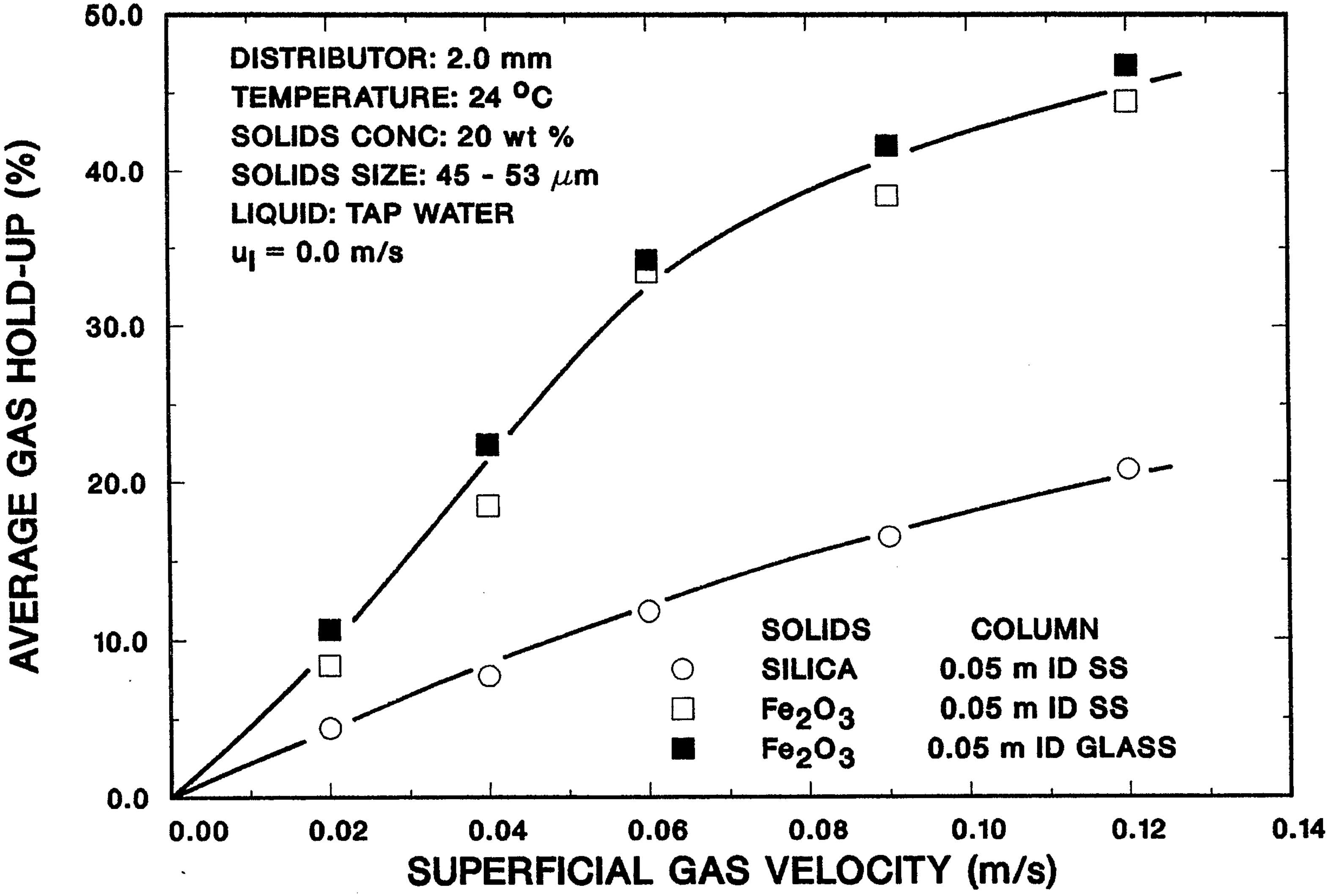

Figure 6. Effect of solids type on average gas hold-up and reproducibility.

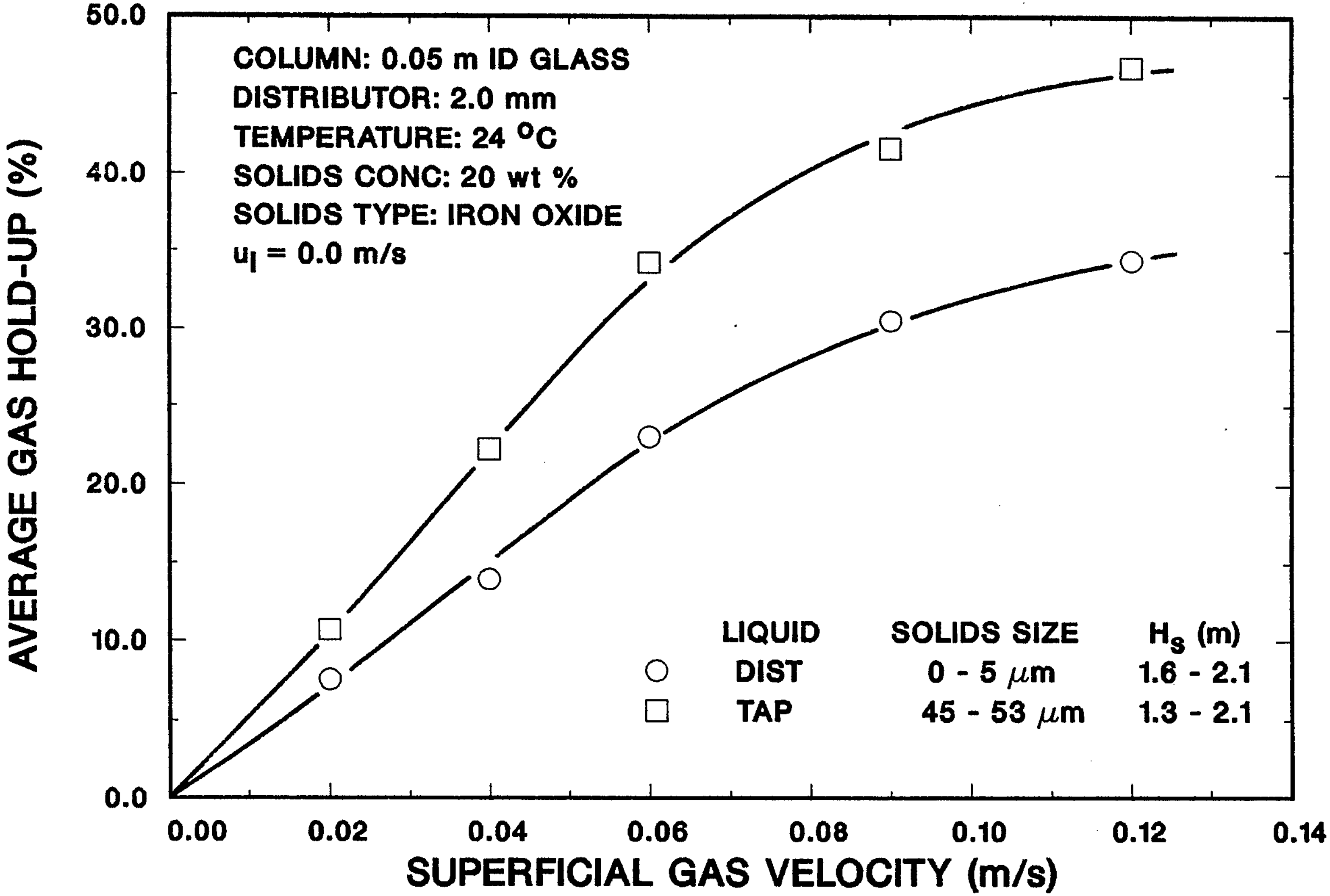

Figure 7. Effect of solids size on average gas hold-up.

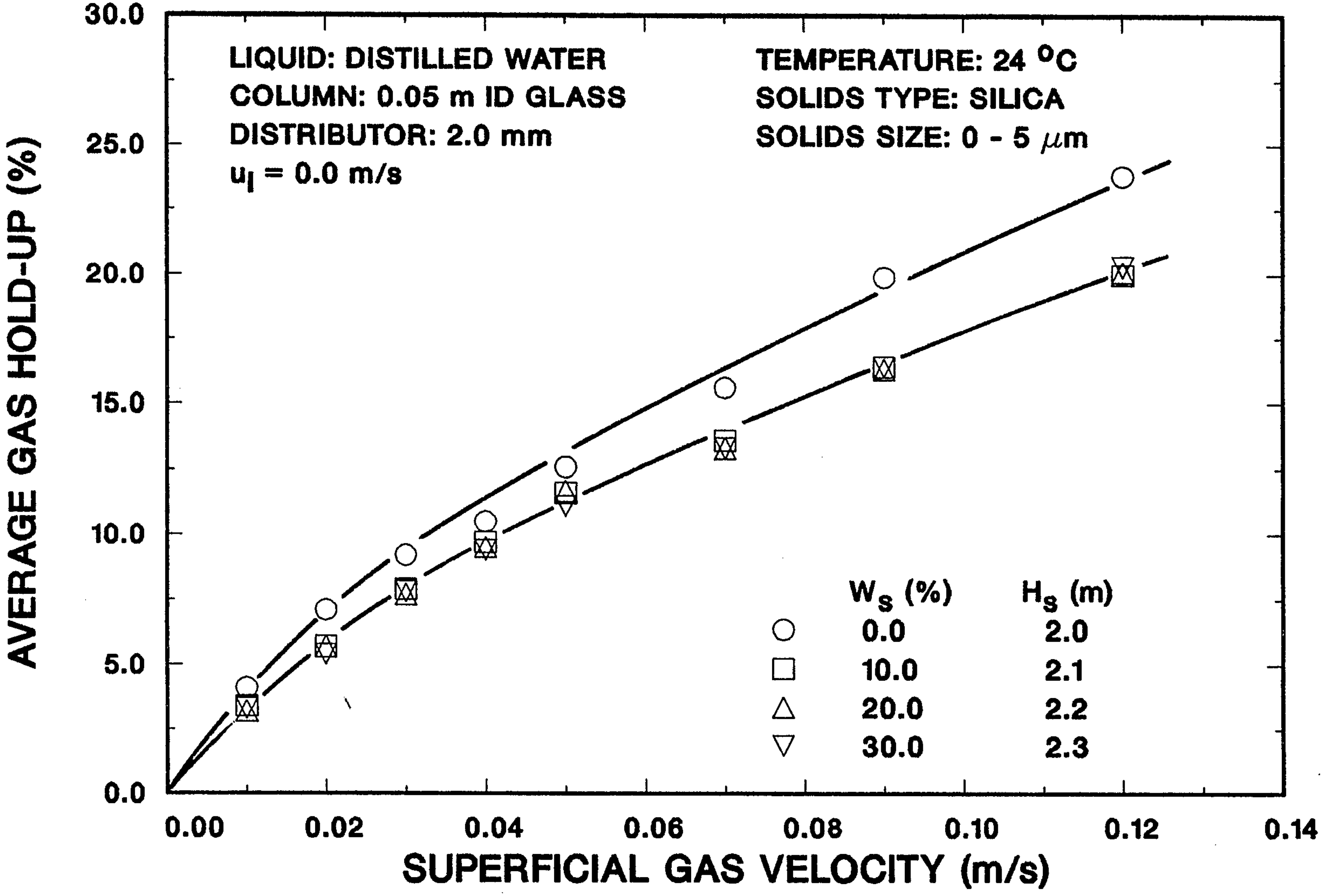

Figure 8. Effect of solids concentration on average gas hold-up.

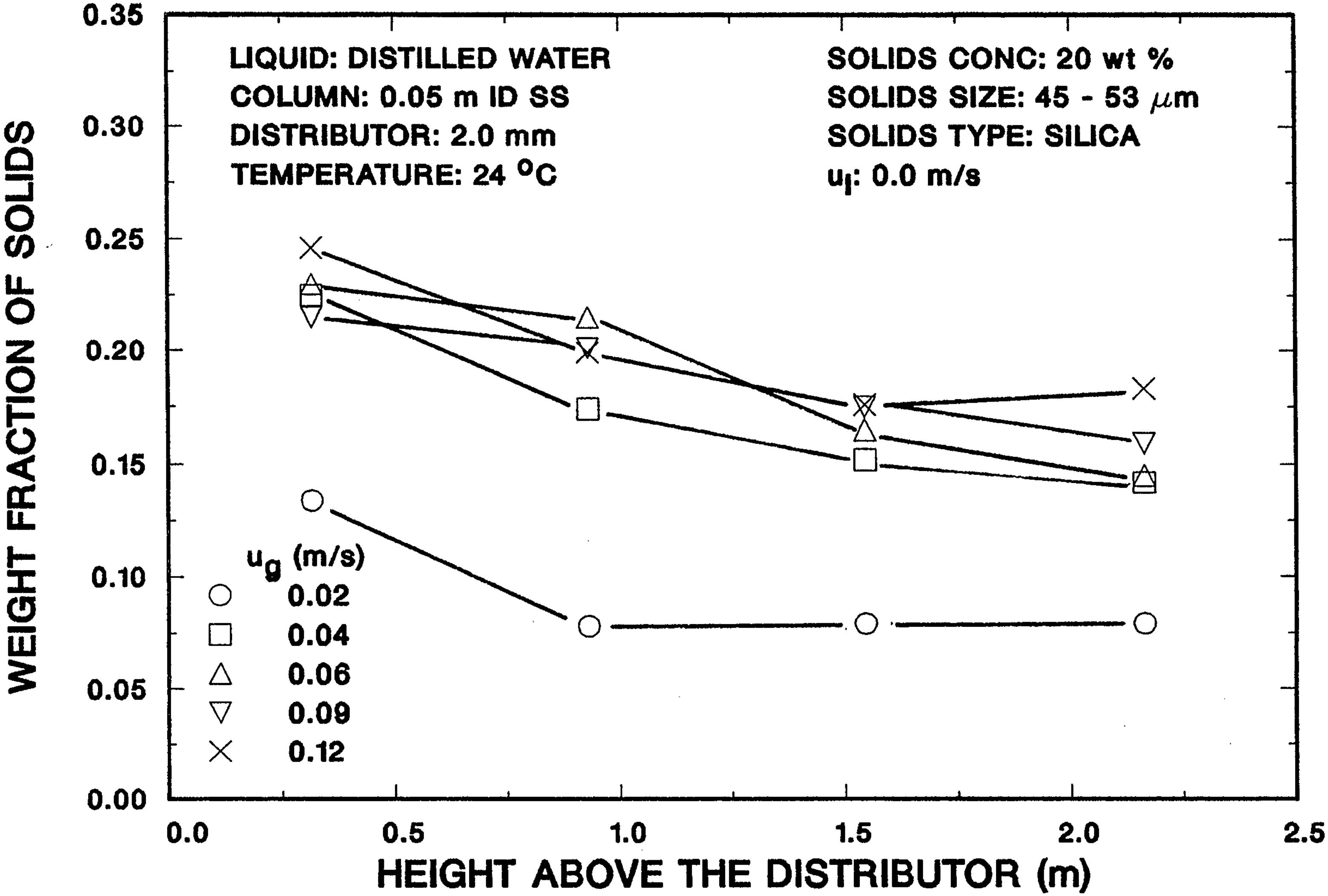

Figure 9. Effect of superficial gas velocity on axial solids dispersion.

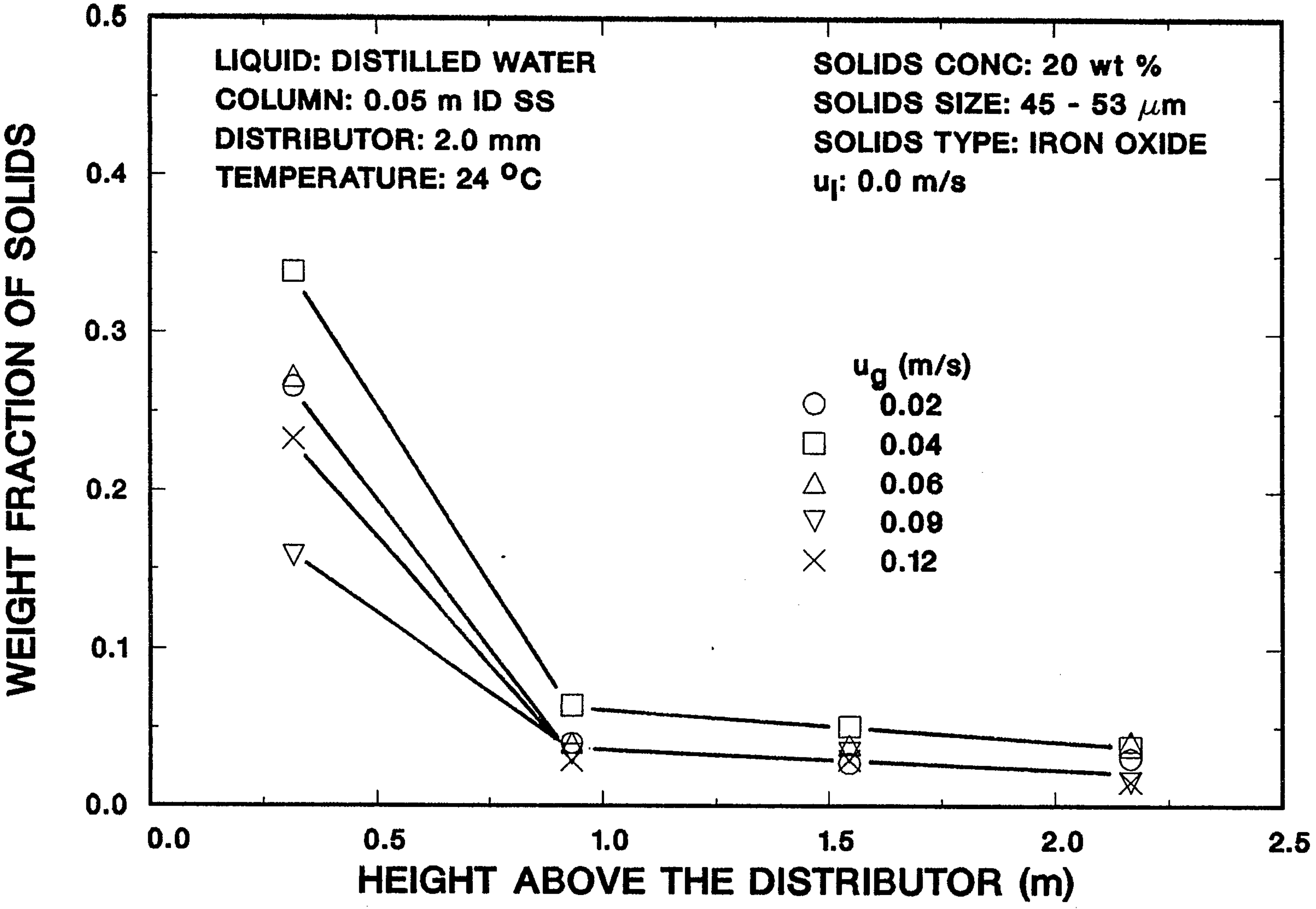

Figure 10. Effect of superficial gas velocity on axial solids dispersion.

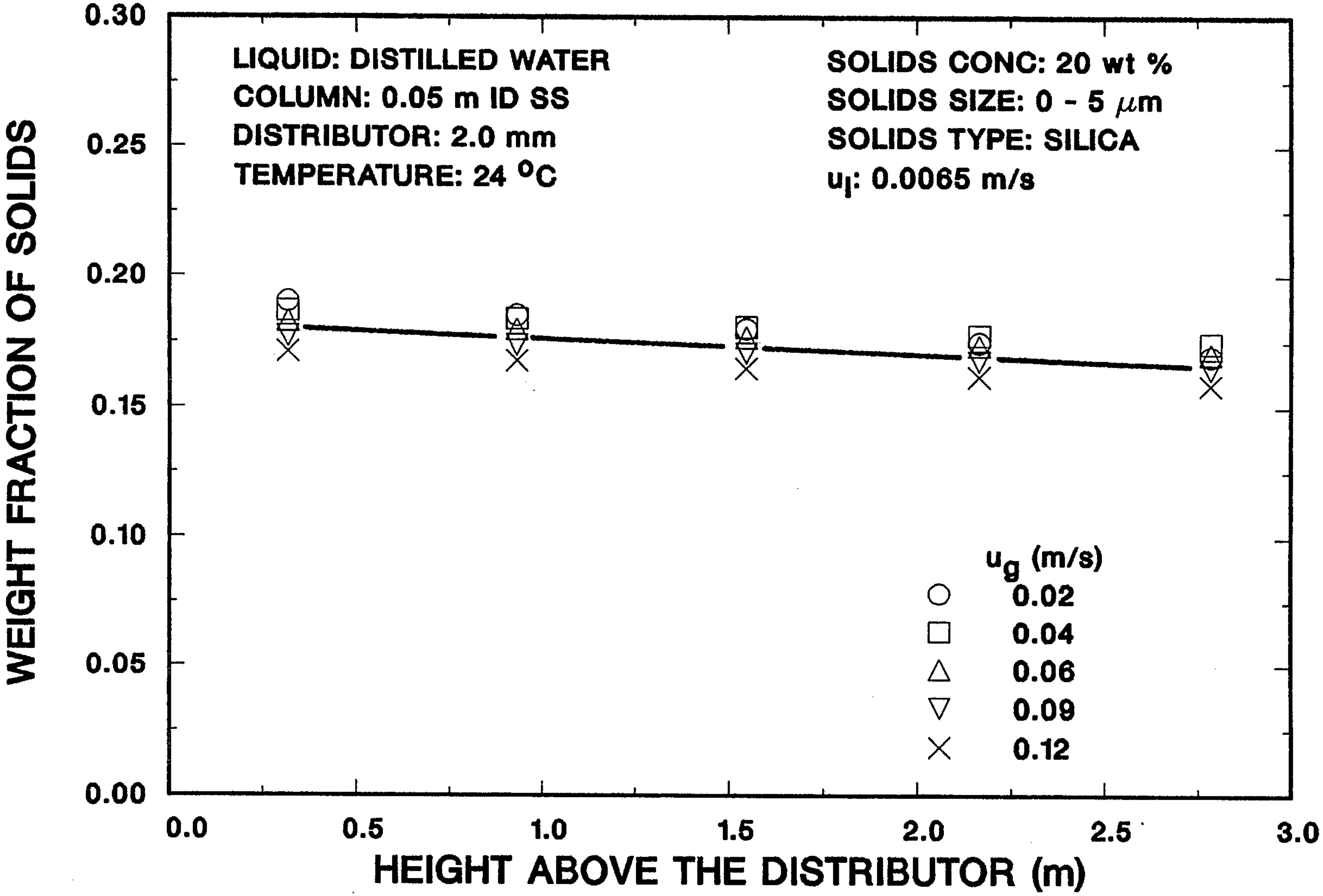

Figure 11. Effect of superficial gas velocity on axial solids dispersion.

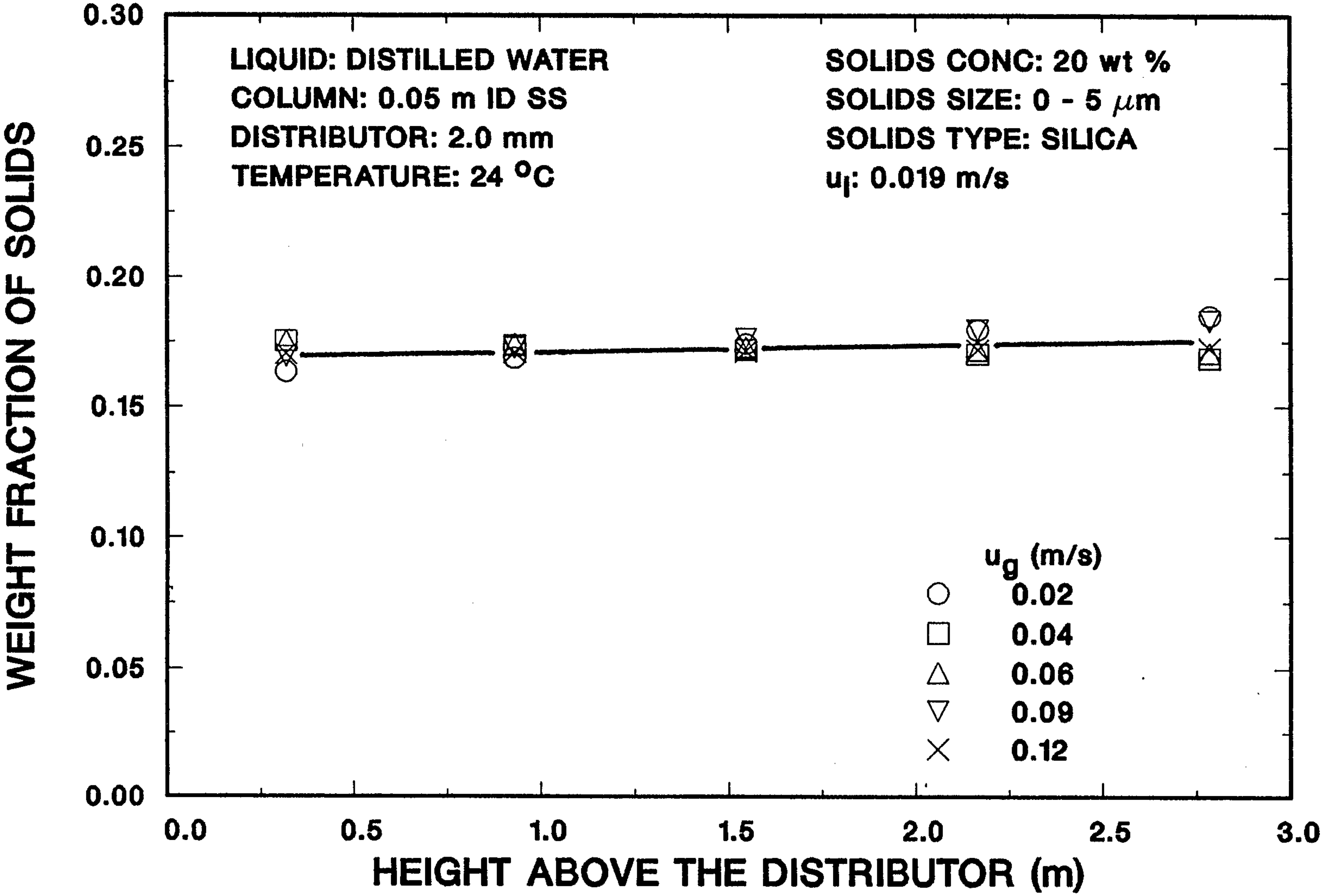

Figure 12. Effect of superficial gas velocity on axial solids dispersion.

DOE/PC/90012-2

Hydrodynamics of Three-Phase Slurry Fischer-Tropsch Bubble Column Reactors

Quarterly Technical Progress Report
for the Period 1 October 1987 - 31 December 1987

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January 31, 1988

Prepared for the Pittsburgh Energy Technology Center,
the United States Department of Energy Under Contract No. DE-AC22-86PC90012
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PATENT STATUS

U.S./DOE Patent Clearance is not required prior to the publication of this document.

TECHNICAL STATUS

This technical report is being transmitted in advance of DOE review and no further dissemination or publication shall be made of the report without prior approval of the DOE Project/Program Manager.

TABLE OF CONTENTS

I. Abstract

Fabrication of all new vessels and modifications of the existing vessels were completed for the small diameter bubble column apparatus. All vessels were installed, and the majority of piping has been completed. We completed ordering new instrumentation and equipment, including high temperature pressure transducers and a slurry pump capable of operating in the presence of solids and at elevated temperatures. We have received all instrumentation and equipment except the high temperature pressure transducers and the cables needed to connect the transducers to the readout. Also, during this period, we have completed the Experimental Design section of the Project Work Plan.

A preliminary design of the movable assembly mechanism needed to transport the nuclear density gauges axially and radially along the large stainless steel column has been completed. Fabrication of the calibration chambers and storage units for the radioactive sources has been initiated. We have begun looking at other possible source combinations which may work better for our system.

II. Objective and Scope of Work

The overall objective of this contract is to determine the effects of bubble column diameter, solids loading and particle size, and operating conditions (temperature, gas and liquid flow rates) on hydrodynamics of slurry bubble columns for Fischer–Tropsch synthesis, using a molten wax as the liquid medium. To accomplish these objectives, the following specific tasks will be undertaken.

Task 1 – Project Work Plan

The objective of this task is to establish a detailed project work plan covering the entire period of performance of the contract, including a detailed program schedule, analytical procedures, and estimated costs and manhours expended by month for each task.

Task 2 – Design and Construction of the Experimental Apparatus

The existing glass and stainless steel columns (0.051 m and 0.229 m in diameter, 3 m tall) that were constructed under our previous DOE contract (DE–AC22–84PC70027), will be modified and additions made in order to study the effect of continuous upward liquid flow. After the procurement of equipment and instrumentation, and construction of the unit is completed, a shakedown of test facilities will be made to verify achievement of planned operating conditions.

Task 3 – Measurement of Hydrodynamic Parameters by Conventional Techniques,

In this task, the effects of operating conditions (liquid and gas superficial velocities, temperature), gas distributor, column diameter, and solids concentrations and particle size on hydrodynamic parameters in the glass and stainless steel columns will be determined. All experiments will be conducted using nitrogen at atmospheric pressure.

The hydrodynamic parameters that will be determined as a function of the independent variables mentioned above are: average gas hold–up, axial solids distribution, axial gas hold–up, flow regime characterization, and qualitative information on bubble size distribution.

Task 4 – Application of a Gamma Radiation Density Gauge for Determining Hydrodynamic Parameters

The objective of this task is to determine hydrodynamic parameters for the three–phase system using a nuclear density gauge apparatus. A movable assembly mechanism and positioning racks for the two nuclear density gauges and detectors will be designed and constructed. Following the interfacing of the apparatus with an on–line microprocessor, the gauges will be calibrated using pure components (liquid wax and solid particles), and with known proportions of liquid and solid. After calibration, the following parameters will be obtained from experiments in the large stainless steel column: axial gas hold–up, axial concentration of solids, and qualitative information on flow regimes and bubble size distributions.

III. Summary of Progress

During this quarter, the Experimental Design section of the Project Work Plan was completed. All new vessels were fabricated and the existing vessels from our previous project (DE–AC22–84PC70027) were modified. All of the vessels were installed on the support structure, and a majority of the associated piping was completed. We were able to locate and order a majority of the instrumentation and equipment, including a high temperature slurry pump and high temperature pressure transducers.

Fabrication of the stainless steel calibration chamber and the storage units which will be used to store the radioactive sources when they are not in use was initiated. A preliminary design of the movable assembly mechanism which will be used to transport the nuclear density gauges radially and axially along the column was completed. Also, we are in the process of examining other possible source combinations which may be applicable to our system.

IV. Detailed Description of Technical Progress

A. Task 1 – Project Work Plan

The Project Work Plan was submitted to the DOE in July, 1987 with the exception of the matrix of proposed experiments. This section of the Project Work Plan was completed during this quarter and is given in Appendix I.

B. Task 2 – Design and Construction of the Experimental Equipment

During this quarter, a gear pump capable of pumping wax slurry at temperatures up to 260 °C was ordered and received. The pump is an ECO gear pump, model G12CCk–KK44–E, which is manufactured by Pulsafeeder. The pump meets all of our needs but imposes limitations on the size particles which can be handled. Due to the close tolerances between the gears and pump casing, the maximum size particle which the pump can handle is 65 μm. Also, we ordered two high temperature pressure transducers (Sensotron Model 101A–10G–250–1–A) which can be mounted directly to the wall of the stainless steel columns. These transducers will be used to obtain wall pressure fluctuations which will be used to give indication of the flow regimes and flow regime transitions in bubble columns. All instrumentation has been received and installed with the exception of the high temperature pressure transducers.

The new auxiliary vessels have been fabricated and installed on the support structure (framework). These vessels include the new large storage/recirculation tank for the large columns, a similar tank for the small columns, and a large capacity scrubber which will be used to clean the outlet gas from the large columns. Modifications of the existing units to accommodate liquid flow and solids were also completed during this period. All vessels have been mounted on the framework. The slurry sample valves and pump have been installed for use with the small stainless steel column.

We have located suppliers of iron oxide and silica particles in the range 0 to 5 μm. We have also located suppliers of iron oxide and silica particles with the majority of particles greater than 53 μm. We are having difficulty finding a company that is willing to grind the particles and give us a narrow sized distribution (45 – 53 μm). For experiments in the small columns, the amount of solids needed in the range 45 – 53 μm is fairly small ($\approx$ 16 pounds of both iron oxide and silica); therefore, we plan to sieve the particles ourselves. However, for experiments in the large columns a significant amount of solids is required ($\approx$ 250 lbs of both iron oxide and silica), and we will have to find a company that will classify particles for us.

C. Task 4 – Application of a Gamma Radiation Density Gauge for Determining Hydrodynamic Parameters

During this quarter, construction of the stainless steel calibration chamber and the vessels which will be used to store the radioactive sources when they are not in use has been initiated. These units as well as the plexiglass calibration chamber will be completed during the next quarter.

We examined other possible source combinations which may be applicable to our system. We wrote a program which would allow us to enter a set of absorption coefficients for a set of sources and then examine the effect of errors in the absorption coefficients on the predicted fractions of each phase (i.e. gas, liquid, and solid). Also, the program allowed us to study the effect of errors in the count rate on the predicted fractions. The various source combinations we have examined are (1) Cesium–137, Americium–241, (2) Cobalt–60, Cobalt–57, (3) Cobalt–60, Barium–133, and (4) Cobalt–60, Cesium–137. The Cesium–Americium system was the most stable. However, we may not be able to achieve an adequate count rate with the Americium source. Since we received a Cesium–137 and an Americium–241 source from SAIC via Catalytic, Inc., we plan to try this source combination before purchasing an alternate set of sources. Of all the alternative sources examined, the system, Cobalt–60 – Cesium–137 appears to be the most promising. Both of these sources are fairly inexpensive and will provide adequate count rates.

During this quarter, James Daly visited Dr. Gidaspow at Illinois Institute of Technology and Dr. Alan Weimer at Dow Chemical Co. in Midland Michigan. The purpose of his visit was to examine their movable assembly mechanisms, which are used to transport the nuclear density gauges along the column, to aid in the design of our assembly. During the next quarter, we plan to purchase the material needed to construct the mechanism and begin fabrication of the unit.

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Kuo, J.C.W., etal. Two–stage process for conversion of synthesis gas to high quality transportation fuels, Final Report of the Department of Energy for Contract No. DE–AC22–83PC60019 (1985)

APPENDIX I

Experimental Design

In this section of the Project Work Plan, we will present the set of conditions we plan to vary throughout the course of our investigations, as well as, the set of experiments we plan to conduct. The purpose of our research is to study the effect of bubble column geometry, distributor type, liquid velocity, solids type, solids concentration, solids size, liquid medium, temperature, and gas flow rate on the hydrodynamic properties associated with Fischer–Tropsch slurry bubble column reactors.

Bubble columns with diameters of 0.051 m and 0.229 m and a height of 3 m will be employed in our studies. Most of the experiments will be conducted in the larger diameter column, where the commercially viable churn–turbulent flow regime can be achieved. The small column will be used to conduct initial experiments to ensure that all equipment is functioning properly. Also, the results from the small column experiments will be compared to those in the large column to study the effect of column diameter and flow regimes on the hydrodynamic parameters.

We plan to use a perforated plate (19 holes 2 mm in diameter) ,as well as a bubble cap distributor in the large columns. Perforated plate and bubble cap distributors are typically employed in slurry reactors. In the small columns, we intend to use a 2 mm single hole orifice plate distributor. This distributor will have approximately the same jet velocity as the perforated plate in the large column.

In order to simulate the return of concentrated slurry from a catalyst/wax separator, a small upward liquid flow will be employed in some of our experiments. We plan to use two liquid velocities. The lower liquid velocity will be 0.005 m/s. This velocity is comparable to that used by Mobil (Kuo et al., 1985) in their bubble column reactor. Also, a superficial liquid velocity of 0.02 m/s will be employed.

Experiments will be conducted with two different types of solids, iron oxide and silica. Iron oxide particles were chosen because their physical properties are similar to those of precipitated iron catalyst particles that are used in Fischer–Tropsch reactors. The silica particles will be used to study the effect of solids density on the hydrodynamic parameters.

The effect of solids loading on hydrodynamic parameters will be studied in the range 0 – 30 wt %, which encompasses the range of catalyst concentrations used in slurry bubble column reactors for Fischer–Tropsch synthesis (Deckwer et al., 1982). The majority of our experiments will be conducted using a solids concentration of 20 wt %; however, some experiments will be conducted using solids concentrations of 10 and 30 wt %.

Narrow–sized solid fractions will be used with particles less than 53 μm. The majority of our experiments will be done with particles 0 – 5 μm; however, in order to study the effect of particle size, we also intend to conduct some experiments with particles 45 – 53 μm. The latter particle size range probably represents an upper limit for this particular application.

We plan to conduct the majority of our experiments with the SASOL reactor wax if available. Some experiments will be conducted using the same type of Fischer–Tropsch derived paraffinic wax (FT–300) that was used in our previous DOE contract.

We plan to use a total of five superficial gas velocities per given set of conditions. These velocities are 0.02 (or 0.03), 0.04, 0.06, 0.09, and 0.12 m/s. For experiments in the large column, in addition to these velocities, a gas velocity of 0.15 m/s will be employed in some runs. The minimum velocity (0.02 or 0.03 m/s) will be determined experimentally based on the extent of solids settling which occurs. If too much settling occurs at 0.02 m/s, then 0.03 m/s will be used as our minimum gas velocity.

Table 1 is a summary of the experimental parameters and range of conditions that will be employed throughout the course of our experiments. Initially, we examined various statistical designs, but encountered difficulties due to the variable levels and non–continuous variables. Thus, in the final selection of experiments, we used a factorial design and then selected the experiments which would enable us to determine the effect of the experimental variables on the hydrodynamic parameters (i.e. gas hold–up, flow regimes and flow regime transitions, etc.). We have designed a set of experiments for each set of columns (small and large). A total of 91 experiments are planned. The experiments proposed for the small columns are shown in Table 2. A total of 41 experiments will be conducted in the small columns. We will periodically repeat some conditions to check for reproducibility of results. The last column in Tables 2 and 3 correspond to the batch of wax which will be used. The letter "F" denotes runs with FT–300, while the letter "S" denotes runs with SASOL. For experiments in the small columns, we do not plan to use a given batch of wax for more than seven experiments (batch F–2).

Table 3 is a list of the experiments we plan to conduct in the large columns. As with the experiments in the small columns, some of the experiments will be repeated periodically to check for reproducibility. For experiments in the large columns, we do not plan to conduct more than eight runs with a given batch of wax (batch F–2 and S–4). All of the experiments listed in Table 3 will be conducted using conventional techniques to obtain axial gas hold–ups and axial solids concentrations. Experiments with the nuclear density gauge will be conducted simultaneously with some of the experiments proposed in Table 3. If the nuclear density gauge is not operational when we begin experiments in the large columns, some experiments may have to be repeated using the nuclear density gauge technique to determine the volume fractions of the gas, liquid, and solids at various positions in the column.

Table 1. Variables and levels of experiments to be performed

Variable	Levels			
	1	2	3	4
Column ID (m)	0.051	0.229		
Distributor	Perf. Plate	Bubble Cap		
Liquid Vel (m/s)	0.00	0.005	0.02	
Solids Type	Iron Oxide	Silica		
Solids Size (μm)	0 - 5	45 -53		
Solid Conc. (wt %)	0.0	10.0	20.0	30.0
Liquid Type	FT-300	SASOL		
Temperature (°C)	200	265		

TABLE 2. SUMMARY OF RUNS PLANNED IN THE SMALL COLUMNS

EXP NO.	WAX TYPE	T (C)	dp (um)	Wc (%)	SOLIDS TYPE	Ul (m/s)	BATCH NO.
BC	SASOL	265	0 – 5	20	IRON OX	0.005	———
1	FT-300	265	———	———	———	0 S	F-1
2	FT-300	265	———	———	———	0.005	F-1
3	FT-300	265	———	———	———	0.02	F-1
4	FT-300	265	0 – 5	10	IRON OX	0.005	F-2
5	FT-300	265	0 – 5	20	IRON OX	0 S	F-2
6	FT-300	265	0 – 5	20	IRON OX	0.005	F-2
7	FT-300	200	0 – 5	20	IRON OX	0.005	F-2
8	FT-300	265	0 – 5	20	IRON OX	0.02	F-2
9	FT-300	265	0 – 5	30	IRON OX	0.005	F-2
10	FT-300	265	0 – 5	10	SILICA	0.005	F-3
11	FT-300	265	0 – 5	20	SILICA	0.005	F-3
12	FT-300	265	45 – 53	20	IRON OX	0 S	F-4
13	FT-300	265	45 – 53	20	IRON OX	0.005	F-4
14	FT-300	265	45 – 53	30	IRON OX	0.005	F-4
15	FT-300	265	45 – 53	20	SILICA	0.005	F-5
16	FT-300	265	45 – 53	30	SILICA	0.005	F-5
17	SASOL	265	———	———	———	0 S	S-1
18	SASOL	265	———	———	———	0.005	S-1
19	SASOL	265	———	———	———	0.02	S-1
20	SASOL	265	0 – 5	10	IRON OX	0 S	S-2
21	SASOL	265	0 – 5	10	IRON OX	0.005	S-2
22	SASOL	265	0 – 5	10	IRON OX	0.02	S-2
23	SASOL	265	0 – 5	20	IRON OX	0 S	S-2
24	SASOL	265	0 – 5	20	IRON OX	0.005	S-2
25	SASOL	265	0 – 5	20	IRON OX	0.02	S-2
26	SASOL	265	0 – 5	30	IRON OX	0 S	S-2
27	SASOL	265	0 – 5	30	IRON OX	0.005	S-2
28	SASOL	265	0 – 5	30	IRON OX	0.02	S-2
29	SASOL	265	0 – 5	10	SILICA	0.005	S-3
30	SASOL	265	0 – 5	20	SILICA	0 S	S-3
31	SASOL	265	0 – 5	20	SILICA	0.005	S-3
32	SASOL	265	0 – 5	20	SILICA	0.02	S-3
33	SASOL	265	45 – 53	10	IRON OX	0.005	S-4
34	SASOL	265	45 – 53	10	IRON OX	0.02	S-4
35	SASOL	265	45 – 53	20	IRON OX	0 S	S-4
36	SASOL	265	45 – 53	20	IRON OX	0.005	S-4
37	SASOL	265	45 – 53	20	IRON OX	0.02	S-4
38	SASOL	265	45 – 53	30	IRON OX	0.005	S-4
39	SASOL	265	45 – 53	30	IRON OX	0.02	S-4
40	SASOL	265	45 – 53	20	SILICA	0.005	S-5
41	SASOL	265	45 – 53	30	SILICA	0.005	S-5

TABLE 3. SUMMARY OF RUNS PLANNED IN THE LARGE COLUMNS

EXP NO.	WAX TYPE	T (C)	dp (um)	Wc (%)	SOLIDS TYPE	Ul (m/s)	DIST TYPE	BATCH NO.	
BC	SASOL	265	0 - 5	20	IRON OX	0.005	PP	———	
1	FT-300	265	———	———	———	0 S	PP	F-1	
2	FT-300	265	———	———	———	0.005	PP	F-1	
3	FT-300	265	———	———	———	0.02	PP	F-1	
4	FT-300	265	———	———	———	0.005	BC	F-1	1
5	FT-300	265	0 - 5	10	IRON OX	0.005	PP	F-2	
6	FT-300	265	0 - 5	20	IRON OX	0 S	PP	F-2	
7	FT-300	265	0 - 5	20	IRON OX	0.005	PP	F-2	
8	FT-300	200	0 - 5	20	IRON OX	0.005	PP	F-2	
9	FT-300	265	0 - 5	20	IRON OX	0.02	PP	F-2	
10	FT-300	265	0 - 5	20	IRON OX	0.005	BC	F-2	2
11	FT-300	265	0 - 5	30	IRON OX	0.005	PP	F-2	
12	FT-300	265	0 - 5	10	SILICA	0.005	PP	F-3	
13	FT-300	265	0 - 5	20	SILICA	0.005	PP	F-3	
14	FT-300	265	45 - 53	20	IRON OX	0 S	PP	F-4	
15	FT-300	265	45 - 53	20	IRON OX	0.005	PP	F-4	
16	FT-300	265	45 - 53	20	IRON OX	0.005	BC	F-4	3
17	FT-300	265	45 - 53	30	IRON OX	0.005	PP	F-4	
18	FT-300	265	45 - 53	20	SILICA	0.005	PP	F-5	
19	FT-300	265	45 - 53	30	SILICA	0.005	PP	F-5	
20	SASOL	265	———	———	———	0 S	PP	S-1	
21	SASOL	265	———	———	———	0.005	PP	S-1	
22	SASOL	265	———	———	———	0.02	PP	S-1	
23	SASOL	265	———	———	———	0.005	BC	S-1	4
24	SASOL	265	0 - 5	10	IRON OX	0 S	PP	S-1	
25	SASOL	265	0 - 5	10	IRON OX	0.005	PP	S-1	
26	SASOL	265	0 - 5	10	IRON OX	0.02	PP	S-1	
27	SASOL	265	0 - 5	10	IRON OX	0.005	BC	S-1	5
28	SASOL	265	0 - 5	20	IRON OX	0 S	PP	S-2	
29	SASOL	265	0 - 5	20	IRON OX	0.005	PP	S-2	
30	SASOL	265	0 - 5	20	IRON OX	0.02	PP	S-2	
31	SASOL	265	0 - 5	20	IRON OX	0 S	BC	S-2	6
32	SASOL	265	0 - 5	20	IRON OX	0.005	BC	S-2	7
33	SASOL	265	0 - 5	30	IRON OX	0 S	PP	S-2	
34	SASOL	265	0 - 5	30	IRON OX	0.005	PP	S-2	
35	SASOL	265	0 - 5	30	IRON OX	0.02	PP	S-2	
36	SASOL	265	0 - 5	10	SILICA	0.005	PP	S-3	
37	SASOL	265	0 - 5	20	SILICA	0 S	PP	S-3	
38	SASOL	265	0 - 5	20	SILICA	0.005	PP	S-3	
39	SASOL	265	0 - 5	20	SILICA	0.02	PP	S-3	
40	SASOL	265	45 - 53	10	IRON OX	0.005	PP	S-4	
41	SASOL	265	45 - 53	10	IRON OX	0.02	PP	S-4	
42	SASOL	265	45 - 53	20	IRON OX	0 S	PP	S-4	
43	SASOL	265	45 - 53	20	IRON OX	0.005	PP	S-4	
44	SASOL	265	45 - 53	20	IRON OX	0.02	PP	S-4	
45	SASOL	265	45 - 53	20	IRON OX	0.005	BC	S-4	8
46	SASOL	265	45 - 53	30	IRON OX	0 S	PP	S-4	
47	SASOL	265	45 - 53	30	IRON OX	0.005	PP	S-4	
48	SASOL	265	45 - 53	30	IRON OX	0.02	PP	S-4	
49	SASOL	265	45 - 53	20	SILICA	0.005	PP	S-5	
50	SASOL	265	45 - 53	30	SILICA	0.005	PP	S-5	

DOE/PC/90012-1

Hydrodynamics of Three-Phase Slurry Fischer-Tropsch Bubble Column Reactors

Quarterly Technical Progress Report
for the Period 1 June 1987 - 30 September 1987

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October 23, 1987

Prepared for the United States Department
of Energy Under Contract No. DE-AC22-86PC90012

NOTICE

PATENT STATUS

TECHNICAL STATUS

TABLE OF CONTENTS

Tables and Figures

I. Abstract

The design of the experimental apparatus for studying three–phase slurry Fischer–Tropsch bubble column reactors, has been completed. The apparatus used during the previous DOE project (DE–AC22–84PC70027) has been dismantled and the support structure modified to accommodate the new/modified apparatus. The design of the new auxiliary vessels have been completed and drawings prepared. Drawings for the modifications of the existing vessels have also been prepared. Fabrication of these units is in progress. Purchase orders for some of the new instrumentation have been issued.

Literature review of the application of nuclear density gauges in two and three–phase studies has been initiated. Design of the nuclear density gauge apparatus is in progress.

II. Objective and Scope of Work

The overall objective of this contract is to determine the effects of bubble column diameter, solids loading and particle size, and operating conditions (temperature, gas and liquid flow rates) on hydrodynamics of slurry bubble columns for Fischer–Tropsch synthesis, using a molten wax as the liquid medium. To accomplish these objectives, the following specific tasks will be undertaken.

Task 1 – Project Work Plan

The objective of this task is to establish a detailed project work plan covering the entire period of performance of the contract, including a detailed program schedule, analytical procedures, and estimated costs and manhours expended by month for each task.

Task 2 – Design and Construction of the Experimental Apparatus

The existing glass and stainless steel columns (0.051 m and 0.229 m in diameter, 3 m tall) that were constructed under our previous DOE contract (DE–AC22–84PC70027), will be modified and additions made in order to study the effect of continuous upward liquid flow. After the procurement of equipment and instrumentation, and construction of the unit is completed, a shakedown of test facilities will be made to verify achievement of planned operating conditions.

Task 3 – Measurement of Hydrodynamic Parameters by Conventional Techniques

In this task, the effects of operating conditions (liquid and gas superficial velocities, temperature), gas distributor, column diameter, and solids concentrations and particle size on hydrodynamic parameters in the glass and stainless steel columns will be determined. All experiments will be conducted using nitrogen at atmospheric pressure.

The hydrodynamic parameters that will be determined as a function of the independent variables mentioned above are: average gas hold–up, axial solids distribution, axial gas hold–up, flow regime characterization, and qualitative information on bubble size distribution.

Task 4 – Application of a Gamma Radiation Density Gauge for Determining Hydrodynamic Parameters

The objective of this task is to determine hydrodynamic parameters for the three–phase system using a nuclear density gauge apparatus. A movable assembly mechanism and positioning racks for the two nuclear density gauges and detectors will be designed and constructed. Following the interfacing of the apparatus with an on–line microprocessor, the gauges will be calibrated using pure components (liquid wax and solid particles), and with known proportions of liquid and solid. After calibration, the following parameters will be obtained from experiments in the large stainless steel column: axial gas hold–up, axial concentration of solids, and qualitative information on flow regimes and bubble size distributions.

III. Summary of Progress

- The Project Work Plan (Task 1) has been submitted to the DOE earlier in this quarter.
- The structure used to support the apparatus during our previous project (DE–AC22–84PC70027) has been modified to accommodate equipment that will be used for the three–phase project.
- Drawing for the bubble column modifications and for the various auxiliary vessels have been completed. The fabrication of these items is in progress.
- Purchase orders for some instruments have been issued.
- We received the nuclear density gauge apparatus (sources, detectors, and accompanying electronics) from DOE via Catalytic, Inc.
- The computer has been cleaned and "boots–up".
- Drawings for the calibration units and storage chambers have been completed.
- The appplication of the License for the Use of Radioactive Materials has been completed.
- Alternative source combinations are being examined.
- Consultation with manufacturers about the movable assembly mechanism has been initiated.

IV. Detailed Description of Technical Progress

A. Task 1 – Project Work Plan

The Project Work Plan was submitted to the DOE in July, 1987.

B. Task 2 – Design and Construction of the Experimental Equipment

The support structure (framework) used to hold the bubble columns and auxiliary vessels during our previous contract (DE–AC22–84PC70027) was completely dismantled and rebuilt. This was done to accommodate the modified apparatus, and also to provide more work space around the individual columns. The electrical wiring for distributing power to the various units (primarily for heating) is nearing completion.

The new auxiliary vessels have been designed and include a large wax storage / recirculation tank for the large columns, a similar tank for the smaller diameter columns, and a large capacity scrubber which will be used to clean the outlet gas from the large columns. The preliminary detailed drawings for these three units have been completed. Figs. 1a and 1b show detailed drawings of the large storage/recirculation tank and its cover plate, respectively. The smaller tank is similar to that shown in Figs. 1a and 1b. The material necessary for the construction of these units has been ordered and received. Fabrication of the vessels is in progress.

In addition to the new vessels, modifications of the existing units are also being made. The expansion units above the two stainless steel columns will be modified to allow for the recirculation of the slurry. Drawings for the two expansion units are shown in Figs. 2 and 3. Fabrication of the two units is in progress.

Technical information on various instruments has been obtained and orders for several of these items have been placed. The stirrers for the two circulation tanks, slurry sampling valves (capable of high temperature operation) and orifice plates, for liquid flow rate measurement, have been ordered. New heating tapes, beaded heaters and voltage regulators have been ordered and received.

We have experienced difficulties in locating suppliers of pressure transducers and slurry pumps capable of high temperature operation in the presence of solid particles. Most of the transducers are capable of operating at temperatures upto 150 °C; whereas, we need transducers that can operate at temperatures in the range 200 – 265 °C. The low temperature transducers cannot be mounted directly on the column wall, and will therefore need a gas purge line to create a buffer between the slurry and the transducer sensing element, or a long tube (5 – 10 cms long) separating the transducer from the column wall. Such a tube would provide a sufficient temperature gradient to allow the operation of the transducer in the vicinity of 150 °C. Nevertheless, we will continue to try to locate transducers capable of high temperature operation, since a transducer mounted directly on the column wall would give a more accurate reading of wall pressure fluctuations than would a transducer with a gas purge or with an extended connecting tube.

We have made extensive inquiries for pumps capable of high temperature operation in the presence of solid particles but have had limited success thus far. Because of the presence of solids and the possibility of tiny gas bubbles entrained in the recirculating liquid, a centrifugal pump cannot be used in our application. With such a pump, cavitation could be a serious problem. We are therefore limited to positive displacement pumps. There are metering pumps available that can be used for our application; however, they are substantially more expensive than the budgeted amount of $9000. Other positive displacement pumps (e.g. gear pumps, lobe pumps, etc.) are not available for handling slurries at high temperatures, particularly because of the low viscosity of our slurry ($<$ 15 mPa.s). The majority of these pumps are rated at maximum temperatures ranging from 150 – 200 °C. Our calculations show that the heating of the slurry from 200 °C (at the pump outlet) to 265 °C (at the bubble column inlet) are very high during the continuous mode of operation. Therefore, we will not be able to use a pump that is rated below 200 °C. We are currently looking into specially designed pumps and expect to locate a suitable pump during the next quarter.

C. Task 4 – Application of a Gamma Radiation Density Gauge for Determining Hydrodynamic Parameters

Table 1 is a list of the equipment received from DOE via Catalytic, Inc. The radioactive sources were shipped separately and arrived on June 10, 1987 at the Texas A&M Radiological Safety Office. The rest of the equipment was received by July 6, 1987 at Zachry Engineering Center, Rm 327.

The DEC PDP 11/23S computer system would not "boot–up" when we first received it; however, after cleaning the circuit boards inside the computer, it appears to be functioning properly. We did not receive the operating system for the computer or the fortran compiler. These will have to be purchased.

The design of the calibration chambers which will be used to determine the attenuation/absorption coefficients and the incident intensities or count rates has been completed. Two calibration chambers will be used and are shown in Figures 4, 5 and 6. One calibration chamber will be constructed from plexiglass (see Figures 4 and 5), and will be used for our initial tests (i.e. cold system). The plexiglass chamber will be approximately 12 in by 12 in by 24 in tall and have 10 sections. The sections are divided in such a way to enable us to insert known volume fractions of the gas, liquid, and solid phases in the range we expect to observe in the column under the proposed operating conditions. The gas and solid phases will be the same as what we intend to use in the column; whereas, the liquid phase will be a hydrocarbon with approximately the same density as wax that is liquid at room temperature. This will enable us to determine to some extent the accuracy with which we will be able to determine the fractions of the various phases. The stainless steel chamber (see Figure 6) will be used to obtain the attenuation coefficients for the wax and solids. This chamber will be constructed from a 24 in section of stainless steel pipe (8 in nominal diameter, schedule 5). The attenuation coefficients for the solids will be compared to those obtained in the plexiglass chamber.

The units that will be used to store the sources when they are not in use have been designed and a schematic of the storage unit for the high energy source is shown in Figure 7. The storage unit for the low energy source is similar to the one for the high energy source. The chambers will be constructed from a stainless steel pipe filled with lead. The sources will be placed inside the chamber when not in use.

We have performed some initial calculations to determine if the sources we obtained from Catalytic, Inc. would be applicable to our system. It appears that the sources do not possess sufficient activity, and new sources will have to be selected. However, we plan to experiment with the sources from Catalytic, Inc. before purchasing new sources. We are in the process of writing a program that will be used to analyze various source combinations. The results from the program will give an indication of the accuracy with which we will be able to determine the various phase fractions for a given set of sources. Based on the results from the program, as well as those obtained from actual measurements with the sources from Catalytic, Inc., we will select our new sources.

We have completed the application for a license permitting the use of radioactive material and will submit it during the next quarter. The license will be for the use of the sources from Catalytic, Inc. and will be updated once we select our new sources.

We have received the information we need to begin designing the movable assembly mechanism and will begin the design of the mechanism during the next quarter.

Table 1. Summary of equipment received from DOE via Catalytic, Inc.

EQUIPMENT	MANU	QUAN.	SERIAL NO.	MODEL NO.
Am NaI Detector	Bicron	2	EF–509, NA	1.5XM.040/1.5B
Cs NaI Detector	Bicron	2	EF–292, NA	1.5M1.5/1.5P
Detector Tube Base	Tennelec	4	240, 249,341, 342	154
Detector Heater Blanket	MINCO	2	NA	HK6070–09B–97.2
Temperature Switch	Elmwood	2	NA	3150-1
NIM BIM	Tennelec	1	428	MB2
NIM BIM Power Supply	Tennelec	1	505	TC 910
High Voltage Supply	Tennelec	2	794, 795	TC 948
Amplifier	Tennelec	2	2142, 2144	TC 248
Discriminator	Tennelec	2	314, 316	TC 453
Ratemeter	Tennelec	2	398, 404	TC 526

NA: Not Available

Table 1. (cont.) Summary of equipment received from DOE via Catalytic, Inc.

EQUIPMENT	MANU	QUAN.	SERIAL NO.	MODEL NO.
Computer	Digital Corp	1	BT01351	OH780 A PDP 11/23S
Terminal	Digital Corp	1	TAJ6555	
VT240	Digital Corp	1	NA	NA
Memory Board	Digital Corp	1	NA	NA
Scaler Board	ADAC Corp	1	NA	1604/OPI
A.D.C. Board	ADAC Corp	1	NA	1023EX
D.A.C. Board	ADAC Corp	NA	1412DA	
Disk Drive	Data Sys. Des.	1	3512 E	110/430
Interface Panel	Bud	1	NA	NA
Terminal Board	Belden	2	NA	NA
Am Radiation Source	Amersham Corp	1	NA	NA
Cs Radiation Source	Amersham Corp	1	NA	NA

Equipment Manuals

Software Manuals

Software

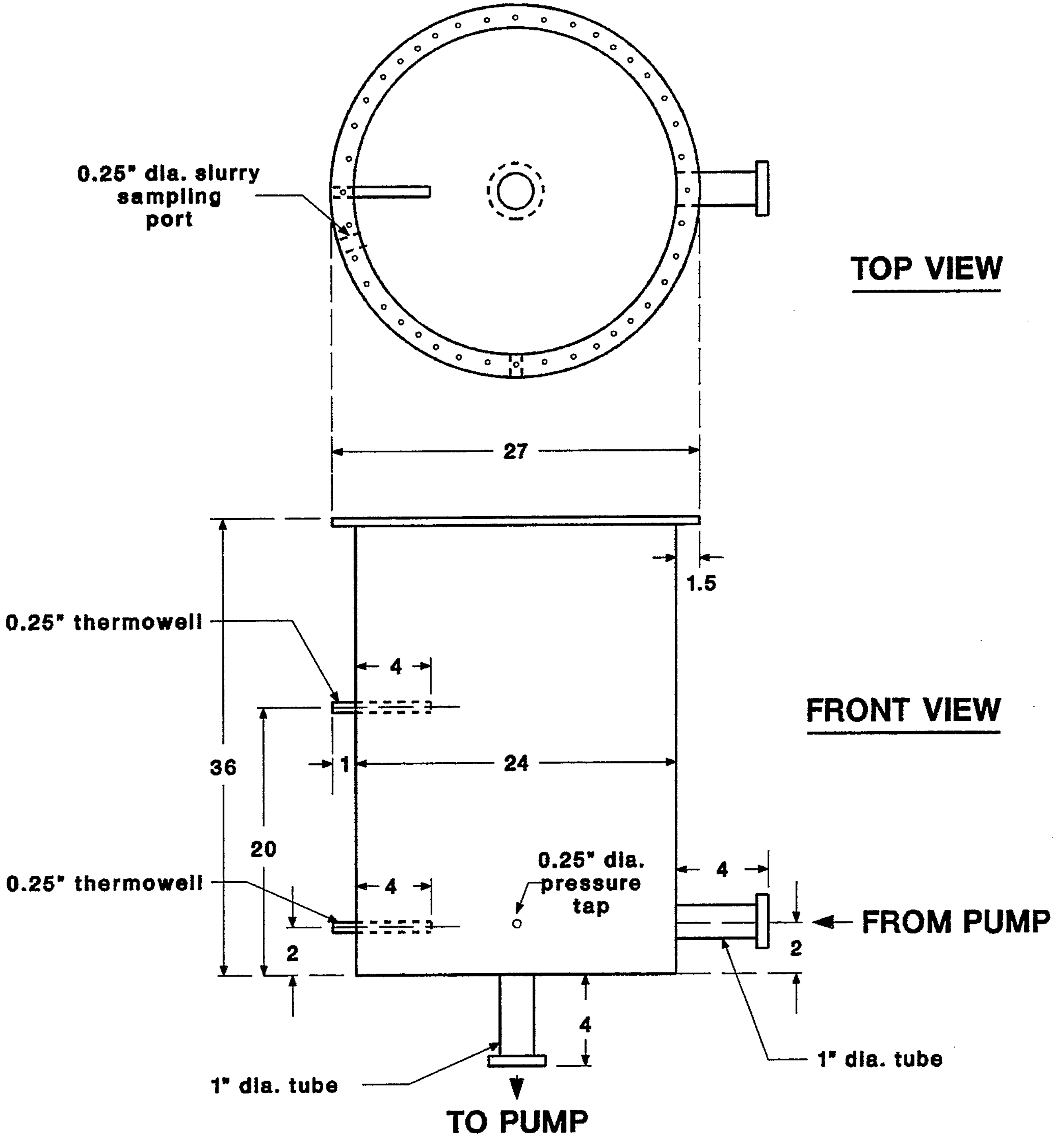
Blueprints

Spool Piece

Mounting rack for spool piece and phsae fraction meter

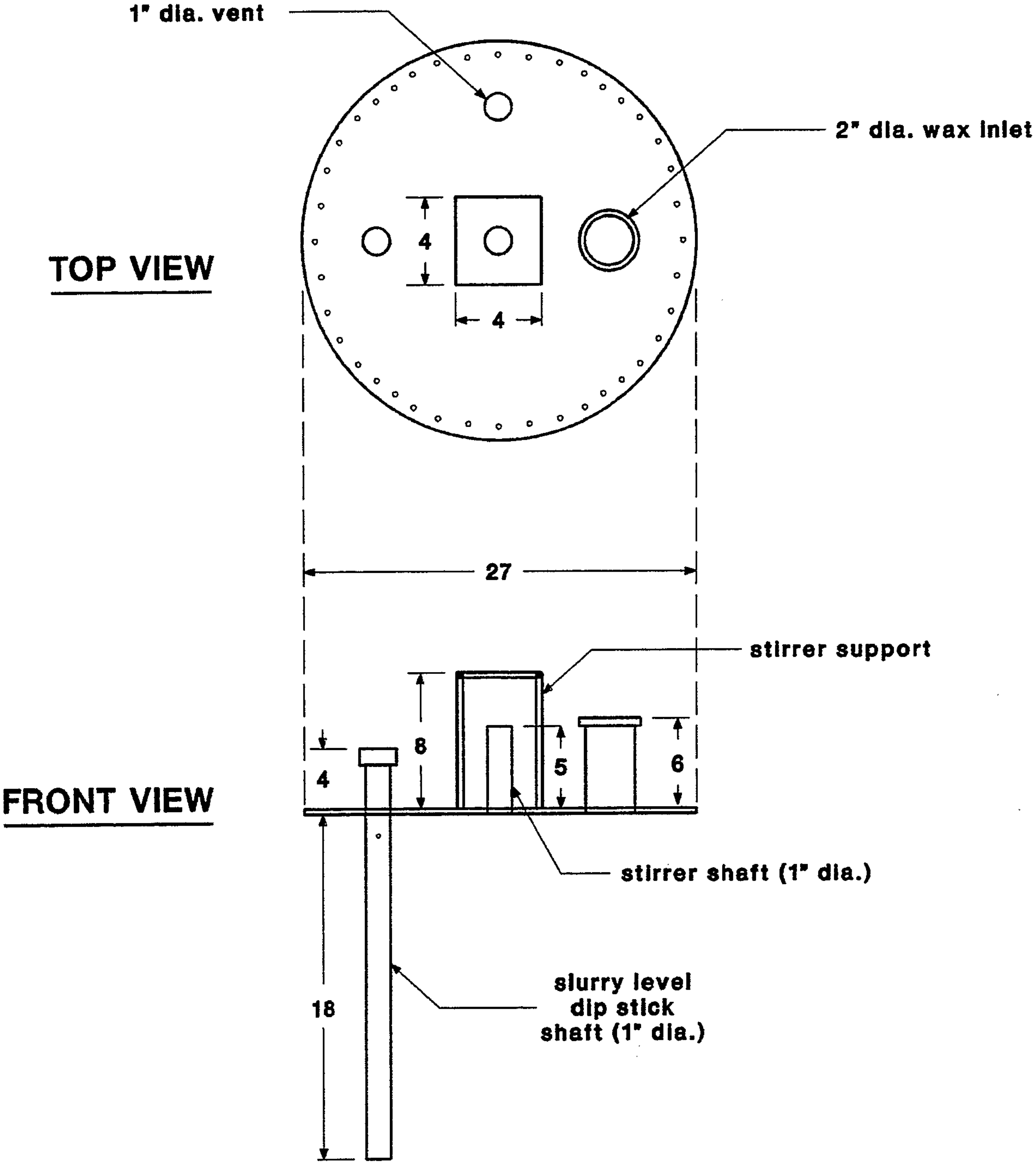
FIGURE 1a

LARGE WAX STORAGE/RECIRCULATION TANK



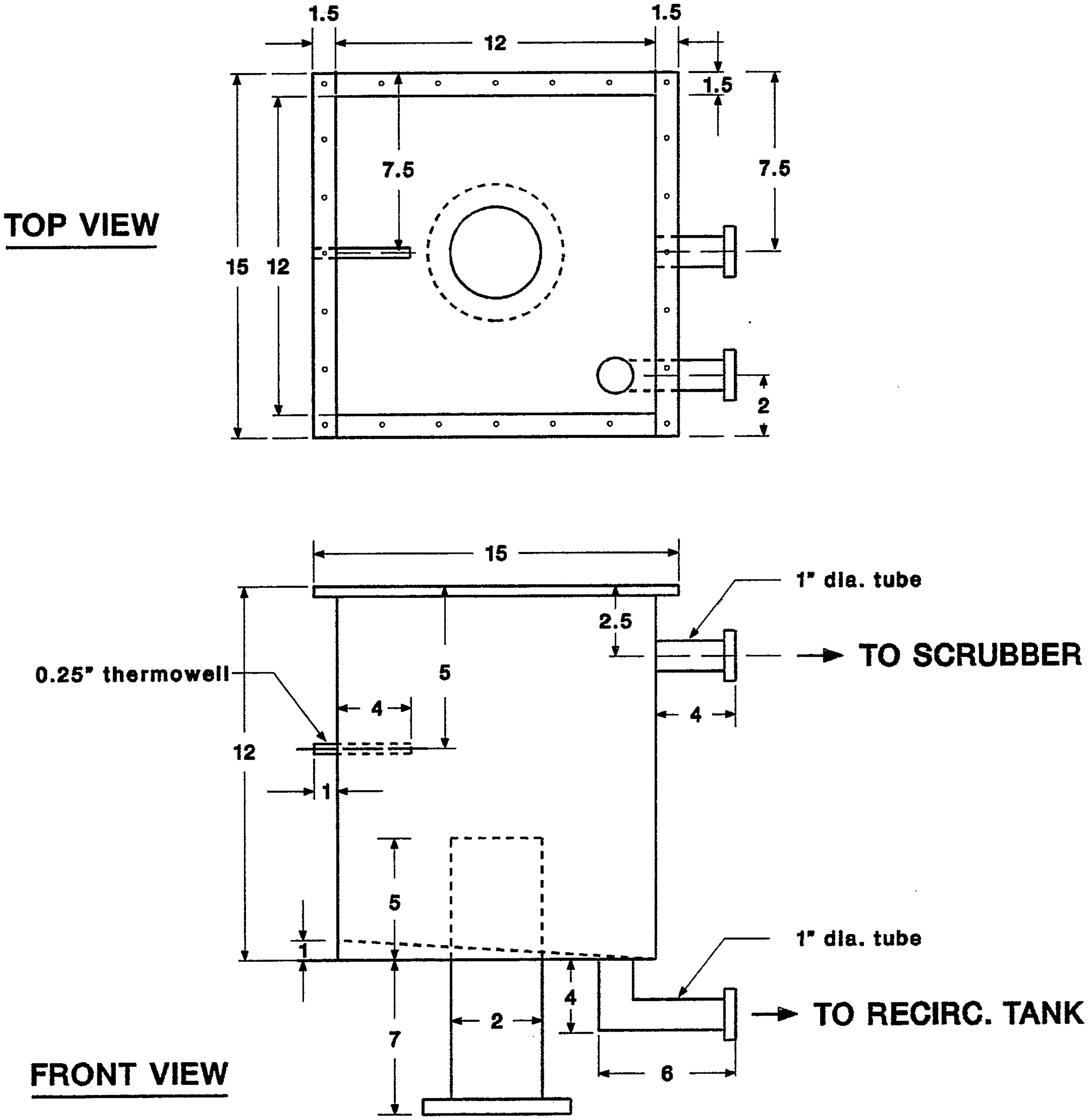
(All dimensions are in inches)

FIGURE 1b
COVER PLATE FOR LARGE STORAGE/RECIRC. TANK



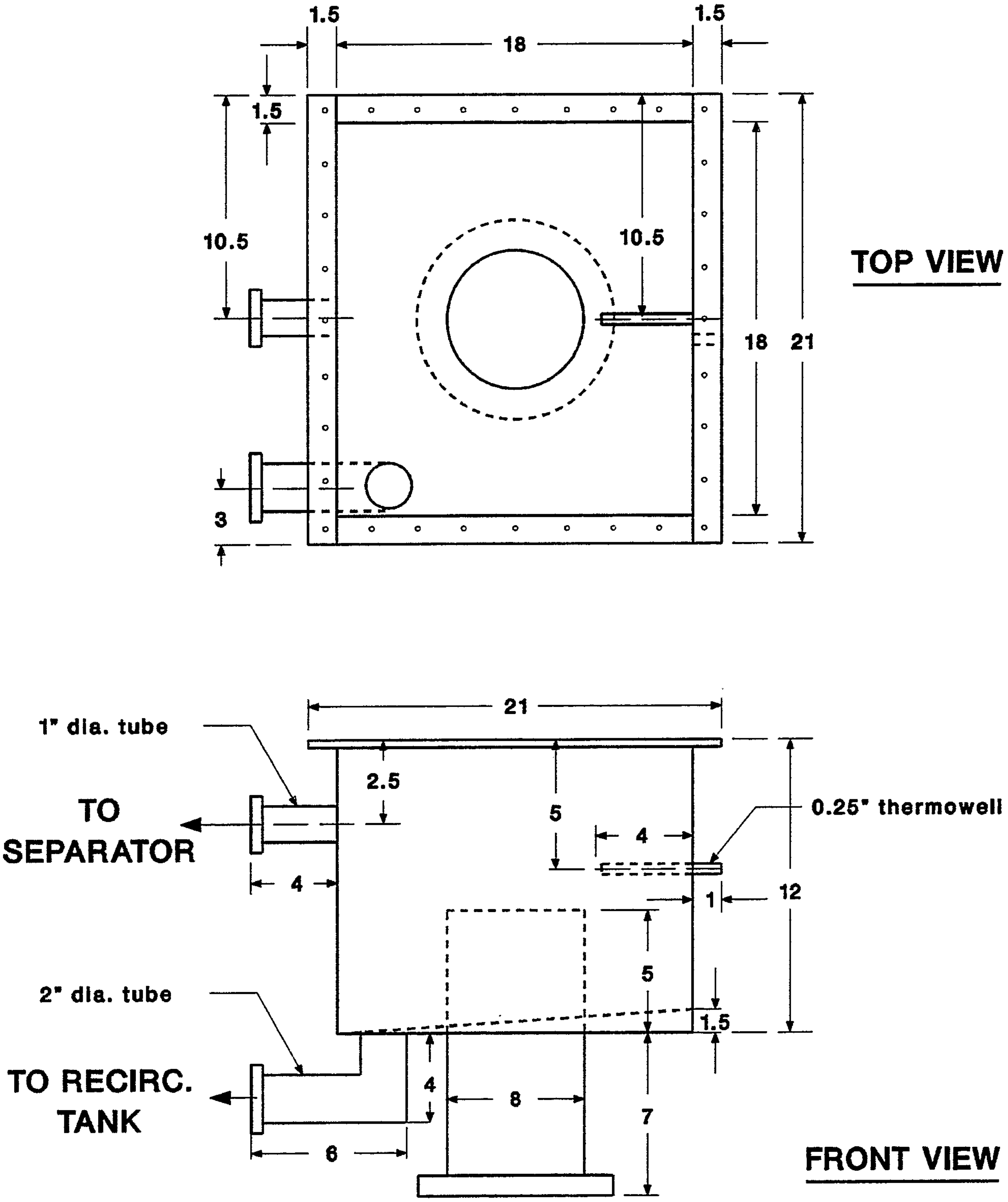
(All dimensions are in inches)

FIGURE 2
EXPANSION UNIT FOR SMALL SS COLUMN



(All dimensions are in inches)

FIGURE 3
EXPANSION UNIT FOR LARGE SS COLUMN



(All dimensions are in inches)

FIGURE 4. CALIBRATION CHAMBER - PLEXIGLASS

SIDE VIEW

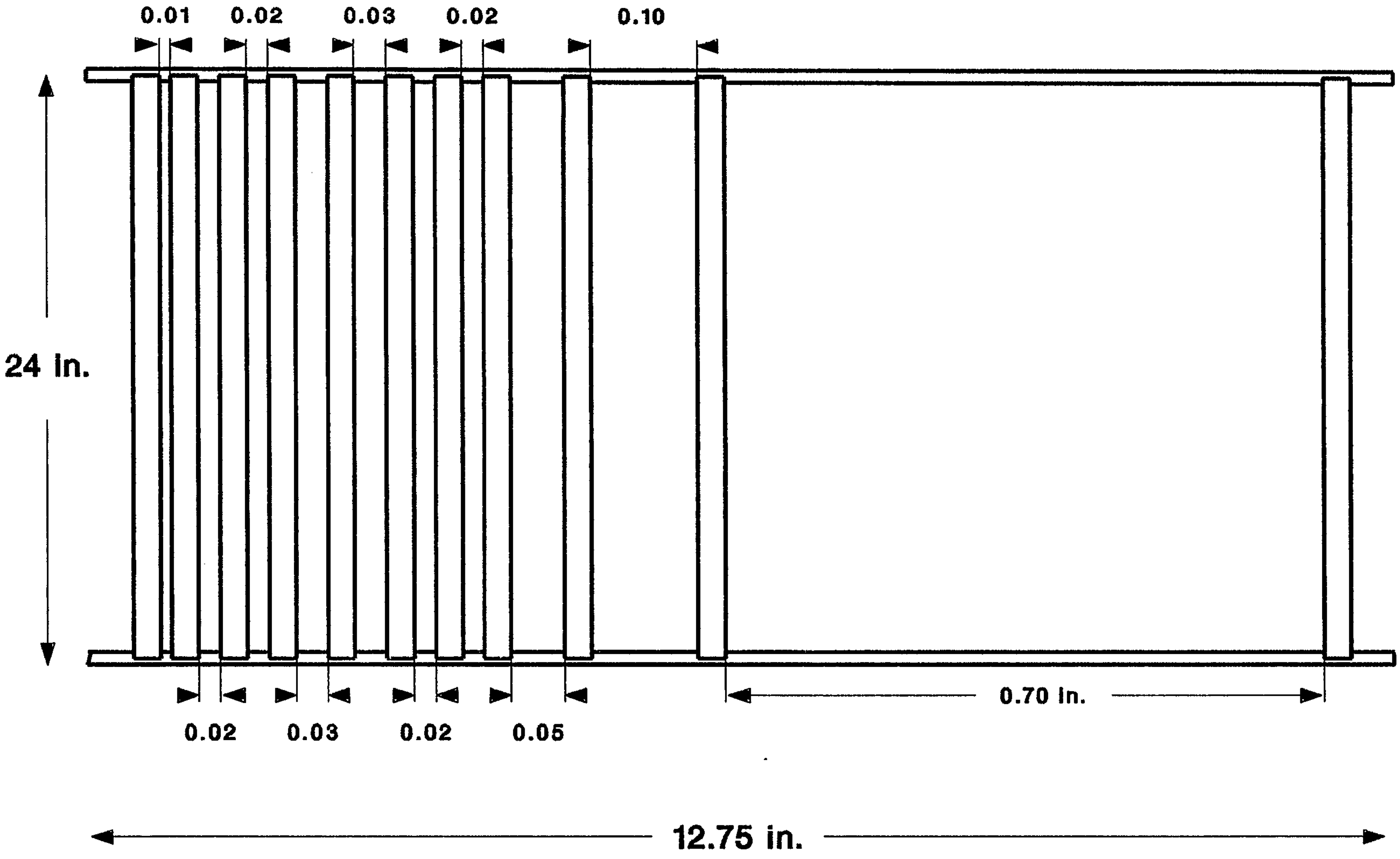


FIGURE 5. CALIBRATION CHAMBER - PLEXIGLASS
TOP VIEW

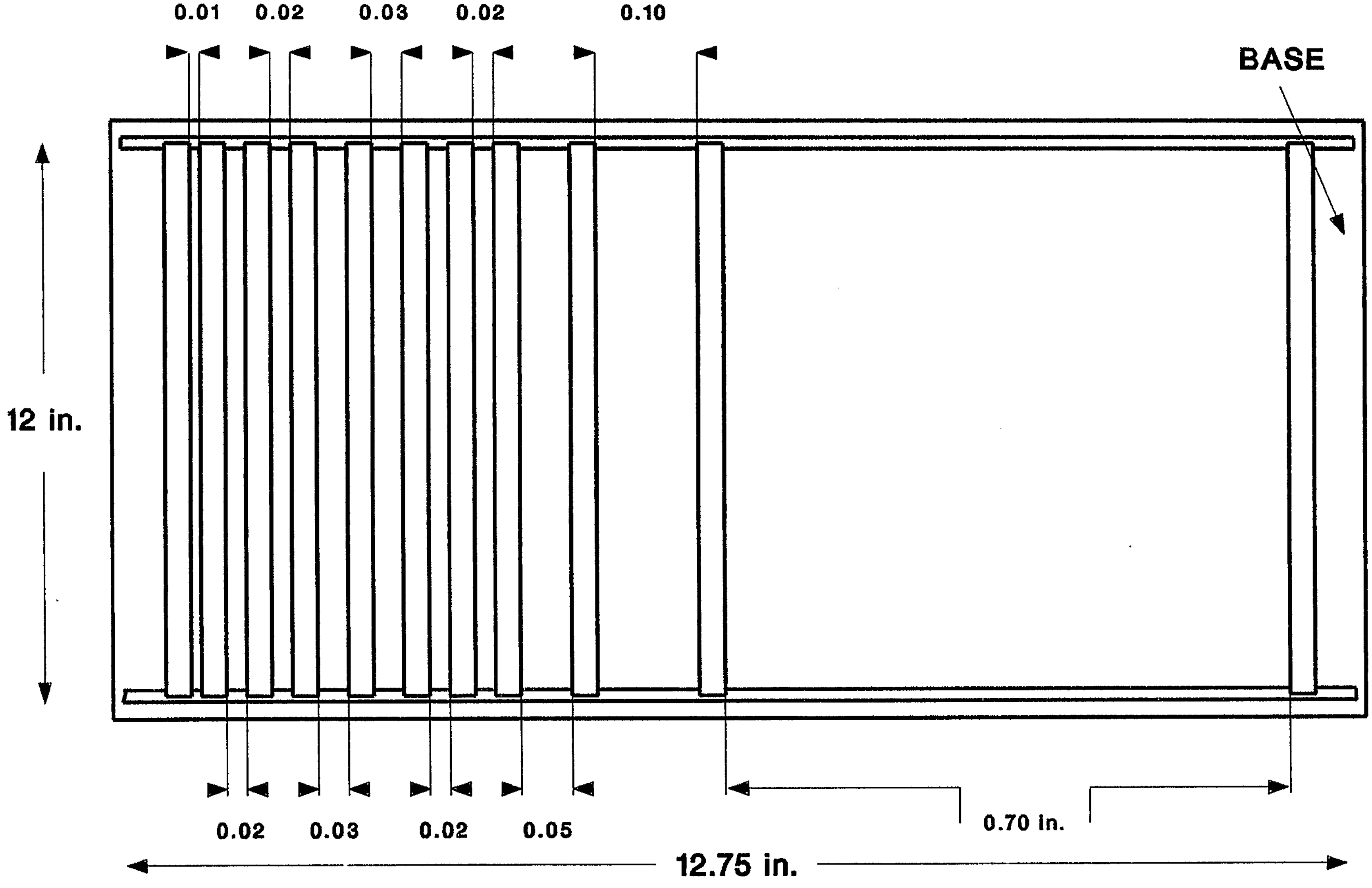


FIGURE 6. CALIBRATION CHAMBER - STAINLESS STEEL

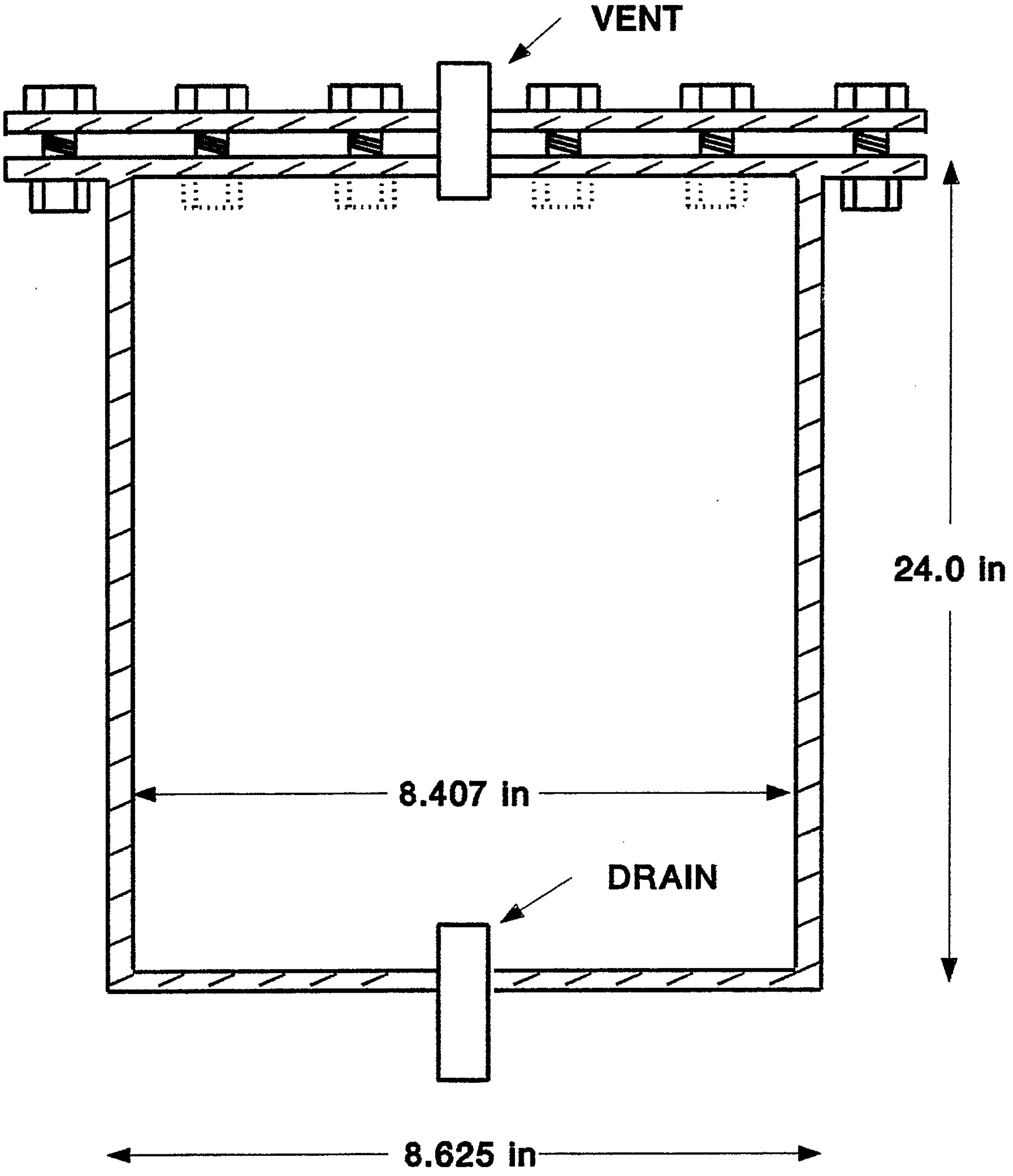


FIGURE 7. STORAGE CHAMBER FOR HIGH ENERGY SOURCE