STUDY OF EBULLATED BED FLUID DYNAMICS FOR H-COAL. MONTHLY PROGRESS REPORT NO. 2, OCTOBER 1--NOVEMBER 1, 1977

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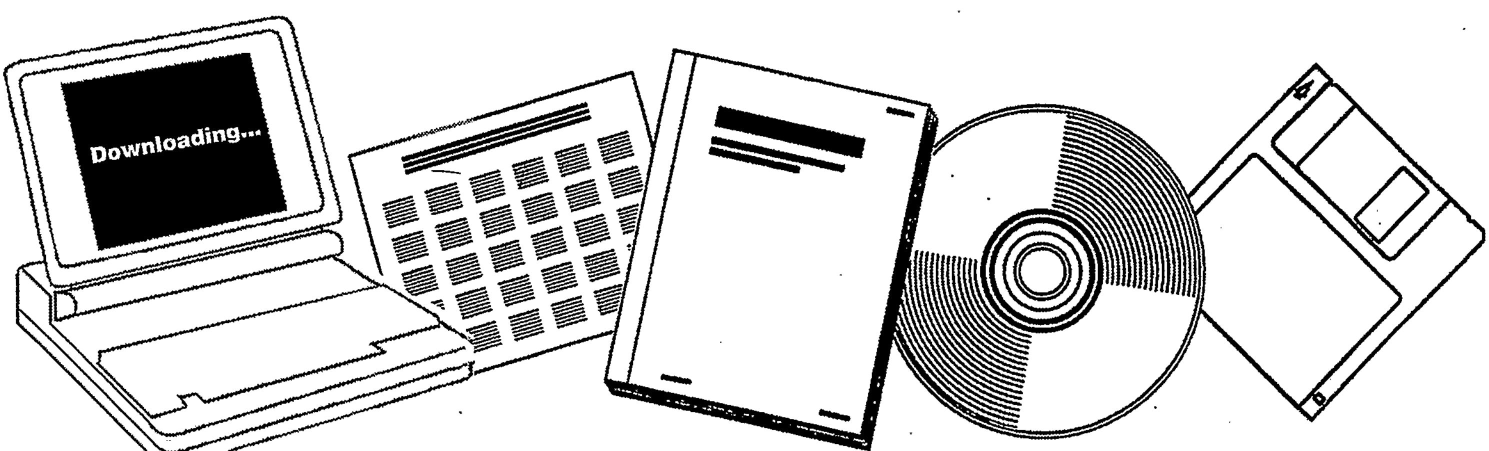

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STUDY OF EBULLATED BED FLUID DYNAMICS FOR H-COAL

MONTHLY PROGRESS REPORT NO. 2
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FOREWORD

The H-Coal process, developed by Hydrocarbon Research, Incorporated (HRI), involves the direct catalytic hydroliquefaction of coal to low-sulfur boiler fuel or synthetic crude oil. The 200-600 ton-per-day H-Coal pilot plant is being constructed next to the Ashland Oil, Incorporated refinery at Catlettsburg, Kentucky under ERDA contract to Ashland Synthetic Fuels, Incorporated. The H-Coal ebullated bed reactor contains at least four discrete components: gas, liquid, catalyst, and unconverted coal and ash. Because of the complexity created by these four components, it is desirable to understand the fluid dynamics of the system. The objective of this program is to establish the dependence of the ebullated bed fluid dynamics on process parameters. This will permit improved control of the ebullated bed reactor.

The work to be performed is divided into three parts: review of prior work; cold flow model construction and operation; and mathematical modelling. The objective of the second progress report is to outline progress in the first two parts.

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SUMMARY

Review of Prior Work

The literature search continued during this month. Major emphasis was placed on reviewing existing HRI data in conjunction with various theoretical models found in the literature. A significant part of this work was also dedicated to extending the understanding of the bubble behavior phenomena which may control the fluid dynamics of the H-Coal system. Experimental techniques useful in the data collection phase of this work were also identified.

Data Collection

Measurement of Physical Properties of H-Coal Liquids.--A visit to the HRI laboratories this month showed that currently there are no provisions to obtain samples from either the PDU reactor or the gas/liquid separator. These samples will be sent to Battelle for viscosity measurements. Further work is necessary to identify the requirements for carrying out these measurements.

Design of the Fluid Dynamics Unit.--The design of the fluid dynamics unit proceeded this month with major emphasis on the reactor vessel. The 6" reactor will be constructed from four 5'-long glass sections connected with stainless steel spool pieces and flanges. Details on the reactor recycle and distributor cup have been finalized with the assistance of HRI.

Progress was also made in various other aspects of the design of the unit. These include support structure, feed tanks, instrumentation, and computerization.

REVIEW OF PRIOR WORK

The status of the review of prior work is presented in Table I. Thus far, 110 papers have been identified as of value to the H-Coal fluid dynamics project. Of these, 77 have been reviewed. HRI reports are also being reviewed during this phase of work.

Liquid-Solid Fluidization

Most of the literature results in this area were reported in the September monthly progress report. During this month, a search of the literature on liquid-solid fluidization revealed one additional set of data for the bed expansion involving cylindrical particles. Blum and Toman (1) measured the expansion of cylinders of the following dimensions:

Diameter	Length
1/8 Inch	1/8 Inch
3/16 Inch	3/32 Inch
3/16 Inch	3/16 Inch

The fluid used in their study was a light mineral oil. The experiments were carried out in a 4" OD tube. A Texas Nuclear device was used to measure the bed density. Their results are presented in plots of bed voidage versus superficial liquid velocity.

These data were analyzed using the Richardson-Zaki (2) correlation. In all cases, the particle Reynolds number was greater than 500. The slope (n) from a log-log plot of ϵ versus U_L/U_T was determined as a function of shape factor, K. The values of K were 0.55 and 0.77. The results were added to the plot of n versus K from last month's progress report, which is Figure 1 in this report. At K = 0.55, the agreement with the results of Richardson-Zaki (2) is good. However, at K = 0.77, the agreement is poor. These results indicate that more experiments need to be performed to determine the effect of particle shape on the Richardson-Zaki index, n.

Gas-Liquid-Solid Fluidization

The HRI data presented in Appendix A of the memorandum entitled "Gas-Liquid-Solid Fluidization" by Wolk (1962) were analyzed to determine if it would support the model of three-phase fluidization presented by Darton and Harrison (3). The data are for the expansion of cylinders of diameters 0.025, 0.050, and 0.063". The length of the cylinders in all cases was 3/16". The liquid and gas were heptane and nitrogen, respectively. The bed diameter was varied from 5/8 to 6".

In these experiments, only the bed height was measured as a function of liquid and gas superficial velocity. Thus, the liquid and gas hold-up (ϵ_L and ϵ_G) must be estimated. Wolk assumes:

$$\epsilon_G = \frac{h - h_0}{h} \tag{1}$$

where h = three-phase bed height.
h_0 = bed height with zero gas flow.

It is believed that this assumption will be true provided the gas bubbles do not create any particle-free wakes in the bed. It is equivalent to assuming that the only effect of the gas is to reduce the area available for liquid flow, thus increasing the interstitial liquid velocity and causing the bed to expand. In some cases this assumption is not true.

The data for 0.025" diameter particles in a 1" diameter bed were analyzed using this assumption. ϵ_G was obtained from Equation 1; ϵ_S (volume fraction of solids) was determined from the bed densities reported. ϵ_L is then given by:

$$\epsilon_L = 1 - \epsilon_S - \epsilon_G \tag{2}$$

The Darton and Harrison model (3), based on the drift-flux approach of Wallis (4) was then used to correlate the HRI data. The gas drift-flux is defined as the volumetric flux of gas relative to a surface moving at the average velocity (gas plus liquid). For a three-phase system it is given by:

$$\nu_{CD} = U_G(1 - \epsilon_G) - U_L\epsilon_G \frac{(1 - \epsilon_G)}{\epsilon_L} \qquad (3)$$

where ν_{CD} = gas drift flux, cm/sec.
U_G = superficial gas velocity, cm/sec.
U_L = superficial liquid velocity, cm/sec.

Darton and Harrison analyzed the data of Michelsen and Ostergaard (5) for the air-water fluidization of glass spheres of 6, 3, and 1 mm diameters. They obtained a plot of ν_{CD} versus ϵ_G. It reveals two flow regimes: the uniform bubbling and the churn-turbulent. In the churn-turbulent regime, large gas bubbles (slugs) rise through the center of the bed. The transition between the two regimes is governed by the relative rates of bubble coalescence and disintegration in the bed. Kim, Baker, and Bergougnou (6) also present evidence for the existence of two distinct flow regimes.

ν_{CD} for the 0.025" diameter cylinders was calculated and plotted as a function of ϵ_G. The results are shown in Figure 2 along with the results of Darton and Harrison (3). As shown in Figure 2, all the HRI data lie in the uniform bubbling regime. No transition to the churn-turbulent regime is observed.

In general, the assumption expressed in Equation 1 will not hold. This is illustrated by the data for the 0.025" diameter cylinder in the 6" diameter bed. ϵ_S for this case is plotted as a function of U_G and U_L in Figure 3. For U_L equal to 6.0 cm/sec, ϵ_S increases with the addition of gas, thus indicating the bed contracts upon the addition of gas. In this case, significant liquid wakes are being formed in the bubble paths.

Review of Literature on Bubble Behavior

As indicated by the above discussion, the behavior of bubbles in three-phase fluidized beds is important in understanding the mechanism of bed contraction and in determining which flow regime is present in the three-phase bed. Any study of this area should also include the behavior of bubbles in gas-liquid systems. During the last month, several articles were reviewed to develop a background in this area. The majority of papers fall into two main categories: the behavior of single bubbles and the behavior of swarms of bubbles.

Davies and Taylor (7) studied the rise of single bubbles in an inviscid fluid. They assumed that flow around the bubble would be given by the potential flow solution for flow around a sphere. This assumption resulted in the following relation between the bubble velocity (U_B) and the radius of curvature of the spherical cap bubble (R):

$$U_B = 2/3 \sqrt{gR} \qquad (3)$$

where g is the acceleration of gravity.

They found Equation 3 adequately predicted the rise of air bubbles in water and nitrobenzene.

Since this study, numerous studies on the effects of fluid properties and bubble shape on bubble velocity have been made. A brief review of these studies has been made by Darton and Harrison (8). In general, these studies determined the drag coefficient of the bubble as a function of bubble Reynolds number and Weber number.

The rise velocity of swarms of bubbles through stagnant liquid columns was studied by Nicklin (9). His most interesting finding was that the bubble velocity declined with increasing superficial gas velocity (increasing gas hold-up). No explanation of this behavior is given, although it may be caused by an increase in the effective viscosity of liquid caused by the presence of other bubbles. Nicklin also determined that continuously generated bubbles rise faster than their buoyance velocity by an amount equal to G/A, where G is the volumetric flow rate of gas and A is the cross-sectional area of the tube. This additional velocity arises from the fact that as the gas enters the tube, it tends to raise the contents of the entire tube at a velocity of G/A. For finite liquid flows, an additional term equal to L/A must also be added. L is the volumetric flow rate of liquid.

Descriptions of bubbles and their wakes in fluidized beds are given by Stewart and Davidson (10) and by Rigby and Capes (11). Stewart and Davidson proposed that the creation of particle-free wakes is the cause of bed contraction with the addition of gas. Darton and Harrison (8) measured the rise of single air bubbles in water-fluidized beds of silica sand. The equivalent diameters of the bubbles ranged from 5 to 25 mm. They present their results in a plot of the bubble drag coefficient versus bubble Reynolds number. The following limiting relationships were found:

$$C_D = 2.7 \qquad Re > 100 \qquad (5)$$

$$C_D = 38\ Re^{-1.5} \qquad Re < 2 \qquad (6)$$

The bubble drag coefficients and Reynolds number are based on bubble velocities relative to the liquid velocity. It was also found that the fluidized bed increased the apparent viscosity of the liquid. Massimilla, Solimando, and Squillace (12) also performed experiments with single bubbles in fluidized beds.

The rising velocity of swarms of bubbles in fluidized beds has been measured by Rigby, et al. (13) and by Kim, Baker, and Bergougnou (6). Both used air-water as the fluidizing medium. Their particles range from 0.12 mm to 0.775 mm, and 2.6 mm to 6 mm, respectively. Very large bubble velocities (20-200 cm/sec) were found in both experiments. These velocities are much greater than those reported by Darton and Harrison (8) and Massimilla, et al. (12). No explanation for these large velocities is given. However, the bubbles are quite large relative to the diameter of the flow equipment used in the studies. Thus, slugging may be occurring.

Several articles were reviewed on the subject of the mechanism of break-up of large bubbles in fluidized beds. Henriksen (14) and Clift (15) propose that bubble break-up occurs by a Taylor (16) instability on the roof of the bubble. Taylor showed that for an inviscid fluid, disturbances on horizontal surfaces between the fluids grow if the upper fluid is more dense than the lower fluid. In the case of bubbles, this growth could lead to break-up of the bubble.

During the next month, the literature review will continue. Emphasis will remain on articles dealing with bubbles and slugs in two- and three-phase systems.

Experimental Techniques

A review of available experimental techniques in multi-phase flow was also undertaken. Seven techniques were identified as having potential for this project. A description of these techniques and their evaluation are presented in Table II.

MEASUREMENT OF PHYSICAL PROPERTIES OF H-COAL LIQUIDS

HRI was contacted with respect to taking samples of H-Coal liquids from the PDU reactor for viscosity measurement. It was determined that currently there are no provisions for taking samples from the reactor or the gas/liquid separator. Samples could be obtained downstream of the "let-down" valve, where the pressure is significantly reduced. However, the reduced pressure will result in a loss of light components. Therefore, sampling will be delayed until provisions can be made to take liquid samples either from the reactor recycle or the gas/liquid separator.

During the next month a high-pressure sampling system will be designed to take samples without the loss of light ends. Battelle will be contacted to assure that the design is compatible with their viscometer and that the sample can be transferred without loss of light ends. Progress will be related to HRI soon after Battelle is contacted.

DESIGN OF THE FLUID DYNAMICS UNIT

Process Design

The design of the fluid dynamics unit continued this month. The 6" ID reactor will be constructed from four 5'-long glass sections connected with stainless steel spool pieces. The spool pieces and glass sections will be held together with flanges. The mechanical design phase of work is well under way. Progress was made in the following areas:

1) Based on blueprints of the recycle and distribution cups obtained from HRI, appropriate mechanical details were incorporated into the reactor design.

2) Finalized design of support structure. Obtained bids.

3) Ordered gas compressor, feed tanks and mixers, gas/liquid separators, and piping.

In the instrumentation area the following were accomplished:

1) Ordered display control consoles.

2) Obtained quotation on gamma-ray scan system.

3) Determined number and location of pressure taps on the reactor.

4) Ordered differential pressure transducers.

During the next month, the following are planned:

1) Finalize mechanical design of reactor, including sampling taps.

2) Order recycle and feed pumps.

3) Continue mechanical design of gamma-ray elevator system.

4) Initiate systems design.

Computerization

During this month the computer requirements were assessed. It was determined that the following items will be read and recorded by the computer as a function of time:

1) Fresh and recycle liquid flow rate.

2) Gas flow rate.

3) ΔP along the reactor.

4) Transmission of gamma rays through the reactor versus reactor height.

5) Temperature versus reactor height.

6) Six analog inputs will also be available for gas or liquid tracer experiments.

Sampling rates on these inputs will vary between 100/sec to 1/min. In addition, the computer will perform the following functions:

1) Operate the elevator for the gamma-ray system.

2) Print and plot data.

3) Data reduction and computations.

During November, efforts will concentrate on the following:

1) A computer system capable of meeting the above needs will be selected.

2) Specific program requirements will be defined.

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TABLE I

SUMMARY TABLE OF PRIOR WORK

Areas	No. of Papers Selected For Review	Number Reviewed
1) Gas-Liquid-Solid Fluidization (Ebullating Beds)	38	25
2) Liquid-Solid Fluidization	13	13
3) Vertical Gas-Liquid Flow (Bubble and Slug Behavior)	14	10
4) Slurry-Gas-Solid Fluidization	10	0
5) Slurry-Solid Fluidization	--	--
6) Experimental Techniques in Multi-Phase Flow	24	24
7) Properties of Coal-Oil Mixtures	11	5
	110	77

DFT/ml
11/3/77

TABLE II

REVIEW OF EXPERIMENTAL TECHNIQUES

Technique	Description	Disadvantages
Gamma-Ray	Based on the differential adsorption of rays by the gas, liquid, and solids phases. Can give quantitative description of two phases.	Technique by itself cannot give quantitative breakdown of hold-up of three phases. Needs to be coupled with other phase measurements to give hold-up information for H-Coal fluid dynamics unit.
Radioactive Tracers (Gas and Liquid)	The flow of the radioactive phase is monitored from outside of the reactor. Will give the hold-up of the tagged phase while traversing the column.	Safety considerations limit the number of tests allowed per year, so cannot be used for daily data collection. Analysis problem with gas-phase tracers because different size bubbles with varying amounts of tracer rise at different rates.
Ultrasonic Scans	Based on differences of the speed of sound through different phases. Can give quantitative description of two phases.	The differences between the speed of sound for several liquid-gas pairs used in these experiments are not large enough to differentiate the phases. Needs to be coupled with other phase measurements to give quantitative hold-up data for three phases.
Conductivity Probe	Based on the electrical conductivity of gases, liquids, and solutions. Could use to differentiate between gas and liquid phases. By adding an electrolyte to the liquid as a step input, the technique could be used to determine liquid-phase hold-up.	Electrolytes may not be soluble in all liquids which we plan to use. The probes have to be in contact with the fluids, so there could be disruption to the flow and several probe locations would have to be put in the reactor. Evaluation is still continuing.
X-Ray Scans	Based on the same principle as gamma-ray scans, but the rays are of much lower energy. Will give additional information since at lower energies the mass absorption of the phases changes.	This technique also needs to be used in conjunction with another phase measurement.
Light Probe	There are several different types of light probes:	
	1) Refractive index principle. The light probe is arranged so that if liquid is at the tip, light is transmitted back up the probe to a detector. When gas is at the tip, the light is refracted at a different angle and is not detected. Both gas and liquid phase hold-ups can be determined.	The probe must be in the flow, which can cause flow pattern interference. There must also be several probe taps in the reactor.
	2) Light transmittance. A liquid tracer method. A step input of dye is added to the liquid stream. The probe consists of two optical fibers separated by a gap. Light transmitted depends on the concentration of dye in the gap. Liquid-phase hold-up is determined.	Coal fines will be used to make a liquid slurry, and light will not be transmitted by the coal fines, so it could be used for only gas-liquid experiments.
	3) Light blockage. Light is blocked by solids, so the light is transmitted only when gas or liquids pass the probe. Two different probes were used in conjunction to measure the size of liquid and gas bubbles as they pass the probe system.	Coal fines in the slurry will interfere with the probe. There also would not be any differentiation between gas and liquid with this type of probe. Flow interference may also be a problem.
Pulse Luminescence	Luminescent particles are used to follow flow of solid or liquid phase. The particles are intermixed with one phase. The particles are excited by a high-intensity light source; then the particles themselves emit light and their flow can be monitored.	The light emitted by the luminescent material will be obscured by coal fines in the slurry, but method could be used for experiments without coal fines.

DMS/ml
11/3/77

Figure 1

RICHARDSON-ZAKI CORRELATION
(n VERSUS K)

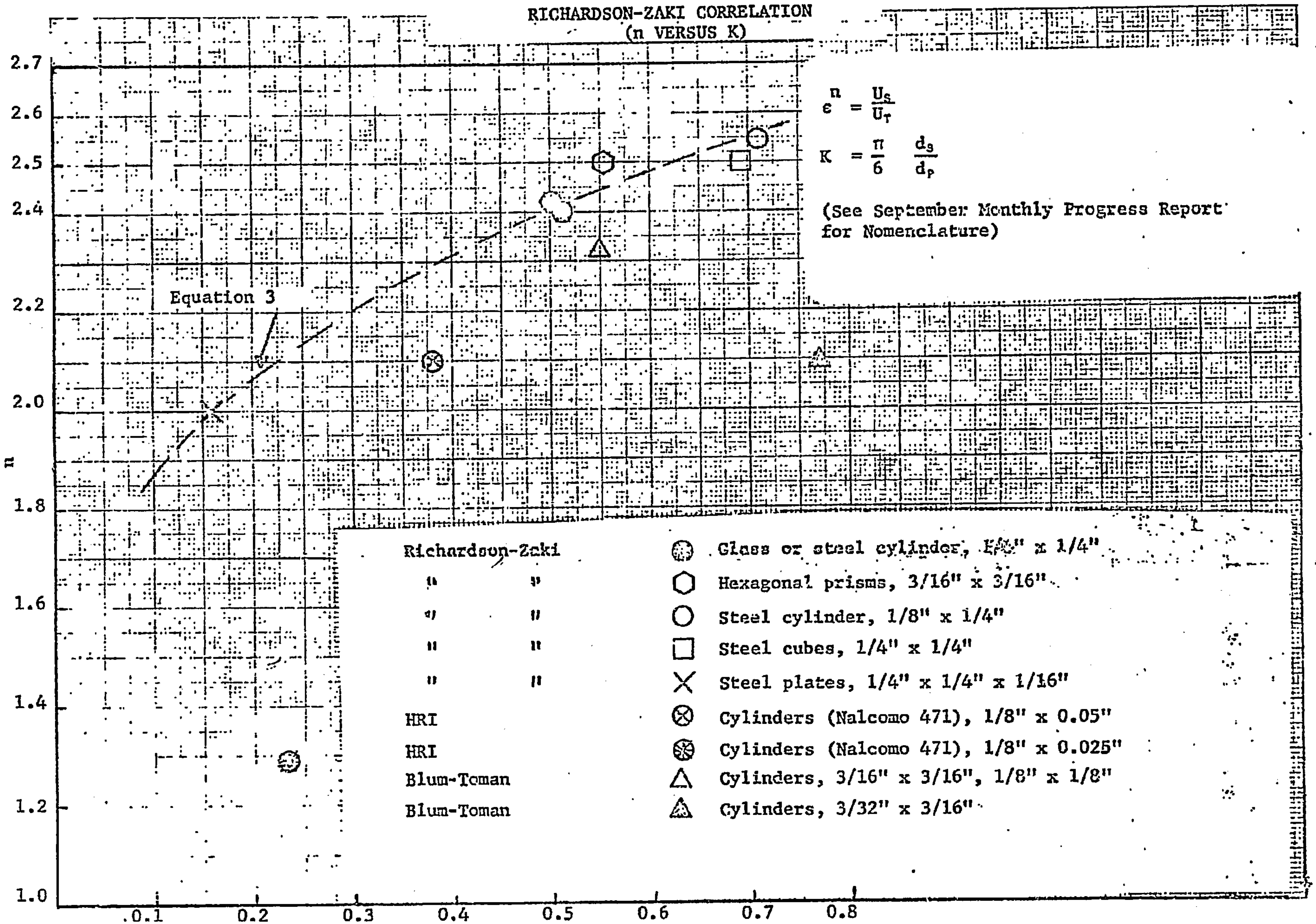

Figure 2

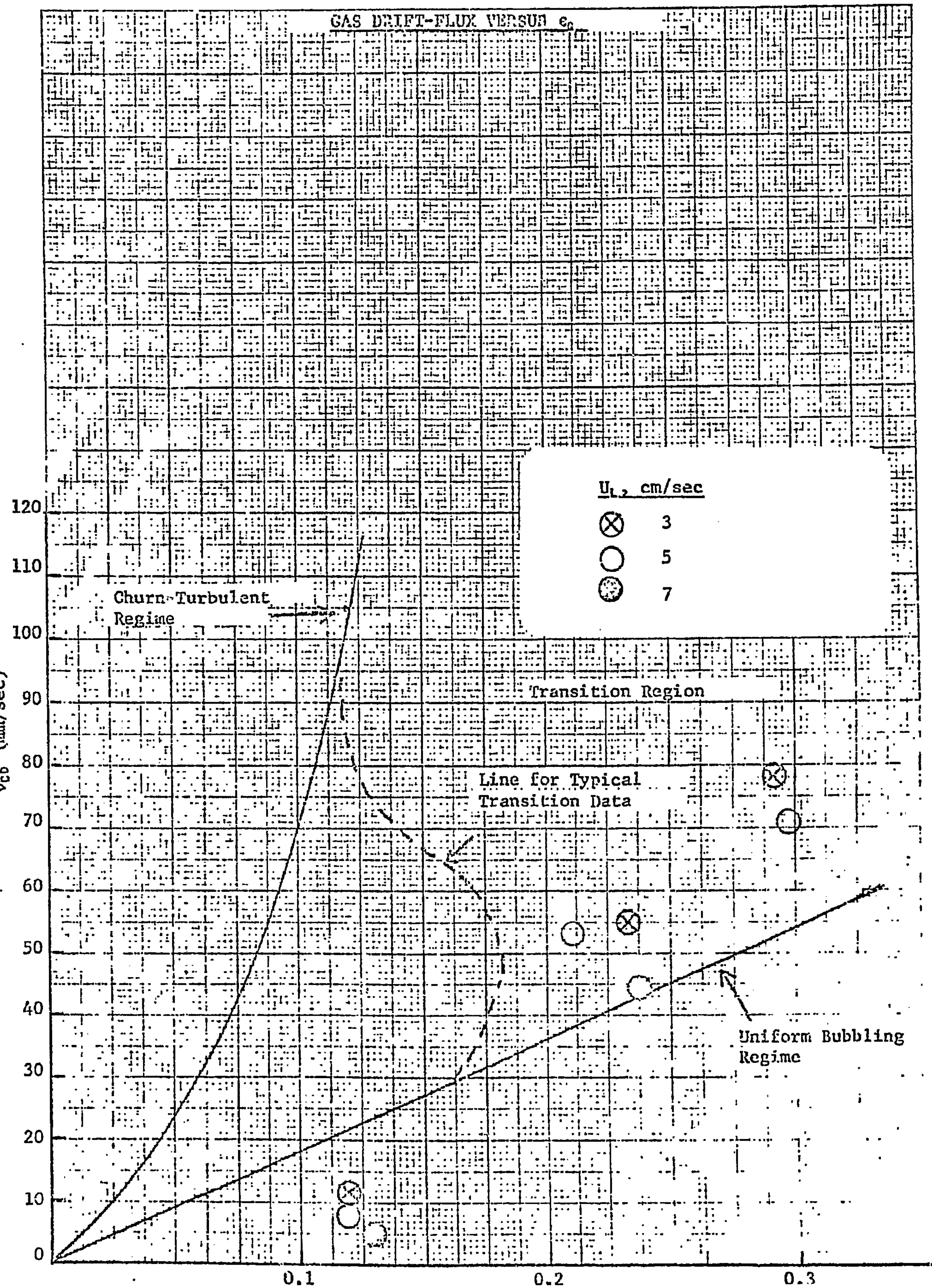

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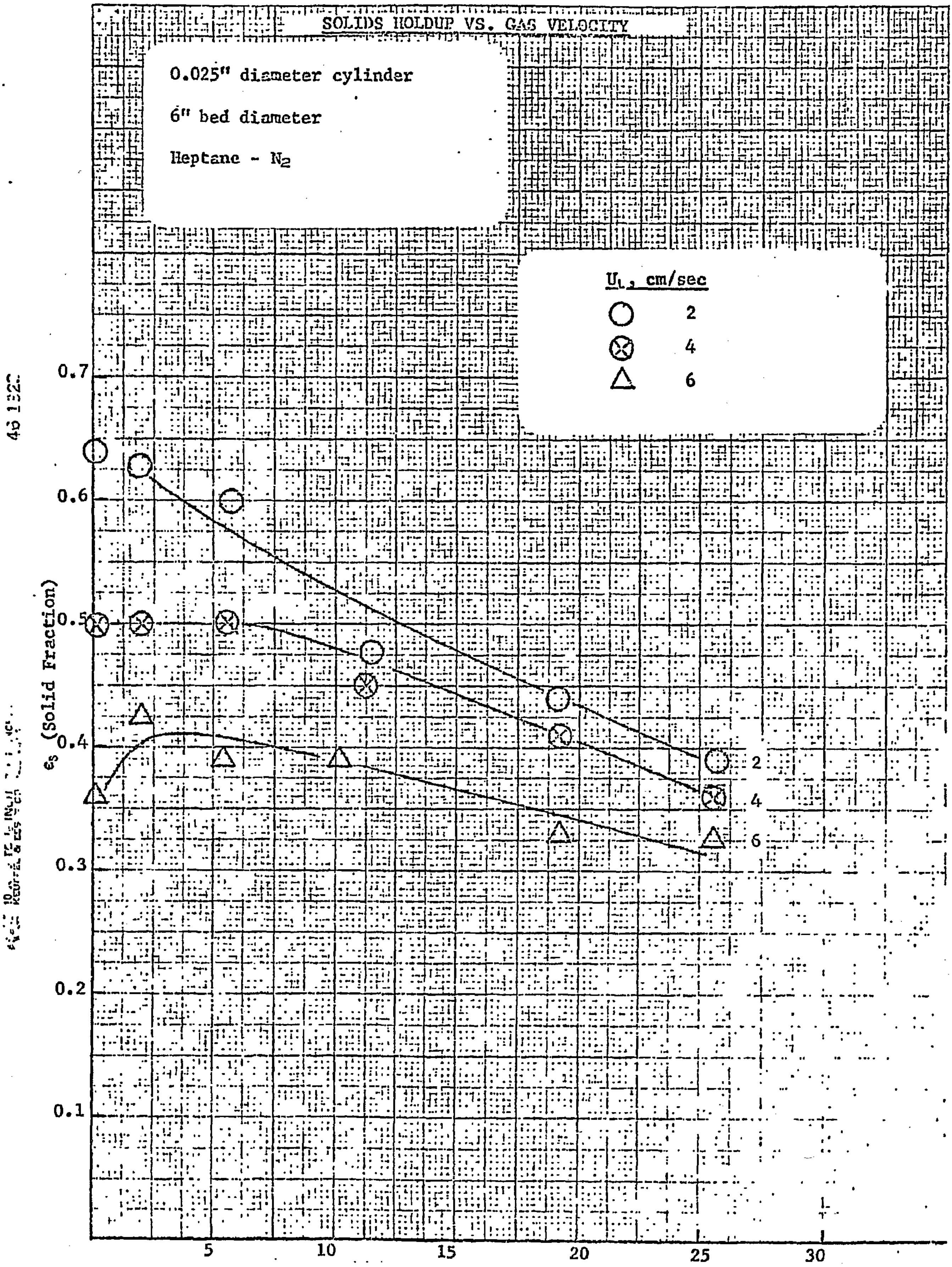
Figure 3



SOLIDS HOLDUP VS. GAS VELOCITY
0.025" diameter cylinder
6" bed diameter
Heptane - N2
UL, cm/sec
2
4
6
e_s (Solid Fraction)
0.7
0.6
0.5
0.4
0.3
0.2
0.1
5
10
15
20
25
30
2
4
6

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