STUDY OF EBULLATED BED FLUID DYNAMICS FOR H-COAL. QUARTERLY PROGRESS REPORT NO. 1, AUGUST 22--NOVEMBER 30, 1977

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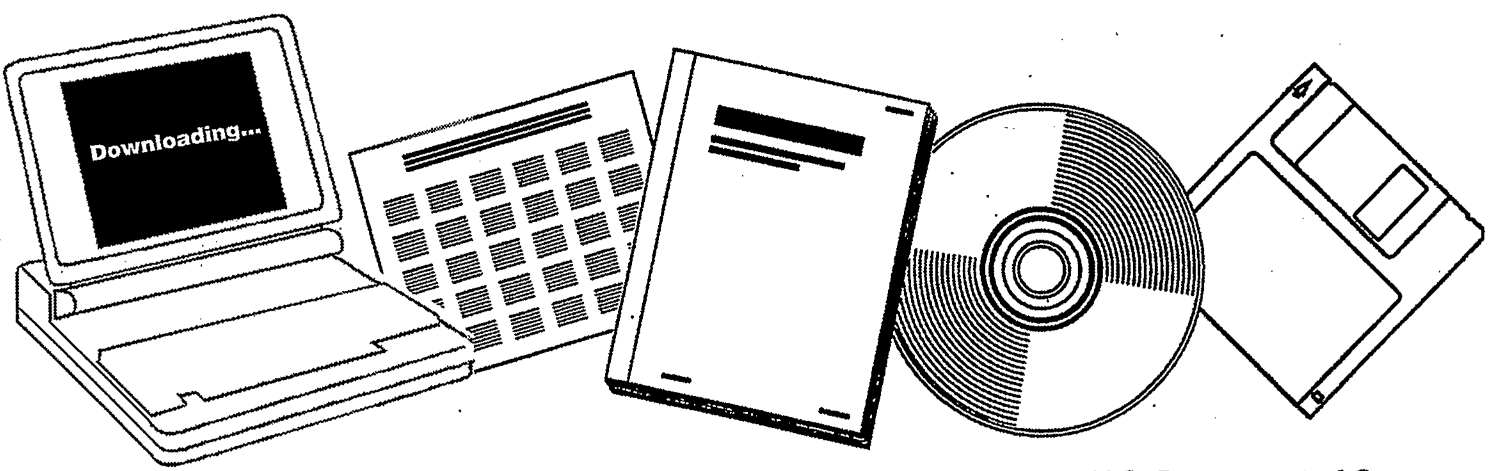

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STUDY OF EBULLATED BED FLUID DYNAMICS FOR H-COAL

QUARTERLY PROGRESS REPORT NO. 1
AUGUST 22-NOVEMBER 30, 1977

BY

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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 operations, and mathematical modelling. The objective of this quarterly progress report is to outline progress in the first two parts during the first three months of the project.

OBJECTIVE AND SCOPE OF WORK

The overall objective of this project is to improve the control of the H-coal reactor through a better understanding of the hydrodynamics of ebullating beds. The project is divided into three main parts: 1) review of prior work in the area of gas-liquid-solid fluidization; 2) construction of a cold flow unit and collection of three-phase fluidization data; and 3) development of a mathematical model. The mathematical model, which will take into consideration information obtained from the literature review and data taken in the cold flow unit, will: a) define steady-state ebullated bed conditions as a function of operating parameters; and b) present criteria for bed stability. Progress which has been made in Parts 1 and 2 during the first three months of this project will be presented in this report.

SUMMARY OF PROGRESS TO DATE

Review of Prior Work

A review of the literature was undertaken to determine what data, models, and correlations exist for describing the behavior of liquid-solid, gas-liquid, and gas-liquid-solid systems. This review also includes reports provided by Hydrocarbon Research, Incorporated (HRI). The status of the review is reported in Table I. Thus far, 155 papers have been identified as of potential value to the H-coal project. Papers which will be included in the literature review report will be selected from these papers. The literature review report will be completed by the first quarter of 1978.

The papers included in Table I were found by searching the following sources: API, Engineering Index, Chemical Abstracts, and ERDA data base.

Unit Construction and Data Collection

Measurement of Properties of H-Coal Liquids.--Various laboratories were contacted to determine their capability of measuring the viscosity and surface tension of H-coal liquids under reactor conditions. A list of the laboratories contacted, the techniques they use to measure viscosity, and the limitations of these techniques was presented in Table III of the September monthly progress report. Battelle Memorial Laboratory was chosen to carry out the viscosity measurements. No laboratory was found which has the capability of measuring surface tension under reactor conditions. Both HRI and Battelle have been visited to determine the requirements for obtaining and transferring the samples.

Design of the Fluid Dynamics Unit

The design of the fluid dynamics unit has started. The process flow sheet was finalized. Major vessels, pumps, the compressor, and process flow lines were sized. The mechanical design of the steel support structure was completed. The 6" reactor will be constructed from four 5'-long glass sections connected with stainless steel spool pieces and flanges. The reactor recycle cup and distributor were designed with the assistance of HRI.

Progress was also made in various other aspects of the design of the unit. Feed tanks and impeller systems were designed, major items for the unit were ordered, techniques for measuring individual phase holdup, and computer requirements were identified.

REVIEW OF PRIOR WORK

Liquid-Solid Fluidization

The most widely used method of correlating liquid-solid fluidization data is that of Richardson and Zaki (1). Their correlation has the following form:

$$\epsilon^n = \frac{U_s}{U_t} \tag{1}$$

where: ϵ = Bed voidage
U_s = Superficial liquid velocity
U_t = Terminal velocity of a single particle.

In general:

$$n = f(Re_t, \frac{d}{D}, \text{particle shape}) \tag{2}$$

where: Re_t = Particle Reynolds number based on U_t
d = Particle diameter
D = Bed diameter

Detailed description of Equation 2 for spherical particles was given in the September, 1977 monthly progress report. For non-spherical particles with $Re_t > 500$, Richardson and Zaki propose:

$$n = 2.7\, K^{0.16} \tag{3}$$

where: $$K = \pi/6 \frac{d_s}{d_p} \tag{4}$$

where: d_s = Diameter of a sphere with the same volume as the particle
d_p = Diameter of a circle of the same area as the projected particle when liquid is its most stable configuration

HRI's data (2) and the data of Blum and Toman (3) for liquid fluidization of cylindrical particles with various length to diameter ratios were analyzed using Equations 1-4. The results are presented in Figure 1, where n is plotted as a function of K. The results indicate that Equation 3 may be inadequate. More experiments need to be performed in order to determine the effect of particle shape on n.

Fouda and Capes (4) suggest the following modification of Equation 1 for very fine particles, aggregated particles, or irregularly shaped particles:

$$\frac{U_s}{U_t} = (1-K'C)^n \tag{5}$$

where: $C = 1-\epsilon$
K' = Parameter which accounts for the effective hydrodynamic volume of the particle.

K' for spheres is equal to unit. They found that K' ranges from 1.7 to 1.43 for equidimensional but irregularly shaped particles. Values of K' as high as 3 were found for flat shaped particles. Fouda and Capes propose:

$$K' = \left[\left(\frac{d_2}{d_1}\right)\left(\frac{S_2}{S_1}\right)\right]^{0.284} \tag{6}$$

where: $\frac{S_2}{S_1} = \frac{\text{Specific surface of particle}}{\text{Specific surface of an equivalent sphere}}$

$\frac{d_2}{d_1} = \frac{\text{Average volume equivalent diameter}}{\text{Particle thickness}}$ for flat particles

or: $\frac{\text{Average circumscribing diameter}}{\text{Average screen diameter}}$ for equidimensional particles

Equation 6 is based on data for flat particles and irregular equidimensional particles. The flat particles had an average screen diameter ranging from 1.3 to 3.6 mm and average thickness ranging from 0.065 to 1.2 mm. The screen size of the irregular particles ranged from 52 to 1825 microns.

Brea, Edwards, and Wilkson (5) studied the flow of non-Newtonian slurries through fixed and fluidized beds of uniform spherical particles. The slurries consisted of titanium dioxide dispersed in water. The titanium dioxide particles ranged in size from 0.18 to 1 micron, and their concentration in water ranged from 10 to 22 wt%. The viscosity of the slurries was measured by an in-line tubular viscometer. The wall shear stress for these slurries was found to exhibit a power law dependency on the shear rate:

$$\tau_{\omega} = M\left(\frac{8\nu_m}{D}\right)^{n'} \tag{7}$$

where: τ_{ω} = Wall shear stress
ν_m = Mean velocity in viscometer
D = Diameter of capillary in viscometer
M = Rheological parameter

The authors developed a model for the friction factor for packed beds. Extension of this model to fluidized systems leads the authors to propose that n in Equation 1 also becomes a function of n' for $Re_{mf} < 40$, but is independent of n' for $Re_{mf} > 40$. Re_{mf} is the minimum fluidization Reynolds number for the bed.

Gas-Liquid-Solid Fluidization

Three-phase systems are more complex than liquid-solid systems. The addition of gas allows for possible gas-liquid flow regime transition to occur in the bed (6,7). It has also been found experimentally that under certain conditions the bed can contract with the addition of gas (8). It is proposed that the contraction occurs because of the formation of particle-free liquid wakes behind the bubbles which move through the bed at the bubble velocity (9). This action reduces the interstitial liquid velocity in the rest of the bed, causing it to contract.

The literature on models correlating the fractional holdup of three-phase systems was reviewed. Darton and Harrison (7) have proposed four criteria which such models should fulfill. These are:

1) The model should reduce to a suitable liquid-solid fluidization model (i.e., Richardson-Zaki) as ϵ_g approaches zero.

2) The model should reduce to a suitable gas-liquid model as ϵ_s approaches zero.

3) $\epsilon_s + \epsilon_g + \epsilon_l = 1.$

4) The model should predict whether the bed will expand or contract upon the addition of gas.

A description of the various models reviewed and comments in light of the above criteria are given in Table II of the September monthly progress report.

The model proposed by Darton and Harrison (7) appears to be the best suited for meeting these criteria. The three-phase fluidization data of HRI (10) was analyzed to determine if it would correlate via the Darton and Harrison model. A discussion of this analysis was presented in the October monthly progress report.

<u>Gas-Liquid Systems</u>

The gas-liquid phase transition and bubble wake formation in fluidized beds indicates the importance of understanding the behavior of gas-liquid systems. An understanding of this area is also important if Darton and Harrison's second criterion is to be met. Papers reviewed in this area have been in the following categories: velocity of single bubbles in liquids and fluidized beds; velocities of swarms of bubbles in liquids and fluidized beds; bubble break-up; bubble coalescence; and bubble wakes. Several papers from the first two areas were discussed in October's monthly progress report. Additional papers from these areas reviewed during November will now be discussed.

Grace (11) has precented an excellent review of the shapes and velocities of bubbles rising in infinite liquids. Data from eight sources are analyzed. For the case where gas density and viscosity are small compared to that of the liquid, bubble shape and velocity are functions of three dimensionless groups:

$$\text{Reynolds Number} = \frac{\rho_c d_c U}{\mu_c}$$

$$\text{Eotvos Number} = \frac{g d_e^2 (\rho_c - \rho_d)}{\sigma}$$

$$\text{Morton Number} = \frac{g \mu_c^4}{\rho_c \sigma^3}$$

where:
- ρ_c = Liquid density
- d_e = Equivalent spherical diameter
- ρ_d = Gas density
- μ_c = Liquid viscosity
- g = Acceleration of gravity
- σ = Surface tension
- U = Terminal bubble velocity

A generalized plot of Reynolds number versus Eotvos number with the Morton number as a parameter is presented in Reference 11. Three distinct bubble shape regions were found: spherical, ellipsoidal, and spherical cap. Estimates of the three dimensionless groups for bubbles in the liquids selected for study in the cold flow unit indicated that bubbles from all three groups could be present.

Lockett and Kirkpatrick(12) have reviewed the numerous models relating the bubble slip velocity (U_s) (bubble swarm velocity - liquid velocity) of a bubble swarm to the gas holdup, ϵ_g, and the terminal velocity of a single bubble, U_{bo}. The various models include the following:

1) $U_s = U_{bo}$ proposed by Turner (13).

2) $U_s = U_{bo}/(1-\epsilon)$ proposed by Davidson and Harrison (14).

3) $U_s = U_{bo}(1-\epsilon)^{n-1}$--a Richardson and Zaki (1) type relationship proposed by Bridges (15).

4) $U_s = U_{bo}(1-\epsilon)/(1-\epsilon^{5/3})$ proposed by Marrucci (16).

Lockett and Kirkpatrick measured U_s as a function of ϵ_g up to $\epsilon_g = 0.66$ for 5 mm diameter bubbles. They found the Richardson-Zaki-type correlation with n = 2.39 gave the best correlation. However, at ϵ_g greater than approximately 0.25, even this correlation had to be multiplied by an empirical correction factor. They suggest that this correction factor is necessary because of bubble deformation at high ϵ_g. Davidson and Harrison (14) also analyzed the data of Bridges (15) and found it correlated best via a Richardson-Zaki-type model. The highest gas holdup analyzed was 0.20. Lockett and Kirkpatrick also explored reasons for deviations from ideal bubble flow. These include liquid circulation, bubble cluster formation, and large bubbles. Hills (17) points out that they are intimately related. Large bubbles can form from clusters and cause increased liquid velocity at the center of the column with downflow at the walls. The radial velocity at the base of the column caused by the downflow tends to cluster small bubbles. Such a phenomenon could explain the large bubble velocities measured by Rigby, et al. (18) and Kim, et al. (19) for flow of swarms of bubbles through fluidized beds.

Henriksen and Ostergaard (20) and also Clift and Grace (21) have proposed that bubbles in fluidized beds break up via Taylor instability which develops on the roof of the bubble. Taylor (22) showed that for an inviscid fluid, disturbances on horizontal surfaces between the fluid grows if the upper fluid is more dense than the lower fluid. Bellman and Pennington (23) further refined Taylor's theory to include the effects of viscosity and surface tension. They showed that surface tension determines the lowest disturbance wave length which will grow, and viscosity determines the disturbance wave length which grows the fastest. In fluidized beds it might be assumed that the fluidized particles introduce disturbances into the upper surfaces of the bubbles as they rise. It might be expected that the wave length of these disturbances would be proportional to the particle size. Thus, if the fluidized particles are sufficiently small, the wave lengths of the disturbances they introduce may be below the critical wave length, and surface tension will dampen the disturbance out. Large bubbles would be expected in a bed of these particles. However, if the fluidized particle sizes are sufficiently large, the wave length of the disturbance they introduce will be larger than the critical value, and the disturbance will grow. Small bubbles would be expected in this bed.

This effect has been illustrated by Ostergaard (24) in air-water fluidized beds of 1 mm and 6 mm glass ballotini.

Bubble coalescense is extremely complex. It can be dominated by gas distributor design (25), the presence of contaminates such as surfactants (26), or inorganic salts (25), or by the flow pattern of the bed or column. Models describing the latter must predict how bubbles cluster and how the film between them drains.

Calderbank, et al. (27) have studied the problem of bubble clustering. Kirkpatrick and Lockett (28) have developed a model describing the rate of film drainage between coalescing bubbles.

Narayanan, et al. (29) have classified the wakes behind bubbles rising in liquids into five different classes. The class is a function of the bubble Reynolds number. For Reynolds numbers less than two (Class I), the bubbles are spherical in shape with a thin, trailing wake. Between a Reynolds number of 2 and 7 (Class II), the bubbles develop a cusp at the rear but still retain a thin, trailing wake. At Reynolds numbers between 7 and 80 (Class III), the bubbles become spherical cusps and develop stable wake vortices. Between a Reynolds number of 80 and 300 (Class IV), the spherical cap begins to oscillate, shedding its vortices. Above a bubble Reynolds number of 300 (Class V), the bubble becomes irregularly shaped and the wake becomes turbulent. Gas bubble wakes from Class III and IV have been observed by Rigby and Capes (30) and by Stewart and Davidson (31) in water fluidized and sand and glass ballotini, respectively. Narayanan, et al. also present a correlation of wake length as a function of bubble Reynolds number for wakes in Class III.

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 were presented in Table II of the October monthly progress report.

CONSTRUCTION OF COLD FLOW UNIT AND DATA COLLECTION

Measurement of Properties of H-Coal Liquid

Application of the results of this project to the H-coal reactor will require knowledge of the surface tension and viscosity of the H-coal liquids under reactor conditions (800-900°F, 2000-3000 psi). Various laboratories were contacted to determine their capabilities for measuring these properties. Table III of the September monthly progress report lists the laboratories and the techniques they use to measure viscosity. None of the laboratories had the capability of measuring surface tension under reactor conditions.

A proposal was submitted by Battelle to carry out the viscosity measurements as a subcontractor. Battelle proposed to determine the viscosity of four different oil samples. These determinations are to be made at no less than four temperatures ranging from 700° to 900°F and at pressures ranging from 2000 to 3000 psi of hydrogen.

The cost of the proposal is about $15,000. The proposal is contingent upon the availability of the apparatus when the H-coal slurries are available. The proposed cost will also increase with time.

HRI and Battelle have been visited to determine the requirements for obtaining and transferring the samples.

Design of the Fluid Dynamics Unit (FDU)

The design of the fluid dynamics unit to study the hydrodynamics of an ebullated bed has started. The unit will consist of a 6" ID reactor made of four 5'-long glass sections connected with spool pieces and flanges. Reactor internals will duplicate those used by HRI in the H-coal reactor. Figure 2 puts in perspective the overall progress and the construction schedule. Details on each item follow:

Process Design.--Figure 3 shows a schematic diagram of the fluid dynamics unit (FDU). The size of the vessels and diameters of the lines were established by using information supplied either from HRI reports or by HRI personnel.

Mechanical Design.--This phase of work is about 50% completed. Preparing machine shop drawings of distributor cup, recycle cup, and spool pieces has been completed. The gas compressor, feed tanks, mixers, gas-liquid separator, piping, and valves were ordered.

A major effort was also placed in the design of the support structure for the reactor system. The supporting structure will consist of four levels. The reactor will be located in a 5' x 3' section in the structure. An elevator housing a gamma-ray scan apparatus will travel vertically along the reactor through the 5' x 3' section. The following is planned for next period:

1) Complete shop construction of spool pieces, distributor, downcomer, and recycle cup.
2) Follow construction of support structure.
3) Order pumps.
4) Complete design of sample taps in reactor spool pieces.

Systems Design.--Blending aspects of the process design, the mechanical design, the electrical design, instrument selection, and automation is the major objective of this phase of work. Recognizing that long delivery items may restrict the construction of the unit on schedule, emphasis was placed on identifying key items for prompt delivery. To date, the following have been completed:

1) Ordered display control consoles and differential pressure transmitters.
2) Evaluated computer and gamma-ray scan systems.
3) Surveyed sonic flow meters for flow measurement of slurries.
4) Evaluated basis for computer requirements.

During the next quarter, the following are planned:

1) Select and order flow meters.
2) Order gamma-ray scan system.
3) Select and order a computer system consistent with the needs of the project.

In addition to the above, systems design drawings will be completed and become available to the craft personnel for field construction.

Data Collection

The unit will be operated at room temperatures using liquids with viscosity and surface tension in the range of those observed in the H-coal process. Potential liquids include: toluene, kerosene, water, and blandol. Gases will include: helium, nitrogen, and Freon 12. A catalyst similar to that used in the H-coal process will be considered.

Measurements will be conducted utilizing differential pressure transmitters, a gamma-ray scan system, radioactive tracers, and direct sampling. These techniques will allow the determination of the bed height, the fractional holdup of each phase as a function of bed height, and the bed behavior when the gas and/or liquid flow is suddenly stopped. Since the collection of the experimental data is not planned until the beginning of May, major emphasis during the next quarter will be placed on identifying limitations of the above experimental techniques. Possible use of other techniques will also be explored.

CONCLUSIONS

A study has been undertaken with the objective of defining the hydrodynamic properties of gas-liquid-solid systems as related to the H-coal process. To date, pertinent literature information on such systems has been reviewed and a cold flow system is being designed. A progress report on the technical progress of the various phases of the design and construction work was presented. Unit completion is scheduled for May 1, 1978.

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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	38
2) Liquid-Solid Fluidization	13	13
3) Vertical Gas-Liquid Flow (Bubble and Slug Behavior)	60	43
4) Slurry-Gas-Solid Fluidization	8	8
5) Slurry-Solid Fluidization	1	1
6) Experimental Techniques in Multi-Phase Flow	35	35
7) Properties of Coal-Oil Mixtures	11	5
	166	143

DFT/ml
12/7/77

Figure 1

RICHARDSON-ZAKI CORRELATION
(n VERSUS K)

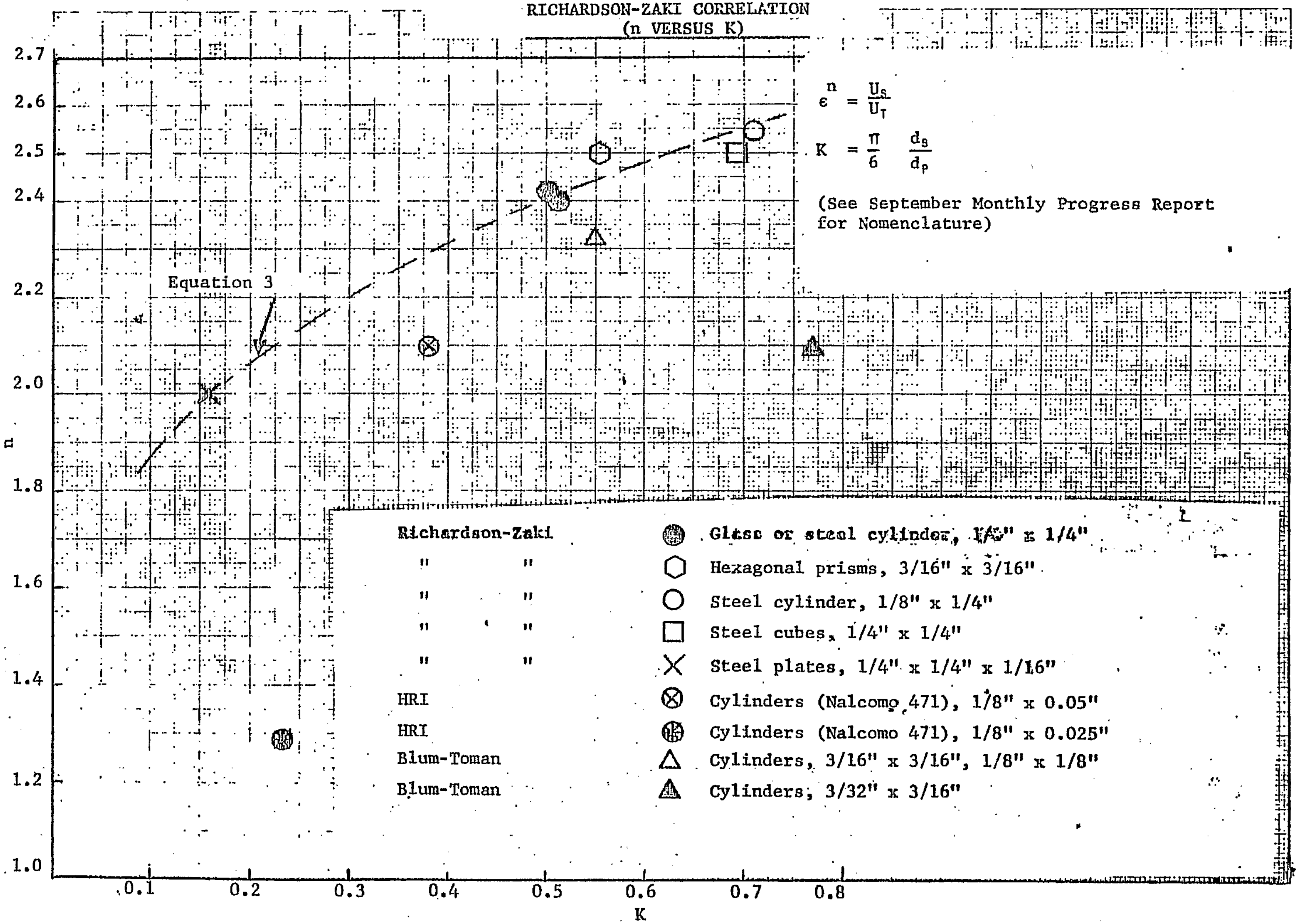
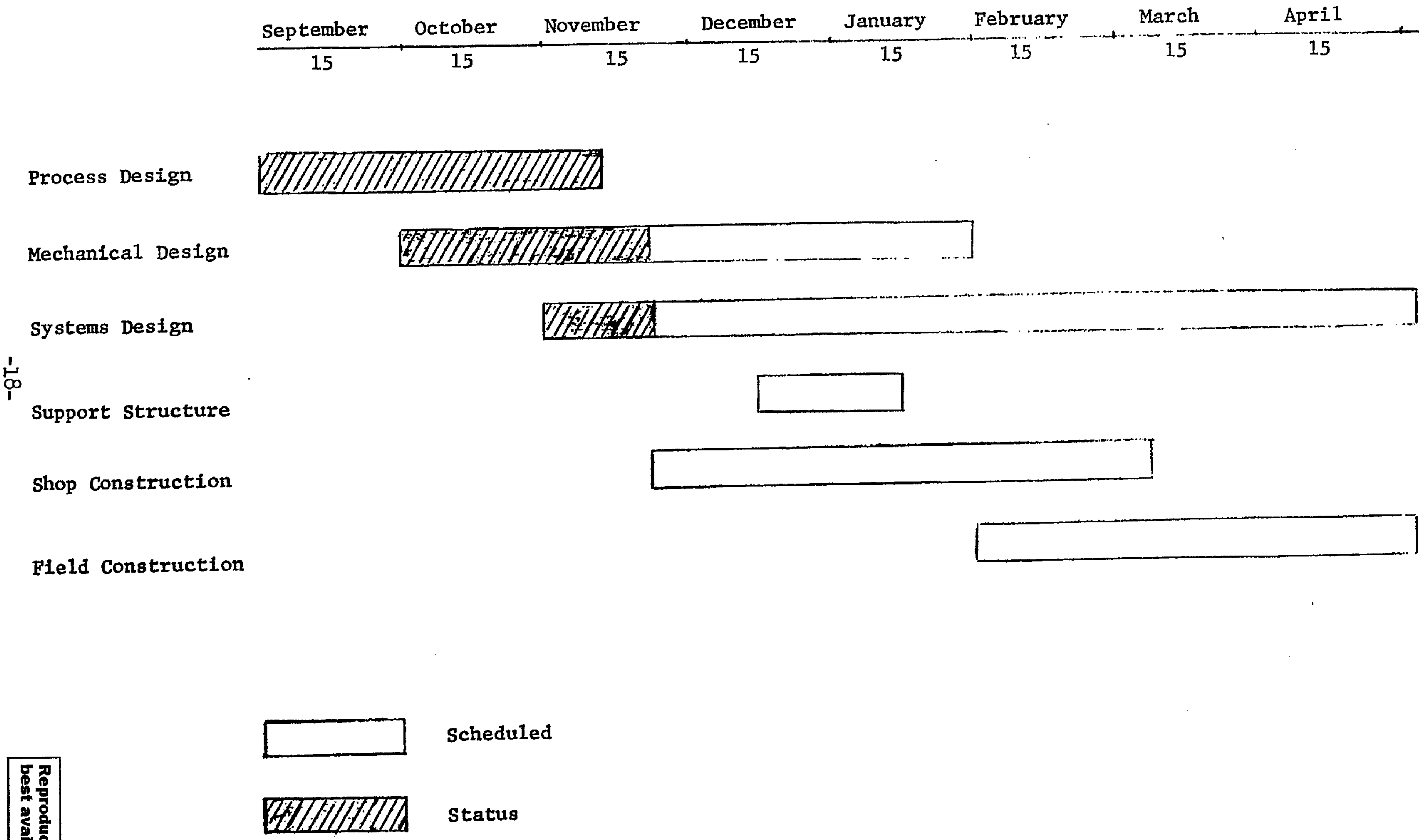


Figure 2

CONSTRUCTION SCHEDULE OF THE FDU



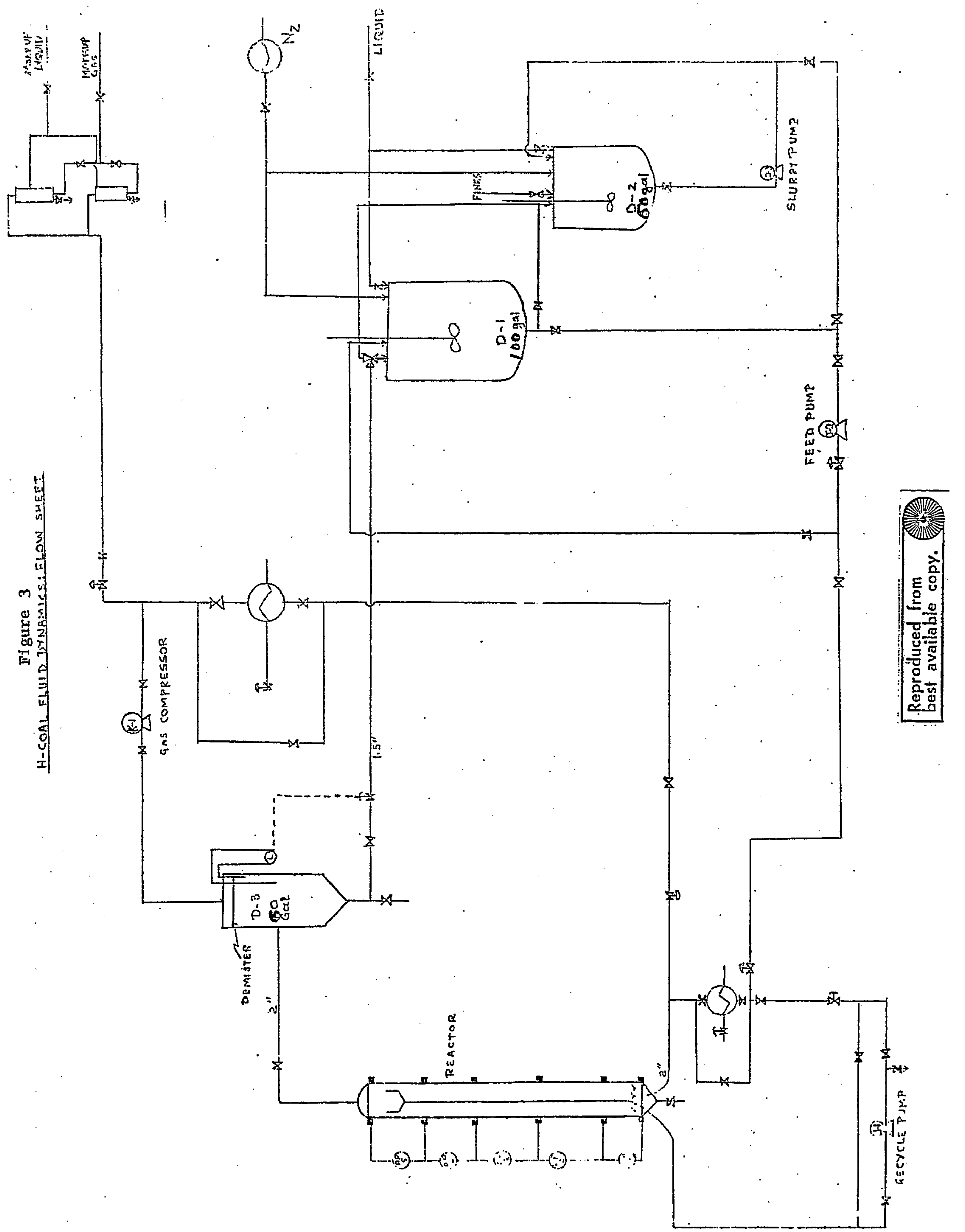


Figure 3
H-COAL FLUID DYNAMICS FLOW SHEET

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