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**LIQUID PHASE METHANOL LAPORTE PROCESS DEVELOPMENT UNIT:  
MODIFICATION, OPERATION, AND SUPPORT STUDIES**

**Topical Report for the Period April 9, 1987–March 9, 1991**

**Task 2.0: Run E-5, Gas Hold-up and Equipment Evaluation Studies**

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## ABSTRACT

Liquid-entrained operations at the LaPorte LPMEOH Process Development Unit (PDU) continued during June and July 1988 under Tasks 2.1 and 2.2 of Contract No. DE-AC22-87PC90005 for the U. S. Department of Energy. The primary focus of this PDU operating program was to prepare for a confident move to the next scale of operation with an optimized and simplified process. Several new design options had been identified and thoroughly evaluated in a detailed process engineering study completed under the LPMEOH Part-II contract (DE-AC22-85PC80007), which then became the basis for the current PDU modification/operating program.

The focus of the Process Engineering Design was to optimize and simplify the LPMEOH process. The proposed process simplifications focused on the slurry loop, which consists of the reactor, vapor/liquid separator, slurry heat exchanger, and slurry circulation pump. Three key conclusions were reached as a result of the detailed process engineering design evaluation: 1) there should be sufficient liquid circulation and turbulence induced by the high synthesis gas velocities to suspend the catalyst particles homogeneously within the slurry, 2) the required internal heat transfer surface should be relatively small so that sufficient heat exchanger area could be installed with little effect on reactor bubble column hydrodynamics, and 3) adequate vapor/liquid disengagement can potentially be achieved by incorporating a reactor freeboard section with a small internal or external demisting device. A new process design capable of operating with one or all of the process simplifications was developed. Fortunately, the PDU process flowsheet modifications were achieved without major reconstruction of existing piping.

Two-Phase Gas Holdup tests began at LaPorte in June 1988 with nitrogen/oil and CO-rich gas/oil systems. The purpose of these tests was to study the hydrodynamics of the reactor, detect metal carbonyl catalyst poisons, and train operating personnel. Any effect of the new gas sparger and the internal heat exchanger would be revealed by comparing the hydrodynamic data with previous PDU hydrodynamic data.

The results of the test showed that, under external slurry loop circulation process conditions, the presence of the new gas sparger and internal heat exchanger provided essentially equivalent gas holdup in the reactor. However, when the external liquid circulation was eliminated, gas holdup increased. Also, from available laboratory and literature data, the low carbonyl levels detected were considered satisfactory for subsequent methanol production.

The "Equipment Evaluation" Run E-5 was conducted at the LaPorte LPMEOH PDU in July, 1988. The objective of Run E-5 was to systematically evaluate each new piece of equipment (sparger, internal heat exchanger, V/L disengagement zone, demister, and cyclone) which had been added to the system, and attempt to run the reactor in an internal-only mode. In addition, a successful catalyst activation with a concentrated (45 wt% oxide) slurry was sought.

The performance of the simplified reactor system was excellent for the entire Run E-5 with only one minor setback. The first attempt at an in-situ catalyst

activation in the new reactor system resulted in only 85% catalyst activation. The activation was considered within the acceptable range and attributed to operating procedures. The remainder of the operating program was exemplary. The presence of an oil phase in the product methanol, seen in previous runs, was eliminated due to repiping of the feed/product heat exchanger and the weight percent oil in the methanol was reduced from 1.3 to 0.6%. Methanol productivity and gas holdup were higher than previous runs at high catalyst loadings. This demonstrated improvement was attributable to the new gas sparger. A higher methanol productivity was achieved without external slurry circulation in comparison to methanol productivity under the same conditions with external slurry circulation. Clearly, in the external loop configuration the catalyst did not contribute significantly to the methanol production while circulating through the slurry loop. Methanol productivity in the new simplified process was greater than autoclave performance, indicating that the design of the new system had eliminated mixing and mass transfer limitations. Also demonstrated was the resilience of the process to unscheduled shutdowns.

ACRONYMS AND DEFINITIONS

CSI	Chem Systems Inc.
DOE	U.S. Department of Energy
EPRI	Electric Power Research Institute
LPMEOH	Liquid Phase Methanol, the technology to be demonstrated
MeOH	Methanol
NDG	Nuclear Density Gauge
PDU	Process Development Unit

## TABLE OF UNITS

%	Percent
Btu	British thermal unit
cc	Cubic centimeters
cm	Centimeters
°C	Degrees Celsius
°F	Degrees Fahrenheit
ft	Feet
gal	Gallons
gm	Grams
gmol/hr Kg	Productivity units: Gram moles of methanol per hour per kilograms of catalyst oxide.
gpm	Gallons per minute
hr	Hours
in	Inches
kg	Kilograms
l	Liters
lb	Pounds
m <sup>2</sup>	Square meters
min	Minutes
mol%	Mole percent
ppb	Parts per billion by volume for gas by weight for liquid
psi	Pounds per square inch
psig	Pounds per square inch gauge
psia	Pounds per square inch absolute
scf	Standard cubic feet
SCFH	Standard cubic feet per hour
sec	Seconds
Sl/hr Kg	Space Velocity Units: Standard liters of feed gas per hour per kilograms of catalyst oxide
SV	Gas hourly space velocity Standard liters per hour per kilogram catalyst oxide
TPD	Tons per day
tons	Short tons (2000 lb)
wt%	Weight percent

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## I. EXECUTIVE SUMMARY

Since 1981, Air Products and Chemicals, Inc. has been providing research and development work to prove the feasibility of the LPMEOH process at the PDU scale as part of the U.S. Department of Energy's (DOE) indirect coal liquefaction program. Chem Systems Inc. (CSI), the inventor of the process, has been the key subcontractor in the program. Air Products has been joined by the Electric Power Research Institute (EPRI) as a private cost sharing participant. This work has been funded under the DOE contracts DE-AC22-81PC30019, DE-AC22-85PC80007, and the current contract DE-AC22-87PC90005.

The primary focus of this PDU operating program was to prepare for a confident move to the next scale of operation with an optimized and simplified process. Although the PDU had been very reliable in prior operations, several new design options had been identified and thoroughly evaluated in a detailed process engineering study which was completed under the LPMEOH Part-II contract (DE-AC22-85PC80007) and became the basis for the current PDU modification and operating programs.

### Process Engineering Design

The focus of the Process Engineering Design was to evaluate options for simplifying and optimizing the LPMEOH process studied under the previous contract. Simplifying the process had the potential to greatly improve the process economics by reducing capital costs and downtime due to equipment failure. The proposed process simplifications focused on the slurry loop, which consists of the reactor, vapor/liquid separator, slurry heat exchanger, and slurry circulation pump. The desired process change was the elimination of the entire slurry loop outside the reactor, which could be achieved if vapor/slurry separation and heat exchange were performed in the reactor vessel. This modification would be a significant process improvement since it eliminated two large high-pressure vessels (the vapor/slurry separator and the heat exchanger shell) and the slurry circulation pump. The 250 gpm centrifugal slurry pump proved to be a highly reliable piece of equipment, however, it required a great deal of preventive maintenance.

Three key conclusions were reached as a result of a detailed process engineering design evaluation: 1) there should be sufficient liquid circulation and turbulence induced by the high synthesis gas velocities to suspend the catalyst particles homogeneously within the slurry, 2) the required internal heat transfer surface should be relatively small so that sufficient heat exchanger area could be installed with little effect on reactor bubble column hydrodynamics, and 3) adequate vapor/liquid disengagement can be achieved by incorporating a reactor freeboard section with a small internal or external demisting device. A new process design capable of operating with one or all of the process simplifications was developed. Fortunately, the PDU process flowsheet modifications were achieved without major reconstruction of existing piping.

## Two-Phase Gas Holdup Test

Two-Phase Gas Holdup tests began at LaPorte in June 1988 with nitrogen/oil and CO-rich gas/oil systems. The purpose of these tests was to study the hydrodynamics of the reactor, detect metal carbonyl catalyst poisons, and train operating personnel. Any effect of the newly installed gas sparger and the internal heat exchanger would be revealed by comparing the hydrodynamic data with previous PDU hydrodynamic data. Hydrodynamic information was measured with a nuclear density gauge. The density gauge was mounted on a track which allows measurements along the axis of the reactor. Gas and liquid holdup were calculated from the absorbance of gamma radiation using Beer's law and adsorption coefficients based on calibration experiments.

Under external slurry loop circulation process conditions, the presence of the new gas sparger and internal heat exchanger provided essentially equivalent gas holdup within the reactor. However, when liquid circulation was in use, gas holdup was suppressed and when external liquid circulation was eliminated, gas holdup was higher. In addition, gas holdup profiles were uniform over the height of the reactor.

Two-phase CO-rich gas studies also served to monitor the production of catalyst poisons, particularly iron and nickel carbonyls. Initial levels of carbonyls, determined by gas chromatography and atomic absorption, were below 70 ppb. After an on-stream time of 20 hours, carbonyl levels dropped to 19 ppb of  $\text{Fe}(\text{CO})_5$  and 4 ppb of  $\text{Ni}(\text{CO})_4$ . From available laboratory and literature data, these low carbonyl levels were considered satisfactory for subsequent methanol production.

## Equipment Evaluation Run E-5

The "Equipment Evaluation" Run E-5 was conducted at the LaPorte PDU in July, 1988. A commercially available catalyst powder (F21/0E75-44) was used. This was a new batch of the same brand and type of catalyst which was used in Runs E-2, E-3, and E-4. The oil used in the catalyst slurry was Drakeol-10 in place of the Freezeze-100 oil that was used in previous runs. This was viewed as a means to qualify an alternate supplier for the white mineral oil used in the catalyst slurry. The objective of Run E-5 was to systematically evaluate each new piece of equipment (sparger, internal heat exchanger, V/L disengagement zone, demister, and cyclone) which had been added to the system, and to operate the reactor in an internal-only mode. In addition, a successful catalyst activation with a concentrated (45 wt% oxide) slurry was attempted.

The first attempt at an in-situ catalyst activation in the new reactor system resulted in only 85% catalyst activation. The level of activation was considered within the acceptable range and attributed to operating procedures and the run was continued. The Drakeol-10 oil appeared to perform as an acceptable substitute for the Freezeze-100 oil previously used.

Production of methanol was stable in Run E-5A after only 18 hours on-stream with syngas. The presence of an oil phase in the methanol seen in previous runs was eliminated due to repiping of the feed/product heat exchanger. The weight percent oil in the methanol was reduced from 1.3 to 0.6%. Methanol productivity and gas holdup were higher than previous runs at high catalyst



loadings. This demonstrated improvement was attributed to the new gas sparger; however, methanol productivity was still slightly below the autoclave curve for high loadings.

The new internal heat exchanger was evaluated in Runs E-5B and E-5C. Again, the reactor performed close to the autoclave results. The overall heat transfer coefficient of the heat exchanger was 74 Btu/hr-ft<sup>2</sup>-°F, close to the design value of 94 Btu/hr-ft<sup>2</sup>-°F. Operation at high space velocities was not possible with the internal heat exchanger because its area was consciously under-designed to achieve measurable temperature differences with a 35 wt% catalyst slurry where a lower temperature differential existed between slurry and heat transfer oil. There was a concern that large temperature differences at 45 wt% slurry and high gas velocities could produce excessive thermal stresses. Therefore, both internal and external heat exchangers were used in Run E-5C to remove the heat of reaction. Daily methanol production ranged from 8 to 9 TPD for the 45 wt% runs, which exceeded the previous production rates of 5 to 7 TPD at 47 wt% slurry.

Run E-5D was the first test of the LaPorte reactor without external circulation. A higher methanol productivity resulted without external slurry circulation compared to the exact same conditions with external slurry circulation in Run E-5B. Clearly, the catalyst in the external loop configuration did not contribute significantly to the methanol production while circulating in the loop external to the reactor. The internal slurry/gas disengagement and demister performed well.

Runs E-5E and E-5F were completed at the end of July with 34 wt% catalyst slurry. Methanol productivity was apparently greater than autoclave performance, indicating that the design of the new system must have eliminated mixing and mass transfer limitations. The overall heat transfer coefficient for the 34 wt% slurry was 95 Btu/hr-ft<sup>2</sup>-°F. Productivity levels achieved in Run E-5F demonstrated that the process was resilient in handling deliberate shutdowns of 1 hour and 24-hour durations. Four unplanned power outages during the month of July also demonstrated the reliability of this process after unscheduled shutdowns.

In summary, the performance of the simplified reactor system was excellent for the entire Run E-5. Additional process changes resulted in reducing slurry oil losses by 75% and substantial improvements in methanol quality. This overall design represents the state-of-the-art for the LPMEOH Process and will be used as the baseline to judge potential performance improvements in the future.

## II. INTRODUCTION AND BACKGROUND

The LPMEOH process was conceived and patented by Chem Systems Inc. in 1975. Initial research and studies on the process focused on two distinct modes of operation. The first was a liquid fluidized mode with relatively large catalyst pellets suspended in a fluidizing liquid, and the second was an entrained (slurry) mode with fine catalyst particles slurried in an inert liquid. The development of both operating modes progressed in parallel from bench scale reactors, through an intermediate scale lab PDU, and then to the LaPorte PDU in 1984. The slurry mode of operation was ultimately chosen as the operating mode of choice due to its superior performance.

Development efforts on the slurry reactor have continued through 1988 both at the LaPorte PDU and in the laboratory. The work done during this period has focused on optimizing all aspects of the LPMEOH process. The key milestones in the development and scale-up of the current LPMEOH process are listed below:

<u>Date</u>	<u>Development Scale and Key Results</u>
1975	LPMEOH concept patented by Chem Systems
1979 - 1981	2 Liter bench scale stirred autoclave Concept Verification Catalyst/Oil Screening Initiated Catalyst Activation Methods Demonstrated
1981-1988	300 cc/liter laboratory scale stirred autoclave Method to Activate Conc. Slurries Developed Catalyst Poisons Studied Feed Gas Composition Effects Determined
1983	4.5 in ID x 7 ft tall Lab. PDU (up to 0.14 TPD MeOH) Reactor Productivity Defined Hydrodynamic Behavior Identified Reactor Modeling Begun
1984 - 1985	2 in ID x 18 ft tall LaPorte PDU (up to 8 TPD MeOH) Reactor Performance Demonstrated Catalyst Life Demonstrated Materials of Construction Defined Operating Experience Base

### A. Process Development Scale Experience

The primary function of the LaPorte PDU is to acquire data using a small, yet representative engineering scale for testing the feasibility of the LPMEOH process. Thus, the PDU was designed to generate and collect plant data over a wide range of operating conditions. The range of operating variables chosen for the original design is shown in Table II.1. In fact, the PDU has operated at flow, space velocity, and catalyst loading conditions well in excess of design.

TABLE II.1

RANGE OF OPERATING VARIABLES FOR LAPORTE PDU

	<u>Minimum</u>	<u>"Typical"</u>	<u>Maximum</u>
Reactor Pressure, psig	500	750	900
Reactor Temperature, °C	220	250	270
°F	428	482	518
Liquid-Fluidized Space Velocity, liter/hr-kg cat	1,000	2,500	4,000
Liquid-Entrained Space Velocity, liter/hr-kg cat	2,000	6,000	10,000
Liquid-Fluidized Catalyst Loading, Settled Bed Height, ft	5	7	7
Liquid-Entrained Catalyst Loading, wt%	10	20	33

NOTE: Space velocity based on standard liters (0°C, 14.7 psia), kg of oxide catalyst, and zero gas holdup in reactor.

The principal reactor feed gas compositions considered during design were:

- CO-Rich Type, in which the H<sub>2</sub> and CO concentrations were not stoichiometrically balanced, but were representative of synthesis gas directly from a modern Texaco coal gasifier. This gas is typical of that for once-through methanol synthesis in a CGCC plant configured to make electric power and coproduct methanol.
- Balanced Type, representative of CO-Rich gas which has undergone shift and CO<sub>2</sub> rejection so that the H<sub>2</sub> and CO concentrations were approximately stoichiometrically balanced (2/1) in order to achieve an "all-methanol" product.
- H<sub>2</sub>-Rich Type, in which the H<sub>2</sub> and CO concentrations were not stoichiometrically balanced, but were representative of synthesis gas from a steam methane reformer.

The compositions of the various gas streams are given in Table II.2. The different reactor feed gas compositions were blended from H<sub>2</sub>, CO, N<sub>2</sub>, and CH<sub>4</sub> supplied by Air Products' adjacent syngas facility at LaPorte. Carbon dioxide is trucked into the plant as a liquid and stored on-site. Since only a portion of the reactor feed is converted per pass, the unconverted synthesis gas is recycled and mixed with fresh makeup gas. The makeup gas is blended so that the reactor feed (makeup plus recycle) simulates either the balanced or CO-rich gas type in once-through operation. Recycling the unconverted synthesis gas reduces gas consumption by 70% for cost-effective operation at LaPorte.

TABLE II.2  
GAS TYPES TESTED AT THE LAPORTE PDU

<u>Component (mol%)</u>	<u>CO-Rich Gas</u>	<u>Balanced Gas</u>	<u>H2-Rich Gas</u>
Hydrogen	35.0	55.0	71.0
Carbon Monoxide	51.0	19.0	18.0
Carbon Dioxide	13.0	5.0	7.0
Methane	0.1	0.1	0.1
Nitrogen/Inerts	0.9	20.9	3.9
H <sub>2</sub> /CO Ratio	0.69	2.89	3.94
$\frac{(H_2 - CO_2)}{(CO + CO_2)}$	0.34	2.08	2.56

B. LPMEOH I and II Operations

A total of five major synthesis runs were conducted at the LaPorte PDU from March 1984 through July 1985. A summary of these campaigns is presented in Table II.3.

TABLE II.3  
LAPORTE PDU OPERATIONS SUMMARY

<u>Run #</u>	<u>Date</u>	<u>Objective</u>	<u>Operation Mode</u>	<u>Catalyst Type</u>	<u>Hours On Syngas</u>
F-1	Mar 84	Shakedown	Hybrid Fluidized/Slurry	Extrudates	248
E-1	Apr/May 84	Activity Maintenance	Hybrid Fluidized/Slurry	Extrudates	964
E-2	Jun 84	High Slurry Conc., High Throughput	Slurry	Powder	145
E-3	May/Jun 85	Activity Maintenance	Slurry	Powder	948
E-4	Jul 85	High Slurry Conc., High Throughput	Slurry	Powder	<u>231</u> 2536

The first PDU run (F-1) was a 10-day shakedown run. The PDU operated smoothly, and the mechanical integrity and process flexibility of the unit were demonstrated. Up to 8 TPD of methanol were produced. The second PDU run (E-1) was a 40-day continuous run on CO-rich synthesis gas ( $H_2/CO=0.7$ ). Stable operation was achieved, but a slow, continuous decline in catalyst activity was observed, in excess of that anticipated from isothermal laboratory autoclave experiments. The accumulation of trace poisons seen on the catalyst was the major cause of this loss of activity (1.1% per day). A third PDU run (E-2) was conducted for 6 days using a commercially available catalyst powder at very high slurry concentration (up to 45 wt%). In-situ activation was performed. The plant operated well mechanically, providing valuable experience for the operations and engineering staff in handling high-viscosity catalyst slurries. Methanol productivity, however, was below the values predicted from laboratory autoclave results. A supporting laboratory program funded by the Electric Power Research Institute (EPRI) determined that inadequate catalyst activation at LaPorte was the reason for the off-performance at the high solids loading in Run E-2. Changes in the activation procedure were identified to remedy this problem.

Analysis of the results of the 1984 operating program indicated that selective upgrading of materials of construction of the PDU would lead to lower levels of trace contaminants. Process improvements that would increase the data gathering capability were also specified. As a result, modifications were made to the LaPorte PDU in early 1985. New equipment was installed to improve the measurement of slurry concentration and methanol product flow. Also, selected vessels and piping were replaced or modified in order to reduce the levels of trace catalyst poisons, primarily iron and nickel carbonyls formed by the reaction of CO with the carbon steel pipe walls. A chemical cleaning program was also undertaken to remove residual contaminants.

Upon completion of these activities, a second 40-day activity maintenance test (Run E-3) using CO-rich gas and a 25 wt% catalyst slurry was conducted in May-June 1985. The reactor conditions matched the earlier extended operating campaign at the PDU (Run E-1). In Run E-3, however, the revised in-situ catalyst activation procedure was successfully applied and, combined with the new metallurgy, the reactor performance matched laboratory predictions for catalyst life and activity. High onstream reliability for the PDU was achieved. The improved performance and catalyst activity maintenance of Run E-3 versus Run E-1 is shown in Figure II.1.

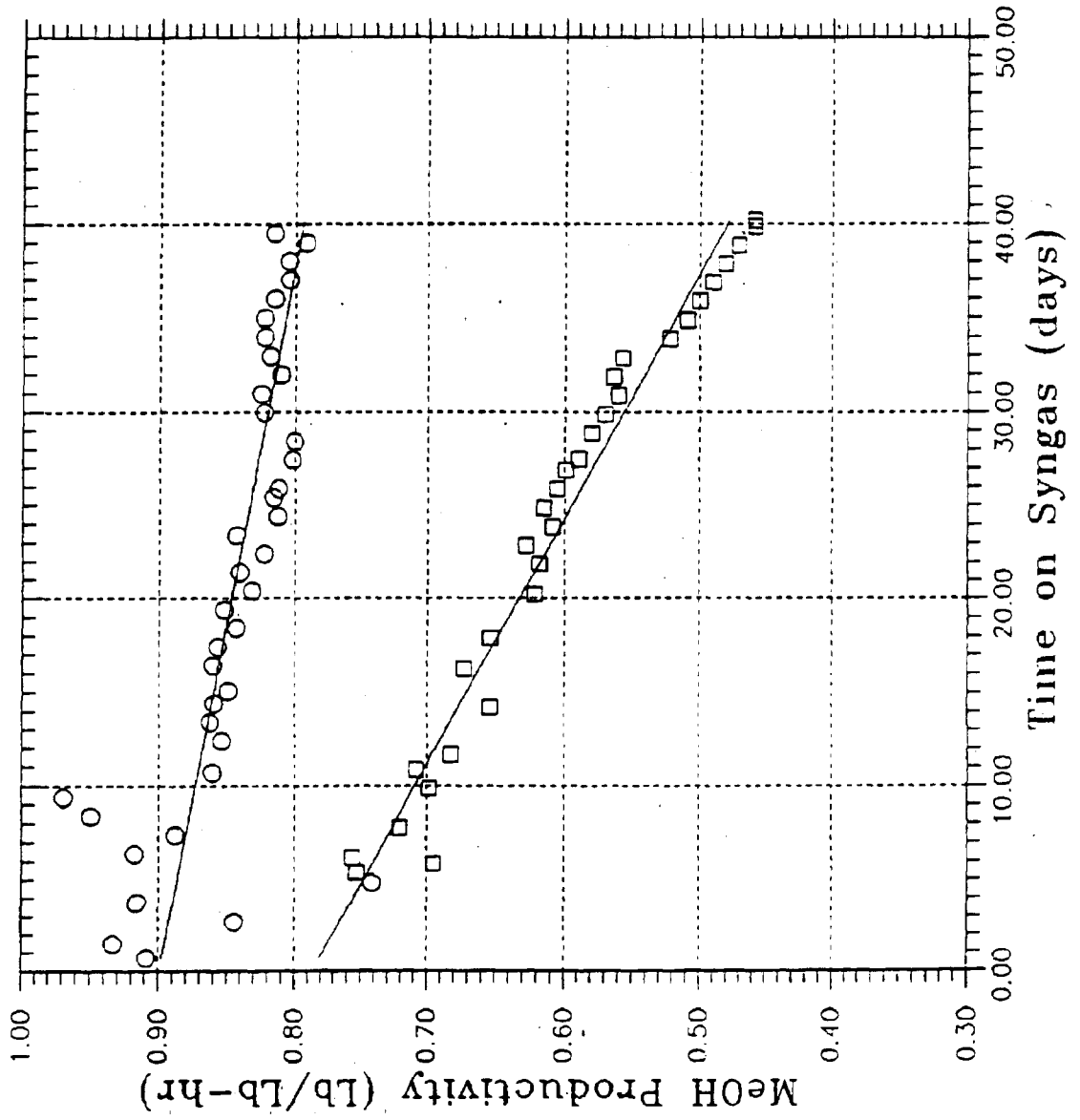
A second operation of the LaPorte PDU at elevated slurry concentrations (again up to 45 wt%) was subsequently performed. A successful catalyst activation was achieved. High operability was again maintained during this 10-day test; the reactor performance exceeded the previous run at these conditions (Run E-2) but catalyst productivity was still less than laboratory predictions. The deficiency in productivity was attributed to a mass transfer limitation and/or inadequate gas/slurry mixing or distribution at these elevated slurry solid loadings.

LaPorte PDU operations successfully demonstrated LPMEOH technology at a representative engineering scale. The PDU accumulated over 2500 hours of methanol synthesis operation with an on-stream factor of 96-100%. Low catalyst deactivation while operating the liquid-entrained system with a 25 wt% catalyst slurry for an extended period of time on CO-rich synthesis gas

Figure II.1

# LAPORTE LPMEOH PDU REACTOR PERFORMANCE

Runs E-1&3: CO-Rich Gas, 750 psig, 482°F



## LEGEND

- Run E-3 - 1985
- Run E-1 - 1984

was a notable achievement. The ability to activate methanol synthesis catalyst powders in an inert liquid at high concentrations was also noteworthy. Methanol production levels as high as 8 TPD for balanced gas feed and 7 TPD for CO-rich gas feed were achieved; the purity of the methanol product from CO-rich gas was consistently higher than 96% wt%, a good fuel-grade quality.

### III. LAPORTE LPMEOH PDU

#### A. Existing PDU Process Description

A simplified process flowsheet for the LaPorte PDU is shown in Figure III.1. The makeup synthesis gas is compressed to the reactor pressure (500-900 psig) by the feed compressor. The compressed makeup and recycle gases are mixed and preheated in the feed/product exchanger before being fed into the methanol reactor. The inert hydrocarbon liquid or slurry that circulates through the reactor is separated from the unconverted synthesis gas and methanol product vapor in the primary V/L separator, and recirculated to the reactor through the slurry heat exchanger. The circulating liquid or slurry can be heated or cooled in the slurry exchanger to maintain a constant reactor temperature, depending upon the level of conversion, system heat losses, and the rate of cold seal flush required by the slurry pump. A utility oil system provides the heating or cooling duty to the slurry exchanger.

The unconverted synthesis gas/product methanol stream leaving the primary V/L separator is cooled against incoming feed gas and the condensed oil is separated in the secondary V/L separator. The uncondensed vapor is further cooled in the product cooler. Condensed methanol is then separated from the synthesis gas and additional condensed oil before being piped to product storage. A small purge stream is sent to flare. The bulk of the unconverted synthesis gas is compressed and returned to the front end of the PDU. Additional systems are present to activate the catalyst, provide seal flush to the slurry pump, and mix the catalyst slurry.

A schematic of the LaPorte reactor showing key dimensions and the location of temperature sensors is given in Figure III.2. Both the feed gas and the recirculated slurry enter a plenum chamber in the bottom of the reactor. This mixture then enters the reaction zone through a bubble cap tray distributor. The three-phase slurry flows concurrently through the reactor and exits to a separate vapor/liquid disengagement vessel.

An external nuclear density gauge is used to monitor the catalyst bed height in the reactor during the liquid-fluidized mode of operation. The gauge is mounted in a mechanical framework which allows it to traverse the reactor in the vertical direction. During the slurry operation, the gauge is used to directly measure three-phase density and subsequently determine hydrodynamic information about the reactor.

#### B. New PDU Process Design

Although the PDU was very reliable in prior campaigns, several new design options were identified and thoroughly evaluated in a detailed process engineering study completed under the LP-II contract (DE-AC22-85PC80007), which became the basis for the current PDU modification/operating program.

The Process Engineering studies done under the LP-II contract examined the feasibility of several proposed modifications to the LaPorte PDU reactor section. These modifications included a continuous catalyst addition/withdrawal system to maintain catalyst activity, replacement of the existing external loop with an internal heat exchanger, use of an agitator for mass



Figure 1.1

# SIMPLIFIED PROCESS FLOWSHEET FOR LAPORTE PDU

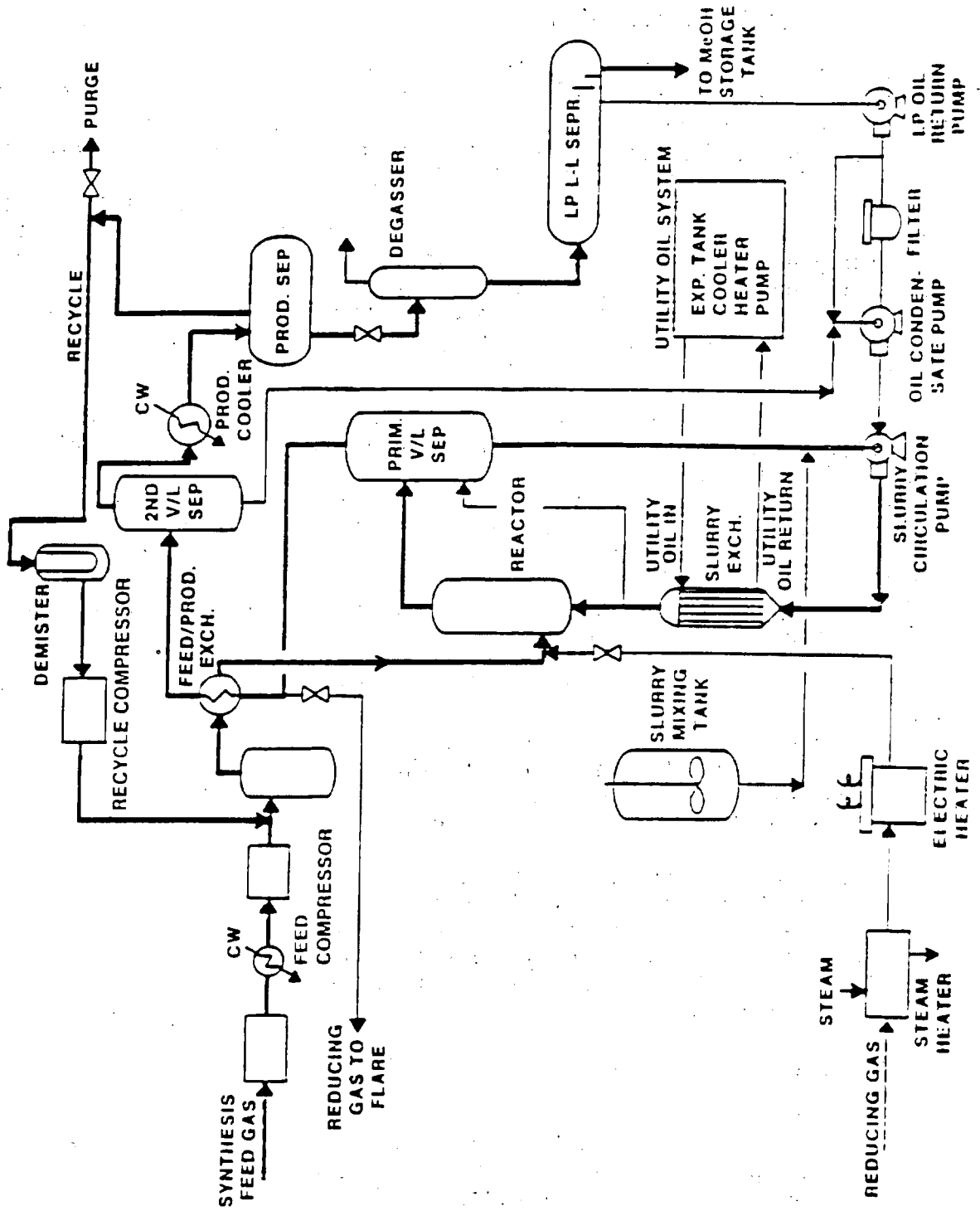
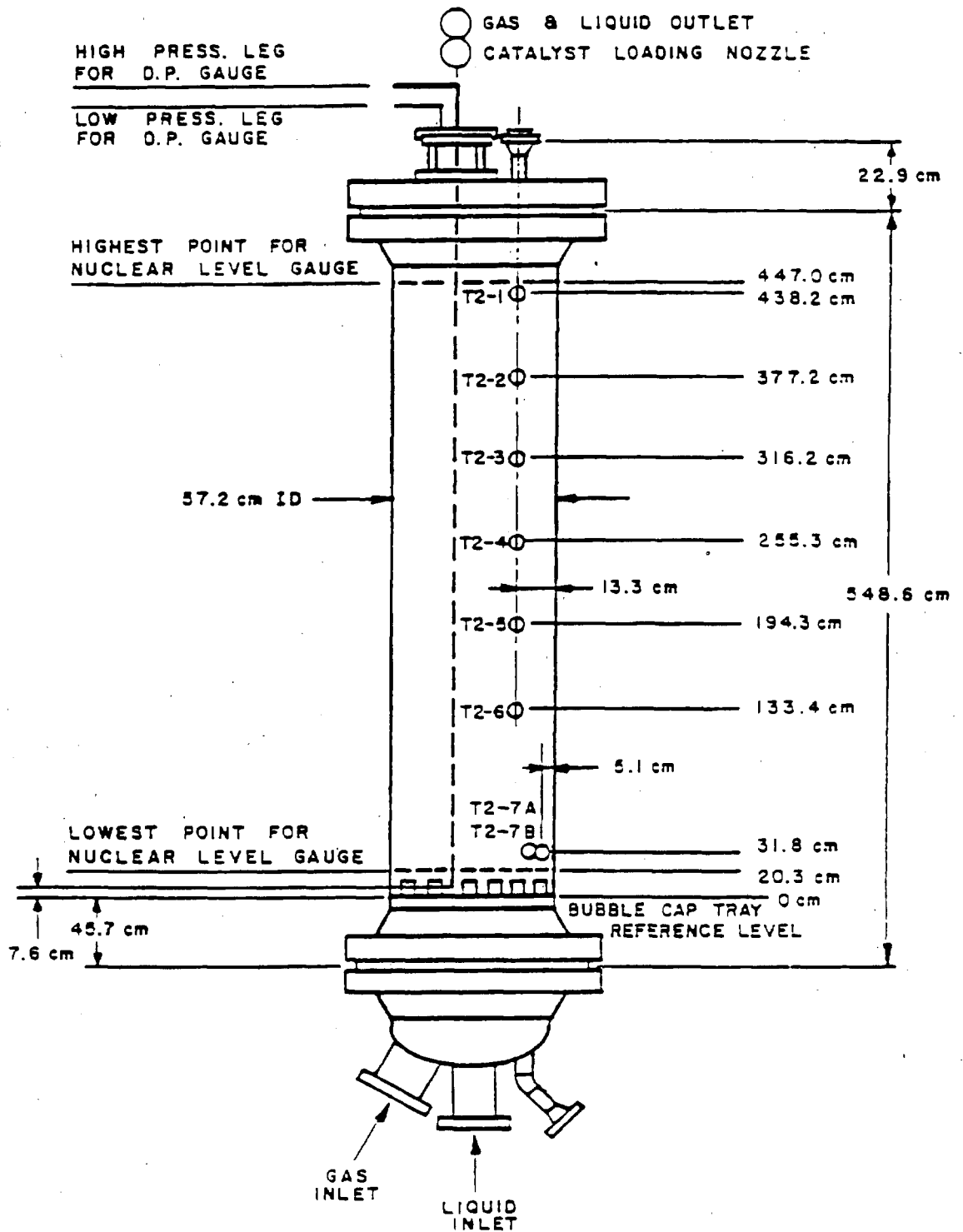


Figure III.2

**LPMEOH PDU REACTOR**



transfer enhancement with concentrated slurries, extension of the reactor height to achieve vapor/liquid disengagement, and installation of either a cyclone or demister to minimize slurry carry-over from the reactor. (See Final Report of DE-AC22-85PC80007 for details of Process Engineering Studies.) The key conclusions reached as a result of the detailed process engineering evaluation were:

- 1) A catalyst addition/withdrawal system has good merit and is worthy of consideration in the continuing PDU operating program.
- 2) The required internal heat transfer surface should be relatively small so that sufficient heat exchanger area could be installed with little effect on reactor bubble column hydrodynamics.
- 3) While an agitator would probably yield the desired enhanced mixing and mass transfer effects, the high cost of testing the unit at the LaPorte PDU and the risk associated with the mechanical seal operating at the severe LPMEOH reactor conditions cannot be justified.
- 4) Adequate vapor/liquid disengagement can potentially be achieved by incorporating a reactor freeboard section.
- 5) An external cyclone and a reactor internal demister should be installed and examined separately in providing extra gas/slurry separation in conjunction with the primary separation step.

Simplifying the process had the potential to greatly improve the process economics by reducing capital costs and the amount of downtime due to equipment failure. The proposed process simplifications were evaluated along with the idea that sufficient liquid circulation and turbulence should be induced by the high synthesis gas velocities to suspend the catalyst particles homogeneously within the slurry without using the external slurry pump. The desired process simplification was the elimination of the entire loop outside the reactor, which could be achieved if vapor/slurry separation and heat exchange were performed in the reactor vessel. This modification would be a significant process improvement since it eliminated two large high-pressure vessels (the vapor/slurry separator and the heat exchanger shell) and the slurry circulation pump. The 250 gpm centrifugal slurry pump proved to be a highly reliable piece of equipment, however, it required a great deal of preventive maintenance.

The slurry pump influences liquid mixing in the reactor by providing a net upward slurry circulation rate through the reactor. This net flow is in addition to the internal circulation within the reactor, which is induced by the high superficial gas velocity through the reactor. In order to determine the impact of elimination of the slurry pump on the liquid mixing, the internal slurry circulation was estimated using bubble column models developed by Clark, et al. (1987) and Kawase, et al. (1986). The predictions from each of these models are shown in Figure III.3. Although the quantitative results from each of the models vary, the conclusions reached are the same for both. The gas induced net liquid velocity is 0 ft/sec, but the upward velocity at the centerline of the reactor is 2.0 to 2.5 ft/sec, and near the walls the downward velocity is -1.0 to -2.0 ft/sec. By comparison, the pump induced