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LIQUID PHASE METHANOL PROCESS DEVELOPMENT UNIT: INSTALLATION, OPERATION, AND SUPPORT STUDIES

Technical Progress Report No. 1

For the Period 28 September 1981 - 31 December 1981

Contractor

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and

Subcontractor to APCI

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ABSTRACT

This was the first period of Contract performance. During this period the Work Breakdown Structure Dictionary was established. Task 1 was completed with submittal of the Project Work Plan and the Quality Assurance Manual. CSI produced basic process design information and a preliminary flowsheet for the LaPorte LPMeOH PDU. APCI developed the flowsheet further and set up the process on APCI's process simulator. The flowsheet development revealed a number of major changes necessary in the existing LPM pilot plant; this has lead to pursuit of a "unified design" concept. This design strategy addresses all modes of operation at the start of the design effort, and results in a single, unified PDU. Approval was requested for the unified design concept as well as advanced schedule for relocation of the LPM unit and advanced procurement of long delivery equipment items. A number of preliminary heat and material balances were calculated for the LPMeOH PDU and preliminary process specifications were prepared for the equipment items. The final design basis was established. The design pressure was set at 1000 psig. Eight design operating cases were defined for the following range of reactor operating conditions: Pressure - 500 to 900 psig, Temperature - 220 to 270°C, Liquid-Fluidized Space Velocity - 1000 to 4000 l/hr-kg catalyst, Liquid-Entrained Space Velocity - 2000 to 10,000 l/hr-kg catalyst, and Liquid-Entrained Catalyst Loading - 0.1 to 0.4 kg catalyst/l oil. The methanol production rate for these cases ranges from 0.2 to 9.7 short tons per day. Preliminary equipment arrangement and site layout drawings were prepared for the PDU. In the laboratories, CSI began autoclave testing of in-situ catalyst reduction procedures. The specification and evaluation of equipment for the CSI laboratory PDU progressed. CSI prepared and issued a Topical Report covering liquid-entrained LPMeOH lab development work accomplished under advance funding. APCI's laboratories progressed with the design of the bench scale slurry reactor.

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PROJECT DESCRIPTION:

On 28 September 1981, Air Products and Chemicals, Inc. (APCI) began a 42-month contract with the U.S. Department of Energy (DOE): "Liquid Phase Methanol (LPMeOH) Process Development Unit: Installation, Operation and Support Studies." This project is aimed to further develop the LPMeOH process invented by Chem Systems Inc. (CSI). Chem Systems is performing as a subcontractor to Air Products.

A DOE-owned, skid mounted pilot plant will be transferred from Chicago, refurbished, expanded for service as the LPMeOH Process Development Unit (PDU), and then relocated to Air Products' LaPorte, Texas facility. Air Products will supply synthesis feed gas to the LPMeOH PDU and operate the unit for a planned 24-month period. Chem Systems is performing the major portion of the laboratory support R&D and is providing technical management for the project. Air Products is providing overall program management and is responsible for engineering design, construction, and operation.

The program is divided into 11 major tasks which are phased to allow progress review and approval to proceed. The 11 major tasks are:

1. Program Planning
2. Engineering and Design Specifications
3. Equipment Procurement
4. LPM Pilot Plant Relocation
5. LaPorte LPMeOH PDU Renovation, Installation and Shakedown
6. Liquid-Fluidized Operation
7. Laboratory Support Program
8. LaPorte LPMeOH PDU Modifications for Liquid-Entrained Mode
9. Shakedown of the Liquid-Entrained Mode of Operation
10. Liquid-Entrained Operation
11. Project Evaluation

The tasks are phased as follows:

Phase	Tasks	Schedule
I	1,2,7,11	28 September 1981 - 28 March 1985 (Months 1-42)
II	3,4,5	1 June 1982 - 1 April 1983 (Months 9-18)
III	6	1 March 1983 - 1 June 1984 (Months 18-32)
IV	8	1 April 1983 - 1 April 1984 (Months 19-30)
V	9,10	1 April 1984 - 1 March 1985 (Months 31-41)

OBJECTIVES:

The overall objective of this program is to demonstrate the technical feasibility of the Liquid Phase Methanol (LPMeOH) process at the Process Development Unit (PDU) scale of operation.

On a per task basis, objectives are to:

Task 1 - Program Planning

Establish a Project Work Plan presenting in detail all activities which will be performed for the successful completion of the program.

Task 2 - Engineering and Design Specifications

a) Conduct a process engineering/design review and safety examination of the existing Liquid Phase Methanation (LPM) pilot plant at its present location in Chicago, Illinois;

b) Obtain permits to install and operate the LPMeOH PDU at LaPorte, Texas;

c) Develop detailed plans and specifications for the repair, modification, and expansion of the existing LPM unit to enable liquid-fluidized (ebullated bed) and subsequently liquid-entrained (slurry) methanol production; and

d) Develop a deactivation plan for the LaPorte LPMeOH PDU.

Task 3 - Equipment Procurement

Purchase, lease, or obtain from DOE inventories the equipment and systems specified in Task 2.

Task 4 - LPM Pilot Plant Relocation

Transfer the existing LPM pilot plant from its present location in Chicago to a vendor's facility for renovation, and subsequently to APCI's LaPorte, Texas site.

Task 5 - LaPorte LPMeOH PDU Renovation, Installation, and Shakedown

a) Renovate the LPM pilot plant to become the LaPorte LPMeOH PDU, according to the specifications developed in Task 2;

b) Prepare the LaPorte site;

c) Install the LPMeOH PDU at LaPorte;

d) Make interconnections and test components; and

e) Conduct an integrated run without catalyst.

Task 6 - Liquid-Fluidized Operation

Operate in the liquid-fluidized (ebullated bed) mode to:

a) Assess the effect of reactor configuration/internals;

b) Identify catalysts which in short runs have acceptable activity and attrition characteristics;

c) Perform process variable scans to determine the effects of temperature, pressure, space velocity, catalyst loading, circulating oil flowrate, and feed gas composition; and

d) Perform a 45-day continuous run to demonstrate short-term process operability, principally at a single set of conditions.

Task 7 - Laboratory Support Program

a) Conduct literature surveys, develop a bibliography of pertinent references, and maintain liaison with others working on related liquid-entrained (slurry) systems:

b) Develop procedures for in-situ reduction of commercial powdered methanol catalysts slurried in oil, vapor phase reduction of commercial granular materials which can subsequently be slurried, and simultaneously screen commercial catalysts and develop data for modeling the liquid-entrained reaction;

c) Synthesize new liquid-entrained catalysts;

d) Screen new liquid-entrained catalysts in a gas phase fixed bed reactor, a liquid-entrained autoclave reactor, and in the Chem Systems' Fairfield laboratory PDU;

e) Construct and operate a cold flow model unit to study the hydrodynamics of the gas-slurry reactor;

f) Modify the existing Chem Systems' Fairfield laboratory PDU to allow liquid-entrained as well as liquid-fluidized operation;

g) Operate the modified CSI lab PDU to perform process variable scans; and

h) Support the LaPorte LPMeOH PDU liquid-fluidized and liquid-entrained operating modes, principally by screening catalysts.

Task 8 - LaPorte LPMeOH PDU Modifications for Liquid-Entrained Mode

Construct a separate skid with equipment necessary for operation of the LPMeOH PDU in the liquid-entrained (slurry) mode, relocate it to the LaPorte site, and perform tie in with the existing PDU.

Task 9 - Shakedown of the Liquid-Entrained Operation

Test components, conduct an integrated run with an inert powder, and conduct an integrated short run with a liquid-entrained methanol catalyst.

Task 10 - Liquid-Entrained Operation

a) Conduct short runs with promising liquid-entrained catalysts;

b) Perform process variable scans to determine the effects of various operating conditions; and

c) Perform a 45-day continuous run to demonstrate short-term process operability.

Task 11 - Project Evaluation

a) Evaluate data from the LaPorte LPMeOH PDU and the laboratories to develop models;

b) Evaluate alternative reactor designs and the two operating modes;

c) Perform detailed process evaluations for commercial-size plants;

d) Develop plans for a larger scale demonstration of the LPMeOH process; and

e) Report on program activites.

RESULTS:

Task 1 - Program Planning

1.1* APCI Management Activites

1.1.1 Project Management - A Work Breakdown Structure (WBS) Dictionary was established which divides the program effort into discrete increments which satisfy reporting requirements. A revised WBS Dictionary incorporating DOE comments was submitted to DOE on 9 November 1981. The cost collection and control system was established. The Project Work Plan and the Quality Assurance Manual were prepared and submitted to DOE on 4 December 1981. Engineering manhour estimates were compiled to support preparation of the manpower and cost plans reflected in the Project Work Plan. This task is completed, subject to DOE approval of the Plan and QA Manual.

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*Refers to Work Breakdown Structure elements.

1.2 CSI Activities

1.2.1 The development of the CSI Project Work Plan was completed and transmitted to APCI on schedule by the 20 November due date. The CSI portion of the Work Breakdown Structure Dictionary was completed. Also, manpower plans, milestone schedules and cost plans as well as a management control system description were prepared and transmitted to APCI on schedule. Internal procedures for project control were established and implemented. This task is now completed.

Task 2 - Engineering and Design Specifications

2.1 APCI Management Activities

2.1.1 Project Management - Coordination was provided for a number of preliminary engineering activities. These included flowsheet development, preparation of process equipment specifications, establishment of the design basis, equipment arrangement, site layout drawings, and LaPorte tie-in requirements.

Contract change requests were submitted to DOE on 11 December 1981 for approval of: a) the concept of a single "unified" design for the LaPorte LPMeOH PDU (the technical benefits of this approach are discussed in section 2.3.1), b) advanced schedule for the dismantling, relocation, and inspection of the existing LPM pilot plant, and c) advanced commitments to purchase specific long lead equipment items (feed gas/recycle compressor and slurry circulation pumps).

The Process Hazards Review team was established.

Preparation began on the specification for disassembly of the LPM pilot plant.

2.1.2 Economic Evaluation - Engineering manhour estimates were compiled and a revision P1 internal Scope Report was prepared. Assistance was provided Project Management with cost impact data supporting the contract change requests submitted to DOE.

2.1.3 Communications - No Significant Activity.

2.1.4 Permit Fees - No Activity.

2.2 CSI

2.2.1 CSI Assistance to APCI

Upon project initiation, CSI provided APCI with basic process design information to facilitate the LaPorte LPMeOH PDU design in the liquid-fluidized and liquid-entrained modes of operation.

CSI recommended consideration of a single unified design for the LPMeOH PDU which would be capable of operating in both modes. This concept has more technical merit than separate units for each operating mode because it enhances operating flexibility while limiting capital costs. A preliminary process flowsheet was developed for the unified design and transmitted to APCI on 9 October 1981. (An updated version of this flowsheet is cited in section 2.3.1.)

A series of process design bases and reactor mass balances were generated for eight LPMeOH PDU operating conditions including both liquid-fluidized and liquid-entrained modes. Three feed gases were considered:

- Lurgi-type, single pass
- K-T-type, single pass
- Balanced-type, recycle

The nominal compositions of these feeds, shown in Exhibit 2.2.1-1, represent synthesis gases from coal gasification processes after acid gas removal and condensation of water. The Balanced-type feed for recycle operation is representative of shifted gas from a high temperature, entrained coal gasifier. From the standpoint of the feed gas entering the LPMeOH reactor, the Balanced-type gas with recycle is similar to the Lurgi-type single pass feed.

APCI was provided with ranges of operating variables suggested for the LaPorte LPMeOH PDU design. (The final operating ranges are cited in section 2.3.1.) A typical design basis for one of the liquid-entrained cases with a Lurgi-type feed gas is presented in Exhibit 2.2.1-2 and the resulting reactor mass balance is shown in Exhibit 2.2.1-3.

2.2.2 Data Acquisition

Preliminary system design and cost estimating were begun to facilitate the program planning effort in Task 1. This system will include a gas chromatograph, a liquid chromatograph, a data logger, a microcomputer and a gas sampling system. The following paragraphs describe the system being considered for the LaPorte LPMeOH PDU.

Gas analyses will be performed on an hourly basis on, at least, two process streams: reactor feed gas and cold effluent gas. If feasible, the hot reactor effluent gas will also be analyzed frequently. The gas sampling system will include timers and automatic switches so that these analyses can proceed virtually unattended. The major equipment items for on-line gas analyses include a chromatograph with thermistor - and filament-type detectors, an electronic integrator and a strip-chart recorder.

The gas components to be analyzed include carbon monoxide, carbon dioxide, methane, hydrogen, nitrogen, methanol, water and, possibly, argon. In addition to the frequent sampling mentioned above, several streams will be monitored on a demand basis. These include fresh feed gas, methanol degasser vapors, catalyst reduction feed gas, and reduction effluent gas. Continuous gas monitors can be used to observe dynamic system behavior, thus reducing the frequency of chromatographic analyses on these streams.

A second chromatograph will be employed for liquid samples, primarily the methanol product. The components to be analyzed include light hydrocarbons, alcohols, esters, aldehydes and water. Up to 20 components will be analyzed. Additionally, this chromatograph may be used on a demand basis to analyze oil samples including make-up oil, condensed oil return and hot circulating oil. These oil analyses may include boiling point curves as well as the component analyses mentioned above. The major equipment items for liquid analyses include a chromatograph with a differential-type detector containing four filaments, an integrator and a strip-chart recorder.

The data logger must monitor and store the temperature, pressure and flow data signals received from the LPMeOH PDU. The minimum number of signals necessary for processing material balances is approximately 35, according to the tabulation in Exhibit 2.2.2-1. This excludes compositional data from the gas and liquid analyses. The data logger will, thus, be sized for 100 channels with programming for averaging and other simple calculations.

A small computer will be utilized to perform material balances. The data logger will dump information into the computer on a regular basis. Analytical results will flow from the integrator to the computer either manually or automatically, depending upon the system chosen. The computer will print out results for LaPorte LPMeOH PDU operation on a regular basis.

Once the APCI revision 0 flowsheet for the LaPorte LPMeOH PDU becomes available, work will begin on specifying instrumentation and data points. In the interim, vendor inquiries will be made to ascertain the type of equipment, as well as its availability and cost, which should best fit the requirements for the LaPorte LPMeOH PDU.

2.3 APCI Design

2.3.1 Integration - CSI's preliminary Process Flowsheet for the LaPorte LPMeOH PDU submitted 9 October 1981 was reviewed and developed further. The latest Process Flowsheet, Revision P1, dated 5 January 1982 is attached as Exhibit 2.3.1-1. A narrative providing background on the development of this flowsheet is given below:

1. Feed Gas Supply

   The simulated synthesis feed gas will be compressed from 150 psia to the LPMeOH loop pressure (about 550 to 950 psia) by the 01.10 feed gas compressor. Synthesis gas compositions will simulate gas from the Lurgi-type and Koppers-Totzek (KT) - type gasifiers without shift (for single pass conversion in the LPMeOH loop), and the high temperature-type gasifiers with shift (so called "Balanced" gas, for recycle operation of the LPMeOH loop).

   Available feed gas components are H_2, CO, CO_2, N_2, and CH_4. The feed methane component is expected to act as an inert in the methanol reaction, however, its presence better characterizes "low temperature" coal gasifier product streams. The proposal for this project considered a natural gas supply from APCI's LaPorte commercial facilities to the LPMeOH PDU. A passive adsorption desulfurizer (not shown on Process Flowsheet) will be required to condition this stream.

2. Feed/Recycle Compressor (01.10/01.20)

   Early in the preparation of heat and material balances for the LaPorte LPMeOH PDU, it became necessary to consider a recycle compressor rather than imposing highly variable conditions to the final stage of feed gas compression or taking the penalty in power costs associated with recompressing the recycle flow from inlet feed gas pressure.

   Based on initial specifications and vendor scanning, it is apparent that the feed gas and recycle compression will be performed on a single reciprocating compressor frame having three cylinders. Two cylinders will perform the two stages of feed gas compression and the third will provide the necessary recompression for the recycled process gas. Both services will be provided with 50% unloading capability with fine capacity control accomplished by recycling from the discharge to the suction of each compression service.

3. Reactor (27.10)

   The existing reactor from the LPM pilot plant is considered for utilization in both the liquid-fluidized and liquid-entrained modes. This differs from the proposal which considered the addition of a new reactor skid as part of the liquid-entrained modifications. Reactor internals may be modified with inserts as necessary for the various operating modes.

4. Vapor-Liquid Separation (27.13)/(27.14)

Due to significant concern over the oil-methanol separation originally planned for the 3-phase separator (22.10) the use of the existing V/L separator from the LPM pilot plant is planned at an intermediate point in the product gas cooling. It is calculated that 75% to 80% of the oil vapor leaving the primary V/L separator (27.13) will be collected at this point. The greatest risk in the 3-phase separator is the potential for formation of an emulsion which would have to be disposed of with the methanol product. Such a condition would reduce the desirability of this methanol product for disposition, and result in an increased oil make-up requirement. Both of these factors will be considered in economic justification of this installation.

A simple condensate trap installation will be used to control the flow of oil from this intermediate separator (27.14) to the oil chamber in the 3-phase separator.

A new primary vapor/liquid separator (27.13) will be required to conform with APCI process standards for both the design and maximum production modes. The sizing of this vessel will also be required to account for the variable process oil volume from cold start through various operating modes since it is impracticable to impose open-loop control over the process oil inventory. In this regard, APCI endorses recommendations made by EPRI in their review of the LPM pilot plant [Reference (1)].

5. Feed/Product Exchanger (21.10)

The E-101 and E-102 product gas coolers from the LPM pilot plant facility cannot handle the increased duty required for methanol production. For this reason reactor product gas will be precooled against incoming feed gas in a new feed/product exchanger (21.10).

6. Steam Generating Circulating Oil Cooler (E-103) Replacement With Oil Cooler (21.40)

Based on operating experience with the LPM pilot plant facility, it is known that the existing steam generating circulating oil cooler was not appropriately sized for this scale of operation. This unit acted as a large heat sink and eventually had to he by-passed during operation in Chicago. Due to the fact that the methanol synthesis reaction produces less of an exotherm than the methanation reaction, it can be expected that this situation would worsen in the LPMeOH PDU. Additionally, due to a lack of available boiler feed water at LaPorte, it is necessary to substitute a water cooled exchanger in this oil cooling service.

7. Unified Design Concept

Due to the effects of the changes noted above in items 4, 5 and 6 and additional considerations listed below, it is apparent that reutilization of the existing skidded equipment piping arrangement has, in large part, been negated. These additional considerations are as follows:

- The reactor and vapor/liquid separator are so tightly positioned within a structural steel frame that access to head flange areas for routine maintenance procedures is not possible.
- The various circulating oil pumps and filters are positioned such that inaccessability not only hinders routine maintenance, but presents a hazard. This was made manifest by a disabling fire at Chicago.
- Catalyst reduction flow is now preferred downflow rather than upflow as presently piped for the existing skidded equipment.

In light of this, and the desirable technical benefits of a full consideration of all modes of operation for such a development unit during its initial design, the "unified design" concept has been proposed. The unified design approach addresses all design modes of operation from the start of the design effort. The result is a single, unified PDU design for both liquid-fluidized and liquid-entrained operation. A basic premise of this concept is that a PDU designed for liquid-entrained operation can, through minor alteration of reactor internals, be operated in the liquid-fluidized mode. In contrast, substantial modifications are required on a unit designed specifically for fluidized operation, to permit later operation in the entrained mode. Presently, a significant modification is required to the existing LPM pilot plant to allow for any operation for methanol production. By considering both fluidized and entrained modes of operation at the start, this modification can be made in a most cost effective manner with respect to the overall program.

Presented below are the major points in this unified concept that differ from the original proposal to first rehabilitate the existing LPM unit specifically for liquid-fluidized operation and later design and install a skidded train of equipment that could theoretically be tied in alongside this first unit to allow liquid-entrained operation.

- Equipment, instrumentation, and valving specifications will include consideration of both modes of operation from day one. Experience with PDU design and operation shows that this design strategy will be a direct aid in avoiding extensive modifications later in the program that would not be identified otherwise.

- The use of slurry pumps is recommended in the initial installation since experience with the LPM pilot plant indicated that the major problem with liquid-fluidized operation is caused by catalyst attrition. Although the LaPorte LPMeOH PDU program will hopefully demonstrate some degree of resolution to this catalyst problem, it can be expected that operation will be extended into circumstances that will be damaging to the existing oil circulation pumps.

- The present plan considers the use of the existing oil circulation pumps in a closed loop oil circuit that will provide the necessary indirect heating and cooling of the slurry circulation stream during the liquid-entrained mode of operation. It is recommended that the required slurry heat exchanger for this service be installed initially along with the closed loop oil heating/cooling system.

8. Slurry Pumps (10.50 A&B)

Seal flush requirements for the slurry pumps are unresolved at this time, however, it is anticipated that the condensed oil flow (flowsheet streams #80, 82, and 83) will provide make-up to a seal flush circulation and injection system. Further development is required in this area of the flowsheet.

9. Pressure Leaf Filter (22.55)

In order to provide for safe handling of the potentially pyrophoric reduced catalyst particulates that will be suspended in the process oil during catalyst change-outs, a pressure leaf filter is recommended. Filtration requirements to one micron should be expected to permit recycling of process oil. A controlled oxidation of the catalyst could be performed on the filter before exposure to atmosphere. It is expected that an economic justification for this equipment will be made on the basis of waste oil disposal and clean oil make-up costs, or subcontracted reprocessing costs.

10. Side Stream Filter (22.50)

During operation in the liquid-fluidized mode, it is recognized that some on-line filtration will be required to control minor catalyst attrition. Since the pressure leaf filter cannot be expected to have a high on-line reliability, it is proposed to utilize the existing circulating oil filters to process a 10% to 20% sidestream bypassing the oil flow control valve.

11. Reactor Temperature Control

Evaluations have been performed of alternative temperature control schemes for the reactor. The clean oil "temperature control system" will be set up to achieve a constant clean oil temperature based on an operator selected set point. Control of the circulating oil/slurry temperature at the reactor inlet will be accomplished by effecting a bypass of the clean-oil around the 21.20 exchanger. The reactor inlet temperature set-point will be cascaded from a reactor outlet temperature controller.

12. Water Addition

Water addition through pumps 02.91 A&B is desirable as a provision for effecting the oil-methanol separation in the 3-phase separator.

13. 3-Phase Separator (22.10)

Control of condensed phase inventories will be accomplished by levels in the 3-phase separator.

14. Process Gas Vent and Recycle

Flow control has been considered for the process gas recycle by measuring the inlet and controlling the discharge on the recycle compressor.

15. Waste Oil Tank (28.40)

In order to provide for safe handling of oil wastes during maintenance or emergency operations a waste oil tank will be situated on the LaPorte LPMeOH PDU. A common header will run past various skid areas to permit temporary hose connections for transfer of oily wastes to this tank.

Accumulated oil containing reduced catalyst particulates can either be treated through the pressure leaf filter noted in item 9 above or disposed of by an approved waste disposal or reprocessing contractor.

16. Catalyst Reduction

The existing reduction gas exchanger 02.82 has been eliminated since the utility of this exchanger cannot be expected to justify the expense of installing it. Process calculations assure that reduction gas heater 02.83 will be sufficient for this service.

Catalyst reduction vessel 02.81 and slurry preparation tank 28.30 are illustrated in a more preliminary manner than the rest of the flowsheet.

Significant changes may be expected in the future as vendor scanning continues. This equipment comprises the only identifiable additions for liquid-entrained operation that will be specified, purchased, and installed on a later schedule than the rest of the system.

APCI's Process Engineering group received the basic process design information from CSI and developed this further. Based on the recommendations of CSI, an awareness of Fluor's conculsions on potential commercial operating conditions, and the practical constraints of the existing LPM pilot plant, a possible range of operating variables for the LaPorte LPMeOH PDU was defined. The range of operating variables is given in Exhibit 2.3.1-2.

The LaPorte LPMeOH PDU flowsheet was set up on APCI's process simulator program. This program is capable of calculating the detailed point·by point heat and mass balance for the flowsheet. A number of preliminary heat and mass balances were developed using the reactor mass balances provided by CSI. The possible operating ranges of the feed gas compressor, recycle compressor, and the slurry circulation pumps were reviewed and preliminary process specifications for these three long delivery equipment items were issued for estimating and vendor screening. Preliminary process specifications were also written for some 30 other equipment items in the LaPorte LPMeOH PDU in order that the Design Engineering group could develop a preliminary layout.

The final process design basis for the LaPorte LPMeOH PDU considers 8 design operating cases, 4 for the liquid-fluidized mode and 4 for the liquid-entrained mode. The operating conditions for these cases are given in Exhibits 2.3.1-3 and 2.3.1-4. A case numbering system has been adopted, using the key shown in Exhibit 2.3.1-5. As seen in Exhibits 2.3.1-3 and 2.3.1-4, the design methanol production rate ranges from 0.2 to 9.7 short tons per day. The final design heat and mass balances for these cases will be released in January 1982.

2.3.2 Equipment - The Machinery Applications group completed preparation of detailed mechanical specifications for the feed/recycle compressor. The Pressure Vessels group evaluated design documentation from the existing LPM pilot plant and determined that the reactor (R101) and the reactor separator (D103) could both be rerated from the current 890 psig to 1000 psig design pressure, leaving the design temperature at 750°F.

Investigation of the slurry pump in the LaPorte LPMeOH PDU lead to consultation with the International Coal Refining Co. on the type seal to be selected. APCI endorses ICRC's recommendations to use tandem welded metal bellows seals in slurry service at temperatures over 400°F. A report prepared by ICRC detailing the technical justification for this choice is attached as Exhibit 2.3.2-1.

2.3.3 Site/Structural - No Activity.

2.3.4 Piping - The Piping Design group prepared preliminary equipment arrangement plans for the LaPorte LPMeOH PDU. As noted in section 2.3.1, it was found that reutilization of the existing equipment arrangements in the LPM pilot plant was impossible given the flowsheet changes, new equipment, and the safety and maintenance requirements in the reactor, pump, and filter areas.

A preliminary site plan was drafted indicating approximate locations for equipment, buildings, storage, and roadways at the LaPorte LPMeOH PDU site. The preliminary equipment arrangement and site layout plans were transmitted to DOE in the Project Status Report for November (18 December 1981).

2.3.5 Electrical - No Activity.

2.3.6 Instrumentation - No Activity.

Task 3 - Equipment Procurement

No Activity.

Task 4 - LPM Pilot Plant Relocation

No Activity.

Task 5 - LaPorte LPMeOH PDU Renovation, Installation, and Shakedown

No Activity.

Task 6 - Liquid-Fluidized Operation

No Activity.

Task 7 - Laboratory Support Program

7.1 APCI R&D

7.1.3 Catalyst Screening and Testing -

Bench Scale Slurry Reactor Set-Up

The 1000 ml slurry reactor schematic is shown in Exhibit 7.1.3-1. The gas supply cylinders will be manifolded outside the laboratory auxiliary equipment will be located inside a ventilated reactor cell to prevent the build-up of toxic or flammable concentrations of H_2 and CO. The gas supplies to the reactor will be automatically shut-off in the event of a ventilation fan failure.

The reactor will be capable of 24 hour continuous operation. Critical operating parameters will be recorded on a multi-point chart recorder. Slurry level in the reactor will be monitored and automatically controlled.

All necessary safety and shut-down devices will be employed. The pressurized portion of the system will be protected with 1100 psig pressure relief valves, and the minimum pressure rating of components under their normal conditions of use will be 1500 psig. The reactor will be able to achieve 1400 psig operation with minor modification if this becomes desirable. All heaters will be protected with upper limit shut-off devices to prevent over-heating in the event of primary controller failure. Over-heating of the reactor contents will force an automatic gas shut-off, automatic heater disconnect, and maximum water flow through the reactor cooling coils. Pressure limit switches will also automatically shut off the inlet gas supply in the event of reactor over-pressurization or pressure loss caused by a significant leak.

1. Gas Manifolds and Inlet Lines

Existing gas manifolds and inlet lines for CO and H_2 supplies will be utilized. A small manifold and associated delivery line will be installed for supply of a CO_2/H_2 blended gas. The pressure of CO, H_2 and CO_2/H_2 feedstock supplies will normally be regulated to 1000 psig. The manifolds will be equipped with 1100 psig pressure relief valves. These relief valves can be reset for 1500 psig if increased pressure operation is necessary. Automatic magnetic excess flow shut-off valves will be used to guard against major leaks.

The gases will be piped into the reactor cell through 1/4" stainless steel tubing. After clean-up stages, the pressure will be further reduced using piston type regulators. The inlet flows of the component gases will be individually controlled by Brooks thermal mass flow controllers and will be recorded and time integrated. After carbonyl removal from the CO stream, the component gases will be mixed and input to the reactor via a preheater section.

With this system, feed gas blends with CO/H_2 ratios between 0.5 and 2.0 can be produced with an absolute accuracy of ±2.7% and maintained to within 0.5%.

2, Reactor

The reactor will be an Autoclave Engineers stainless steel, 1000 ml autoclave equipped with a Magnadrive II stirrer capable of delivering 16 in. lbs. of static torque and 0.63 hp at 2500 rpm. The reactor will contain a stainless steel cooling coil, thermocouple well and baffle assembly, and will be fitted with a rupture disc. The gas inlet point will be beneath the 6-flat-bedded turbine impeller, which, because it is a hollow shaft Dispersimax type, will induce gas backmixing from the gas space above the slurry.

Exit gas will pass through an air cooled partial condenser with a controlled temperature of 200°C to reflux the slurry oil back to the reactor. The exit gas is then directed through a heated line to a heated dome loaded back pressure regulator to be let down to near atmospheric pressure.

3. Reactor Temperature Control

The reactor will be heated under thermostatic proportional control to the typical operating temperature of between 200 and 270°C. The heat produced by the exothermic methanol synthesis reaction will be removed by evaporation of cooling water in the internal cooling coil. Loss of inlet water pressure will cause a shut-off of the inlet gas supply to prevent overheating of the reactor. The flow of water to the cooling coil will be controlled by a solenoid and manual regulating valve in series, the solenoid valve activated by the reactor thermocouple alarm system. A high temperature alarm will be installed which will control a solenoid valve installed to by-pass the manual regulating valve and allow full water flow to the cooling coils. As already described, this high temperature alarm will also interrupt the reactor heaters and gas supply.

4. Slurry Level Control

A high temperature, differential pressure (dp) transducer, connected between the gas inlet and outlet lines, will be used to monitor the slurry level within the reactor. A decrease in slurry inventory will be compensated for by pumping fresh oil or slurry into the reactor, utilizing a Bran and Lubbe N-P31 piston metering pump constructed of 316 stainless steel and tungsten carbide. A rise in differential pressure above a set point, caused by an increased slurry inventory, will open an electrically operated ball valve and allow slurry to transfer back to the slurry reservoir via throttling valves. A high/low alarm on the dp transducer will shut off the slurry pump if either a malfunction of the transducer or an abnormal slurry level should occur.

The glass reservoir will be maintained under an inert atmosphere and will be protected by a 3/4" in-line relief valve, which is sized to allow a maximum pressure of 10 psig in the event of a malfunction of the slurry level system.

5. Exit Gas Analysis

After pressure let-down, the exit gas will be directed via heated lines to heated solenoid valves which select flow to either a heated, oil filled wet test meter for flow rate measurement, or to the analytical system for component analysis. The analytical system consists of a Carle process gas chromatograph (GC), which analyzes for CO, CO_2, H_2, H_2O, methanol, ethanol, methane, ethane, propane, and dimethyl ether. The GC is controlled by a Perkin-Elmer Sigma 15 computing integrator, which allows complete analysis automation and data reduction. The analysis system will operate unattended and store both raw and reduced data on a micro floppy diskette system. This analytical system will also be utilized for the gas phase testing reactors. An automated sample stream selection valve(s) will be used to switch between the reactors.

7.1.4 Catalyst Preparation - A total of 22 to 25 catalyst formulations were established in the following three categories: unsupported mixed oxides, supported mixed oxides and Raney alloys. Mixed heterogeneous-homogeneous dual systems were excluded from the original plan. So also were catalysts because of the safety concern with their radioactive nature. There is perhaps some technical potential for thorium-based catalysts but the risk/benefit ratio of large-scale applications of $ThCu_6$ catalyst needs to be evaluated prior to its preparation.

A request for contract change was submitted on 10 November 1981 proposing that 3 commercial catalysts be used for baseline data in the screening subtask 7.1.3. To hold the number of potential catalyst test candidates to 25, it was proposed to synthesize 22 rather than 25 new catalysts. It is believed that 22 new catalysts are sufficient to cover the basic compositions of interest; it is judged that the screening effort will benefit from the establishment of strong baseline data.

7.2 CSI R&D

7.2.1 Literature Review - A review of published information in three-phase reaction systems for the LPMeOH liquid-entrained mode of operation was to have been initiated during the reporting period. This work was deferred, however, until adequate funding is committed.

7.2.2 Autoclave Tests - An autoclave program with powdered methanol catalysts suspended in an inert hydrocarbon liquid was initiated. This work represents a supplement to a program begun under prior funding. The equipment to be utilized in this study has been described in Reference (2).

The first phase of the autoclave program is an optimization of the in-situ catalyst reduction procedure. The productivity of catalysts reduced while suspended in the hydrocarbon liquid will be compared to that of a base case vapor-phase reduced catalyst at various conditions.

The initial range of variables to be examined are as follows:

- Maximum Reduction Temperature - 180 to 275°C
- Reduction Pressure - 110 to 7,000 kPa
- Reduction Gas Composition - 2 to 98% hydrogen by volume
- Gas Flow - 250 to 1,000 l/hr-kg cat.

The catalyst loading will be maintained at 0.2 kg of unreduced powder per liter of oil. Agitator speed will be 1,200 rpm and the duration of reduction will be 24 hours or until gas uptake is completed.

The first five experiments involve temperature and pressure variations at a hydrogen concentration of 98 percent and a gas flow of 1,000 l/hr - kg cat. All of the reduced catalysts will be evaluated at four standard conditions using a Lurgi-type feed gas having a composition of 50 percent hydrogen, 25 percent carbon monoxide, 10 percent carbon dioxide and 15 percent argon. The four productivity-evaluation conditions are within the following range of variables:

- Temperature - 250° to 275°C
- Pressure - 3,600 to 7,100 kPa
- Flow - 4,000 to 8,000 l/hr - kg cat.

These experiments have begun and will be reported in the upcoming Project Status Reports and in the next Technical Progress Report.

7.2.5 Cold-Flow Model

This subtask involves the construction and operation of a cold-flow unit which is to be sized comparable to the LaPorte LPMeOH PDU reactor. The objective of this work is to determine the hydrodynamics of the reactor in both liquid-fluidized and liquid-entrained modes of operation. The results would be used for reactor modeling purposes and for determining the necessity and type of reactor internals required for efficient LPMeOH PDU operation.

Initial work in this area involved determining the equipment specifications and costs as well as planning the experimental program as required in the Task 1 effort. Further work on the cold-flow model has been deferred until adequate funding is committed. APCI is also reviewing the applicability of separate but related cold flow modeling being performed for liquid phase Fischer-Tropsch development [Reference (3)].

7.2.6 CSI Lab PDU Modifications - This subtask involves the construction of a liquid-entrained reactor system which is to be added to the existing CSI lab PDU containing a liquid-fluidized reactor system. A description of the liquid-entrained lab PDU has been presented in Reference (2). Exhibit 7.2.6.-1 is a simplified process flow diagram for the CSI liquid-entrained lab PDU.

Work in this area has concentrated on the preparation of specifications and evaluation of bids for the slurry pumps and the vessel skid. The evaluation of slurry pump alternatives is nearly completed. Requests for quotations on the vessel skid have been sent to several fabricators.

7.2.7 CSI Lab PDU Experimental Program - No Activity. This subtask will not be initiated until the CSI lab PDU modifications have been completed.

7.2.8 Support for LaPorte LPMeOH PDU - This subtask involves support for the LaPorte LPMeOH PDU program, primarily testing catalysts to be used in the larger-scale equipment. The initial effort in this regard will be to test liquid-fluidized catalyst candidates in the CSI lab PDU. The first of these runs will be conducted during the next quarter. In the interim, the CSI liquid-fluidized lab PDU has been reconditioned and is now ready for use.

Task 8 - LaPorte LPMeOH PDU Modifications for Liquid-Entrained Mode

No Activity.

Task 9 - Shakedown for Liquid-Entrained Operation

No Activity.

Task 10 - Liquid-Entrained Operation

No Activity.

Task 11 - Project Evaluation

11.1 APCI Management Activities

11.1.1 Project Management - Project Management coordinated the preparation of reports.

11.1.2 Economic Evaluation - See section 2.1.2.

11.1.3 Travel and Living - CSI and APCI met on several occasions in Allentown, and once in Fairfield. APCI's Operations Manager from LaPorte attended meetings in Allentown and visited the LPM pilot plant in Chicago.

11.2 CSI Activities

11.2.1 Data Evaluation - No Activity.

11.2.2 Design and Economics - No Activity.

11.2.3 Process Scaleup - No Activity.

11.2.4 Reporting - A Topical Report covering the liquid-entrained LPMeOH laboratory development work accomplished under advance funding was issued (2). The monthly Project Status Reports for October, November, and December 1981 were prepared and issued.

11.3 APCI Design

11.3.1 Integration - Machinery Applications provided review and comment to CSI on CSI's slurry pump bids for the CSI lab PDU.

11.4 APCI R&D

11.4.1 Corporate Development Department - Review and comment were provided to CSI on their Topical Report. Input was provided to the Monthly Status Reports.

11.4.2 Process Systems Group R&D - Review and comment were provided to CSI on their Topical Report. Input was provided to the Monthly Status Reports.

REFERENCES

(1) Hoffert, F., "Report on the Inspection of the LPM Unit at IGT," prepared for EPRI (30 November 1978).

(2) Topical Report, "Liquid-Entrained LPMeOH, Advance Funded Work, Technical Data," prepared by Chem Systems Inc. for the U.S. DOE under Contract No. DE-AC22-81PC30019 (December 1981).

(3) "Catalyst and Reactor Development for a Liquid Phase Fischer-Tropsch Process," being performed by APCI for the U.S. DOE under Contract No. DE-AC22-80PC30021 (October 1980 - October 1983).

ACKNOWLEDGEMENTS

The Chem Systems work is under the direction of the CSI Program Manager, R.L. Mednick. M.I. Greene is performing as the CSI Research Manager. At Air Products the responsible Research Managers are S.A. Butter and M.S. Chen. Lead engineering performers at Air Products are D.J. Silkworth - Project Engineering, T.R. Tsao - Process Engineering, and J.L. Henderson - LaPorte Operations. The assistance of R. J. Burna of ICRC with slurry pump seal information is gratefully acknowledged.

ATTACHMENTS

Exhibit 2.2.1-1
2.2.1-2
2.2.1-3
2.2.2-1
2.3.1-1
2.3.1-2
2.3.1-3
2.3.1-4
2.3.1-5
7.1.3-1
7.2.6-1

E. P. Holley
Project Manager

J. Klosek

J. Klosek
Program Manager

p

ATTACHMENTS

EXHIBIT 2.2.1-1

LaPorte LPMeOH PDU Fresh Feed Gas Compositions

	Lurgi-type Feed	KT-type Feed	Balanced-type Feed
H_2	50.0 Mol %	38.0 Mol %	65.8 Mol %
CO	25.0	60.0	32.9
CO_2	10.0	1.0	0.6
CH_4,C_2H_6	13.0	-	0.3
N_2	1.8	0.8	0.2
Ar	0.2	0.2	0.2
	100.0	100.0	100.0

EXHIBIT 2.2.1-2

CSI Process Design Basis for the LaPorte LPMeOH PDU Liquid-Entrained Reactor at High Space Velocity and Moderate Catalyst Loading

- 7,000 kPa, 250°C
- S_V: 10,000 liters/hr - kg. cat.
- Reactor: 22.5" ID X 15' T-T
- CO conversion: 34%

  CO_2 conversion: 5%
- Catalyst: "A" powder
- Loading: 0.2 kg oxide/liter cold oil
- Reactor volume: 41.4 FT^3 = 1172.5 liters
- Selectivity: to CH_3OH, 98%

  to C_2H_5OH, 2%
- Lurgi-type feed gas

EXHIBIT 2.2.1-3

CSI Gas-Phase Material Balance for the LaPorte LPMeOH PDU Liquid-Entrained Reactor At High Space Velocity and Moderate Catalyst Loading

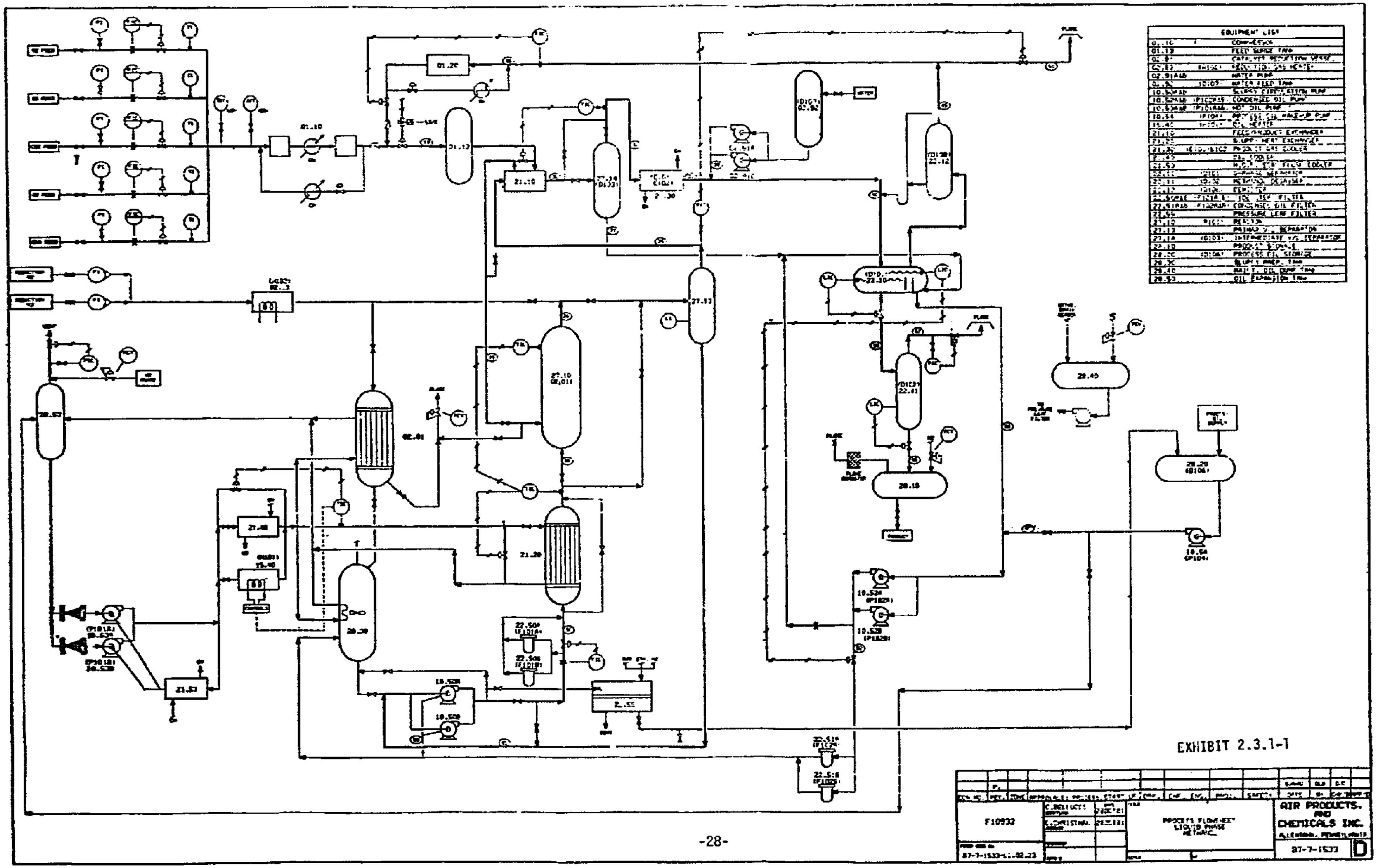
	REACTOR FEED		REACTOR PRODUCT		LIQUID PRODUCT		UNCONDENSED GAS	
	lb M/hr	lbs/hr	lb M/hr	lbs/hr	lb M/hr	lbs/hr	lb M/hr	lbs/hr
H_2	89.65	179.3	56.48	113.0	-	-	56.48	113.0
CO	44.83	1,255.2	29.59	828.5	-	-	29.59	828.5
CO_2	17.93	788.9	17.03	749.3	-	-	17.03	749.3
CH_4+N_2	26.90	430.4	26.90	430.4	-	-	26.90	430.4
H_2O	-	-	1.06	19.1	0.92	16.6	0.14	2.5
CH_3OH	-	-	15.84	506.9	14.96	478.7	0.86	28.2
C_2H_5OH*	-	-	0.16	7.4	0.16	7.4	-	-
Total	179.30	2,654.6	147.04	2,654.6	16.04	502.7	131.00	2,151.9
SCFH	67,955		55,728				49,648	

*Includes higher alcohols

EXHIBIT 2.2.2-1

LaPorte LPMeOH PDU Data Logger
Minimum Information

Data Point Description	Temp.	Press.	Flow
1. Feed Carbon Monoxide	1	1	1
2. Feed Hydrogen		1	1
3. Feed Carbon Dioxide		1	1
4. Feed Nitrogen		1	1
5. Feed Methane		1	1
6. Feed Argon		1	1
7. Recycle Gas	1	1	1
8. Combined Reactor Feed Gas	1	1	1
9. Reactor	5		
10. Circulating Oil/Slurry	1		1
11. V/L Separator		1	
12. Hot Reactor Effluent Gas	1	1	1
13. Purge Gas			1
14. Water Addition			1
15. Methanol Product			1
16. Methanol Degasser Vapor			1
17. Condensed Oil Return			1
Totals	10	10	15



EQUIPMENT LIST		
01.10		COMPRESSOR
01.13		FEED SURGE TANK
[illegible]		CATALYST REDUCTION VESSEL
02.82	(H102)	[illegible] GAS HEATER
02.91A&B		WATER PUMP
02.92	(D107)	WATER FEED TANK
10.50A&B		SLURRY CIRCULATION PUMP
10.52A&B	(P102A&B)	CONDENSED OIL PUMP
10.53A&B	(P101A&B)	HOT OIL PUMP
10.54	(P104)	PROCESS OIL MAKE-UP PUMP
15.40	(H105)	OIL HEATER
21.10		FEED/PRODUCT EXCHANGER
21.20		SLURRY HEAT EXCHANGER
21.30	(E101/E102)	PRODUCT GAS COOLER
21.40		OIL COOLER
21.53		[illegible] COOLER
[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	METHANOL DEGASSER
[illegible]	[illegible]	[illegible]
22.50A&B	(F101A&B)	[illegible] FILTER
22.51A&B	(F102A&B)	CONDENSED OIL FILTER
22.55		PRESSURE LEAF FILTER
27.10	(R101)	REACTOR
27.13		PRIMARY V/L SEPARATOR
27.14	(D103)	INTERMEDIATE V/L SEPARATOR
28.10		PRODUCT STORAGE
28.20	(D106)	PROCESS OIL STORAGE
28.30		SLURRY PREP. TANK
28.40		[illegible] OIL DUMP TANK
28.53		OIL EXPANSION TANK

EXHIBIT 2.3.1-1

EXHIBIT 2.3.1-2

Range of Operating Variables for LaPorte LPMeOH PDU

	Minimum	"Normal"	Maximum
Reactor Pressure, psig	500	700	900
(kPag)	(3,443)	(4,827)	(6,206)
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
(m)	(1.5)	(2.1)	(2.1)
Liquid-Entrained Catalyst Loading, kg. cat/liter cold oil	0.1	0.2	0.4
(kg. cat/kg cat-oil slurry)	(0.11)	(0.20)	(0.33)

NOTES:

1. LaPorte LPMeOH PDU design pressure 1000 psig (6,896 kPag).
2. Liter at standard conditions 20°C (68°F), 14.696 psia (101 kPa).
3. Catalyst kg based on catalyst in oxide state, prior to reduction.

EXHIBIT 2.3.1-3

Design Cases for Liquid-Fluidized Mode:
Operating Conditions for LaPorte LPMeOH PDU

Case No.	Space Velocity 1/hr kg	Press, psig	Temp. °C	Catalyst Bed. Ht. ft	Catalyst Loading kg/l	%CO Conv. (Single pass)	%CO_2 Conv. (Single pass)	% CH_3OH Select.	% C_2H_5OH Select.	% CO_2 Select.	Reactor Feed lb-Mol/hr	Fresh Feed lb-Mol/hr	Recycle Flow lb-Mol/hr	Purge Flow lb-Mol/hr	MeOH Product Rate ST/D
FB-4930.8	4,000	900	230	7	-	25	3	98	2	0	247	53	194	1	6.5
FL-3750.8	2,500	700	250	7	-	20	2	98	2	0	155	30	125	10	2.6
FK-2750.6	2,000	700	250	7	-	10	0	83	12	5	124	46	78	20	3.3
FK-1570.3	1,000	500	270	7	-	7	0	83	12	5	61	41	20	36	0.5

EXHIBIT 2.3.1-4

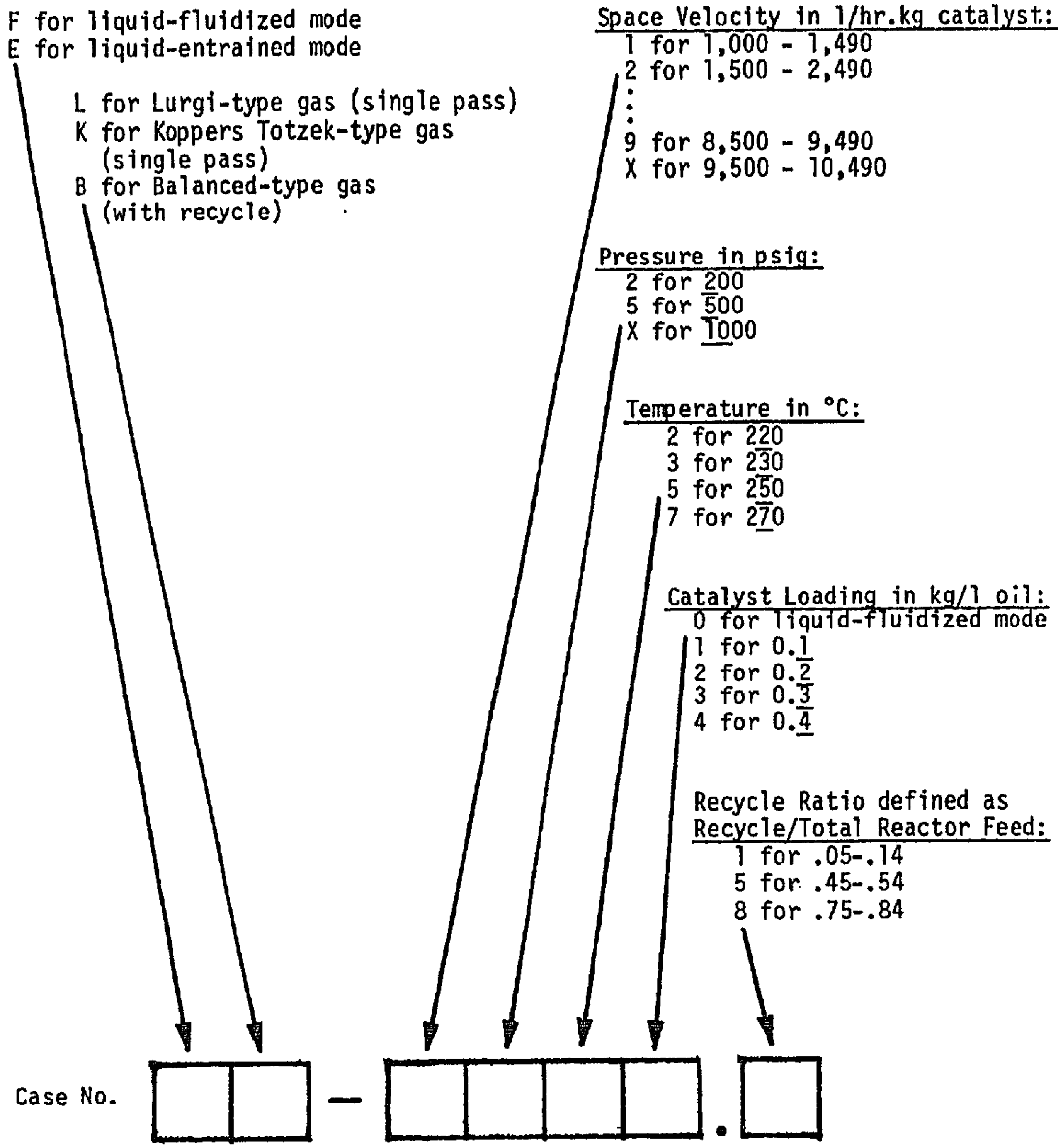
Design Cases for Liquid-Entrained Mode:

Operating Conditions for LaPorte LPMeOH PDU

Case No.	Space Velocity 1/hr kg	Press, psig	Temp. °C	Catalyst Bed. Ht. ft	Catalyst Loading kg/l	%CO Conv. (Single pass)	$\%CO_2$ Conv. (Single pass)	% CH_3OH Select.	% C_2H_5OH Select.	% CO_2 Select.	Reactor Feed lb-Mol/hr	Fresh Feed lb-Mol/hr	Recycle Flow lb-Mol/hr	Purge Flow lb-Mol/hr	MeOH Product Rate ST/D
EB-X954.8	10,000	900	250	-	0.4	30	3	98	2	0	350	77	273	0.5	9.7
EL-6752.7	6,000	700	250	-	0.2	24	2	98	2	0	107	27	80	10	2.3
EK-4752.7	4,000	700	250	-	0.2	12	0	83	12	5	72	22	50	10	1.6
EK-2571.0	2,000	500	270	-	0.1	7	0	83	12	5	18	18	0	17	0.2

EXHIBIT 2.3.1-5

Case Numbering System



F for liquid-fluidized mode
E for liquid-entrained mode

L for Lurgi-type gas (single pass)
K for Koppers Totzek-type gas (single pass)
B for Balanced-type gas (with recycle)

Space Velocity in 1/hr.kg catalyst:
1 for 1,000 - 1,490
2 for 1,500 - 2,490
⋮
9 for 8,500 - 9,490
X for 9,500 - 10,490

Pressure in psig:
2 for 200
5 for 500
X for 1000

Temperature in °C:
2 for 220
3 for 230
5 for 250
7 for 270

Catalyst Loading in kg/l oil:
0 for liquid-fluidized mode
1 for 0.1
2 for 0.2
3 for 0.3
4 for 0.4

Recycle Ratio defined as Recycle/Total Reactor Feed:
1 for .05-.14
5 for .45-.54
8 for .75-.84

EXHIBIT 2.3.2-1
SLURRY PUMP SEAL SELECTION

Service: High Temperature Fluids (Greater Than $400^{\circ}F$, Solids)

Solids concentration greater than 0% under design conditions. The lined slurry pump applications are included in this group.

SRC Technology recommends the use of tandem welded metal bellows seals in high temperature fluids greater than $400^{\circ}F$ with solids. The tandem metal bellows seals (Fig. 1) have the following advantages over other proposed seal arrangements (Fig. 2, 3, & 4):

TANDEM WELDED METAL BELLOWS SEAL

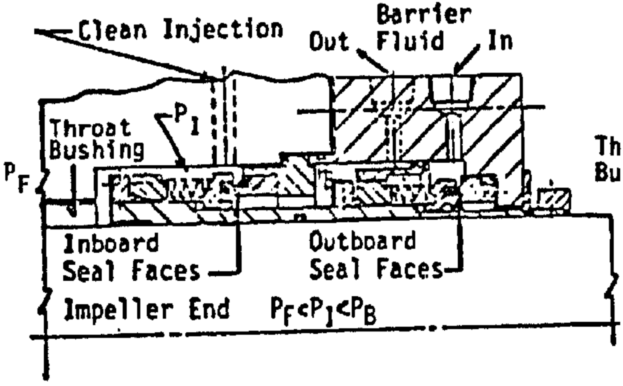


Figure 1

TANDEM PUSHER TYPE SEAL

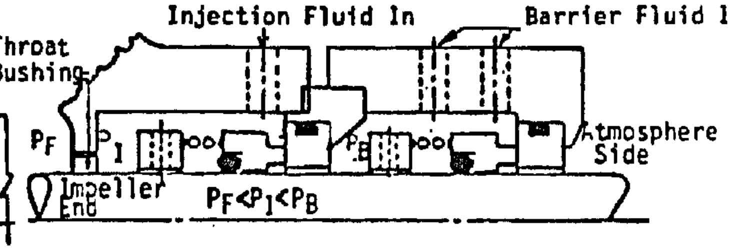


Figure 2

DOUBLE, BACK-TO-BACK WELDED METAL BELLOWS

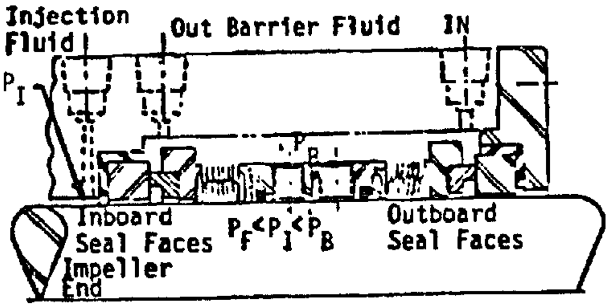


Figure 3

DOUBLE, BACK-TO-BACK PUSHER TYPE SEAL

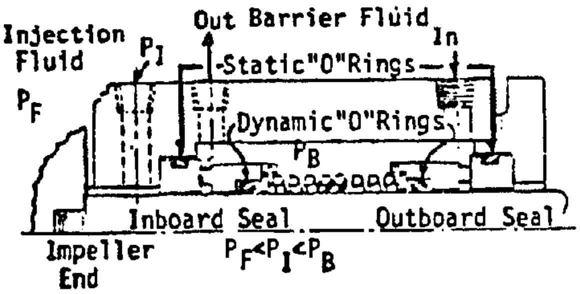


Figure 4

P_F = Abrasive, Hazardous Fluid Or Slurry Pressure

P_I = Clean, But Hazardous, Injection Fluid Pressure

P_B = Clean, Non-Hazardous Circulating Barrier Fluid Pressure

In a tandem seal (Fig. 1 & 2), abrasive fluid is kept out of the stuffing box because clean injection pressure, P_I, is normally greater than abrasive fluid pressure, P_F. Barrier fluid pressure, P_B, is normally kept slightly higher than injection pressure to enable the barrier fluid to act as an inboard seal face lubricant and to decrease the chance of abrasive or dirt intrusion between the inboard seal faces. Under normal operating conditions, small traces of barrier fluid leak into the injection fluid then into the abrasive fluid stream; however, no harm occurs since all three fluids are compatible. When injection pressure is lost, abrasive fluid pressure exceeds injection pressure and the abrasive fluid begins to enter the stuffing box. Since the abrasive fluid pressure is not as high as the design injection pressure and since barrier fluid pressure remains constant, the pressure differential across the inboard seal faces increases causing more barrier fluid to leak into the area between the inboard seal faces and the pump throat bushing. Increased barrier fluid inleakage is beneficial because it contains the abrasive, hazardous fluid and hazardous injection fluid in the stuffing box between the inboard seal faces and the throat bushing and gives the operators time to fix the injection fluid pressure system. Furthermore, contamination of the barrier fluid circulation system and leakage of hazardous injection fluid and abrasive, hazardous fluid to the atmosphere does not occur.

If injection pressure is lost in a tandem welded metal bellows seal, the abrasive particles that do enter the stuffing box remain at the outer diameter of the bellows of the inboard seal. Since centrifugal force tends to throw these particles out towards the wall of the stuffing box, the chance of particles packing the area between the convolutions of the bellows is decreased and bellows flexibility is maintained. A flexible bellows is necessary to help compensate for seal face wear and axial shaft movement. In a tandem pusher type seal, centrifugal force also helps prevent abrasive particles from packing the area around and below the springs, thus spring flexibility is maintained. In both the bellows and pusher type tandem seals, centrifugal force also tends to hurl abrasive particles away from the inboard seal faces thus decreasing the chance of abrasive intrusion between the faces.

In a double, back-to-back seal (Fig. 3 & 4), abrasive fluid is also kept out of the stuffing box because barrier fluid pressure, P_B, is greater than injection pressure, P_I, and injection pressure is greater than abrasive fluid pressure, P_F. If injection pressure is lost, abrasive fluid comes in intimate contact with the inside diameters of the inboard seal. Even though barrier fluid pressure remains higher than abrasive fluid pressure, centrifugal force and capillary action may urge abrasive particles between the rotating and stationary faces of the inboard seal causing possible seal face wear. Furthermore, when injection pressure is lost in a double, back-to back pusher type seal, abrasive particles tend to lodge under the rotating head and dynamic secondary seal (shown in Fig. 4 as a dynamic elastomeric O-ring located between the rotating head and shaft). To compensate for inboard seal face wear or any axial shaft movement, the rotating head of the inboard seal tries to move axially to the left to keep the faces closed. However, the abrasive particles lodged under the rotating head and dynamic secondary seal prevent this axial movement. Consequently, this rotating head and secondary seal "hang up" causes the inboard seal to leak. Upon loss of injection pressure in a double, back-to-back metal bellows seal (see Fig. 3), abrasive particles accumulate in the inside diameter of the bellows. Centrifugal force tends to pack the convolutions of the bellows with abrasive particles causing bellows inflexibility. When bellows become inflexible, they cannot compensate for seal face wear or axial shaft movement. Hence seal leakage results.

If barrier fluid pressure is lost in a tandem seal, barrier fluid no longer acts as an inboard seal face lubricant and traces of barrier fluid no longer leak into the injection fluid. In fact, injection fluid becomes the inboard seal face lubricant. Negligible traces of hazardous injection fluid may leak into the barrier fluid, but no abrasive, hazardous fluid should enter the barrier fluid, or even the stuffing box, since injection pressure is still higher than abrasive fluid pressure. Consequently, tandem seals tend to increase plant reliability since only a concurrent loss of injection pressure and barrier fluid pressure can cause seal failure.

If barrier fluid pressure falls below injection pressure and abrasive, hazardous fluid pressure in a double, back-to-back seal, seal failure will occur. If there is a loss of barrier fluid pressure or if there is a system upset causing abrasive fluid pressure to rise, barrier fluid pressure may fall below injection pressure and abrasive fluid pressure ($P_B<P_F<P_I$). If this happens, the rotating head of the inboard seal is forced axially to the right since injection pressure is higher than barrier fluid pressure and the inboard seal faces open.

Abrasive, hazardous fluid and hazardous injection fluid enter the stuffing box and contaminate the barrier fluid circulation system. Furthermore, abrasive fluid becomes the outboard seal face lubricant. These faces soon wear and hazardous fluid is leaked to the atmosphere. Both double, back-to back bellows and pusher type seals are susceptible to this type of seal failure.

In fact, several pilot plants have already reported double, back-to-back pusher type seal failures similar to those described above. Wilsonville reported that they had trouble maintaining barrier fluid pressure which caused the inboard seal to open. Exxon Coal Liquefaction Plant reported a double, back-to-back seal failure when a process upset raised slurry pressure above barrier fluid pressure causing contamination of the barrier fluid system when the inboard seal faces were forced open.

Tandem pusher type and double, back-to-back pusher type seals typically employ dynamic elastomeric secondary seals (see Fig. 2 & 4). Elastomeric secondary seal materials have a temperature limitation of 500-550°F (for Kalrez). However, some hazardous, abrasive fluid streams in the Demonstration Plant have a design operating temperature exceeding 700°F, far above the allowable limit for elastomers. Dynamic secondary seals made of graphite have been tried in double, back-to-back pusher type seals at high process solvent temperatures at Wilsonville, but Wilsonville reported that the dynamic graphite secondary seals failed in the presence of this hot process solvent. Wilsonville also reported that static graphite secondary seals did not fail in the presence of hot process solvent. There is a strong chance that dynamic elastomeric secondary seals in tandem pusher type seals would also fail when subjected to hot process solvent at high temperature (greater than 550°F).

Tandem or double, back-to-back bellows seals are commonly available with temperature limitations up to 800°F. Secondary sealing is accomplished by statis graphite gaskets or metal wedges which are not susceptible to breakdowns in the presence of hot process solvent.

Since tandem seals appear to offer greater operating reliability than double, back-to-back seals and since welded metal bellows can operate at higher temperature than pusher type seals, SRC Technology recommends the use of tandem welded metal bellows seals in high temperature(greater than 400°F) hazardous, abrasive fluid centrifugal pump applications. Double, back-to-back seals are only recommended where tandem seals cannot be made to fit in the stuffing box of a pump requiring a double seal.

If solids are definitely present in an environmentally hazardous fluid but exist in very low concentrations, a tandem metal bellows seal should again be employed, and the injection fluid and barrier fluid systems connected and operated. However, if operating experience proves the injection fluid system to be superfluous, then it may be shut off. If only low concentrations of solids are present in a non-hazardous fluid, then operating experience may prove that the injection fluid system and barrier fluid system may be shut off.

Environmental Considerations

In addition to their ability to operate effectively in fluids containing solids, tandem seal arrangements may be required for environmental reasons. Environmental standards appearing in the Federal Register/Vol. 46 No. 2/ Monday, January 5, 1981/Proposed Rules state in part the following:

> The proposed standards would require pumps in light liquid service to be equipped with dual mechanical seal systems that include a barrier fluid system. The barrier fluid would be required to be something other than a light liquid or gaseous VOC[1]. Light liquids are defined as VOC liquids with vapor pressures greater than 0.3 kPa at 20°C. Each barrier fluid system would be equipped with a sensor so that failure of the inner and outer seals could be detected. In addition, each barrier fluid system would be operated at a pressure greater than the seal area pressure or would be equipped with barrier fluid degassing reservoir. The degassing reservoir would be connected by a closed vent system to a control device having a VOC control efficiency of at least 95 percent. The proposed standards would also require weekly visual inspections of the seals on light liquid pumps in order to identify failure of the outer seal. Repair of the pump would be required within 15 days after a seal failure or leak was detected unless repair would require a process unit shutdown. The first attempt at repairing the pump would be required within 5 days after detection of the leak. If a pump could not be equipped with dual mechanical seals and a barrier fluid system, a closed vent system would be required to transport leakage to a control device having a VOC control efficiency of at least 95 percent.

[1] Volatile Organic Compound (VOC)" means any organic compound which participates in atmospheric photochemical reactions. "In VOC Service" means that a fugitive emission source contains or contacts a process fluid that is at least 10% VOC

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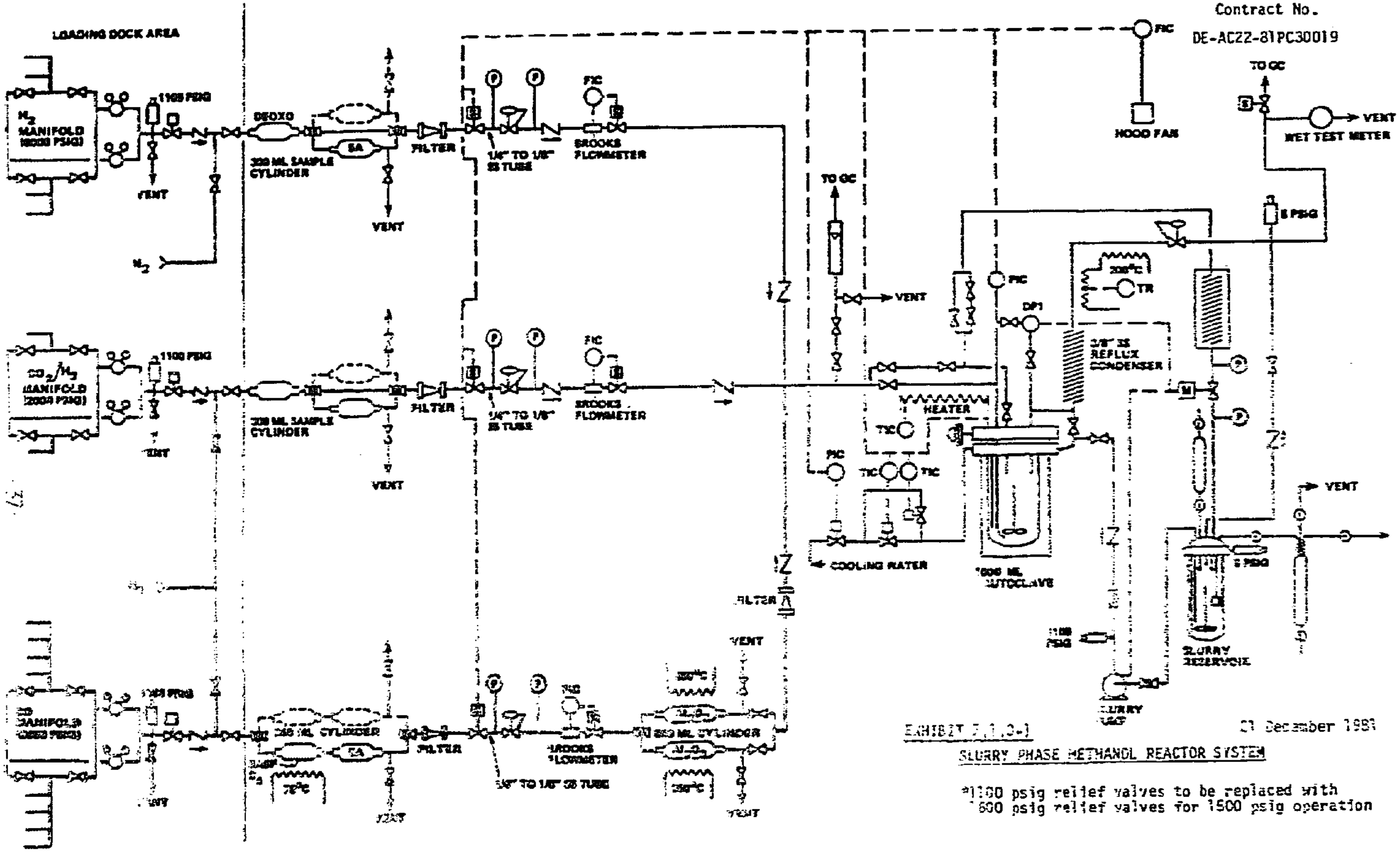

EXHIBIT 7.1.3-1

31 December 1981

SLURRY PHASE METHANOL REACTOR SYSTEM

*1100 psig relief valves to be replaced with 1600 psig relief valves for 1500 psig operation

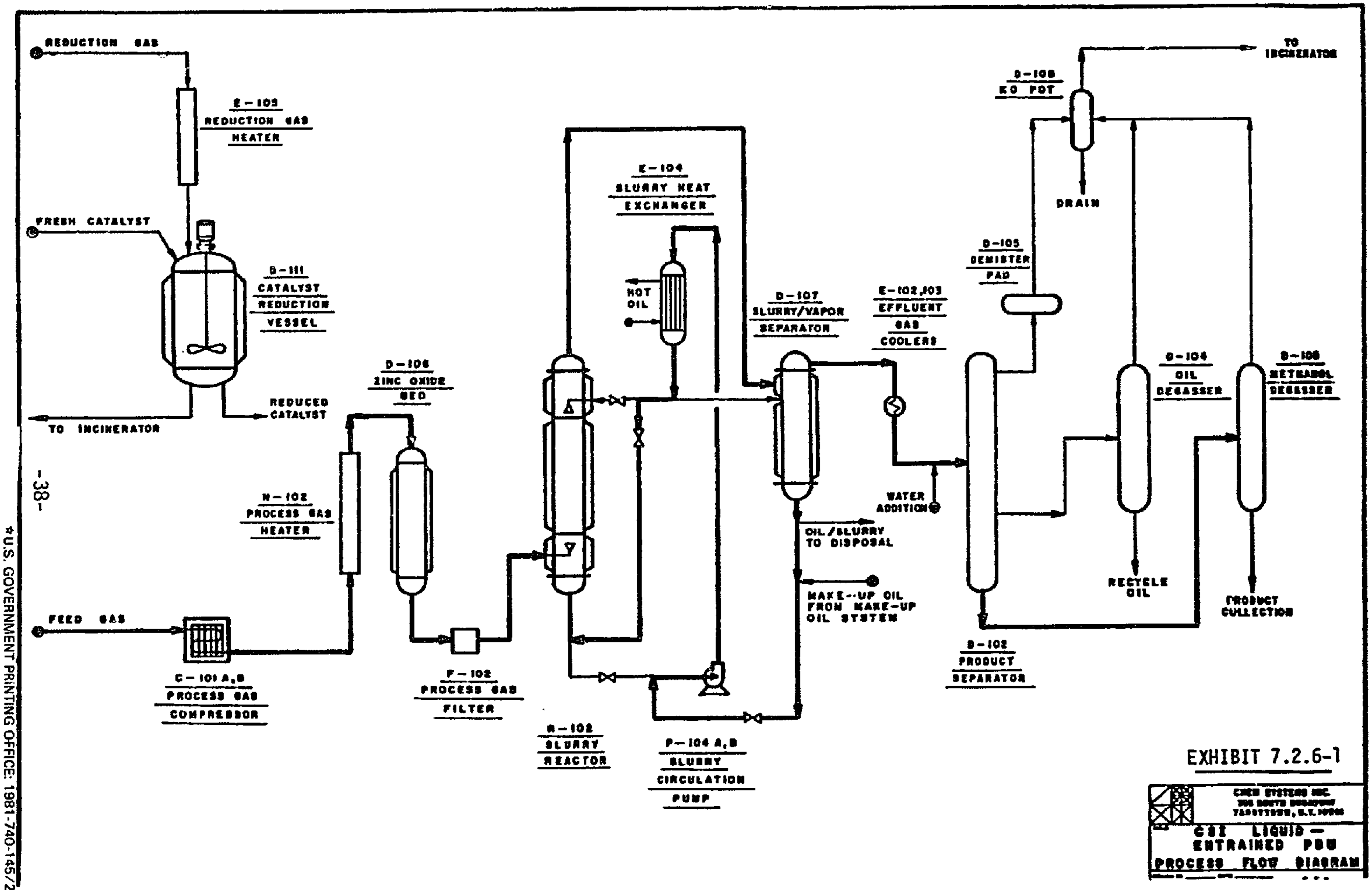

EXHIBIT 7.2.6-1

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