DRAFT

LIQUEFACTION OF COALS USING ULTRA-FINE PARTICLE,
UNSUPPORTED CATALYSTS: IN SITU GENERATION BY
RAPID EXPANSION OF SUPERCRITICAL FLUID SOLUTIONS

QUARTERLY TECHNICAL PROGRESS REPORT

for the period

October 1, 1990 - December 31, 1990

Date Prepared

August 1991

prepared for

U.S. DEPARTMENT OF ENERGY
Pittsburgh Energy Technology Center
Pittsburgh, Pennsylvania 15236-0940

prepared by

TEXTRON DEFENSE SYSTEMS
a subsidiary of Textron Inc.
2385 Revere Beach Parkway
Everett, Massachusetts 02149

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Under Contract No.: DE-AC22-90PC90053

DRAFT

Q4Q90

TABLE OF CONTENTS

<u>Section</u>		<u>Page</u>
1.0	SUMMARY	1
2.0	TECHNICAL APPROACH	5
	2.1 Program Objectives	5
	2.2 Concept Description	6
	2.3 Program Approach	8
3.0	TECHNICAL PROGRESS	14
	3.1 Task 1 - Project Work Plan	14
	3.2 Task 2 - Experiment Design and Fabrication	14
	3.3 Task 3 - Solute/Solvent Optimization	17
	3.4 Task 4 - Catalyst Preparation and Evaluation	18
	3.5 Task 5 - Reporting and Documentation	18
4.0	CONCLUSIONS AND FUTURE WORK	19
	4.1 Conclusions	19
	4.2 Future Work	20
5.0	REFERENCES	21

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

1.0 SUMMARY

The program objective is to generate ultra-fine catalyst particles (20 to 400 Å in size) and quantify their potential for improving coal dissolution in the solubilization stage of two-stage catalytic-catalytic liquefaction systems. It has been shown that catalyst activity increases significantly with decreasing particle size for particle sizes in the submicron range.

Ultra-fine catalyst particle generation will be accomplished using a novel two-step process. First, the severe conditions produced by a supercritical fluid (e.g., supercritical H_2O or CO_2) will be used to dissolve suitable catalyst compounds (e.g., Fe_2O_3 , FeS_2 , and/or $\text{Fe}(\text{CO})_5$). Sulfur containing compounds may be added to the supercritical solvent during catalyst dissolution to enhance the catalytic activity of the resulting ultra-fine, iron based, catalyst particles.

Downstream of dissolution stage of the ultra-fine catalyst particle generator, the supercritical solution will be allowed to expand rapidly from the severe thermodynamic conditions of the supercritical state to ambient pressure. The extremely rapid expansion process results in an extreme density decrease producing a highly super-saturated, highly nonequilibrium condition. Under these conditions, nucleation and particle growth take place. These processes continue for a very brief time (of order 1 μs) because strong mixing processes leading to rapid dilution of the expanding solution occur very close to the nozzle discharging the supercritical solution. Since nucleation and particle growth are allowed to take place for only a very brief time, the expanded supercritical solution will contain extremely small catalyst particles (20-400 Å in size). In situ impregnation of coal materials (e.g., coal

particles or coal derived liquids) will be accomplished by allowing the post-expansion, ultra-fine catalyst particle laden gas stream to come into intimate contact with pulverized coal or coal-liquid droplets. Liquefaction activity testing using the catalyst impregnated coal materials will be performed to determine the influence ultra-fine catalyst particles have on coal solubilization.

In order to demonstrate ultra-fine catalyst particle generation, optimization of catalyst particle characteristics, impregnation of coal materials with ultra-fine catalyst particles, and performance of activity testing on resulting catalyst impregnated coal samples, a twelve-month program involving analytical and experimental efforts is being conducted. This is a report on the technical progress made towards the above goals during the first quarter of this program.

The technical progress made during the first quarter of this program fell short of that initially planned. This was due to prior commitments that this project's Program Manager had to attend to before proceeding with this program. The main effort applied during this quarter was preparing and delivering the Program Management Report, which was submitted to DOE on October 25, 1990, and the Project Work Plan (Task 1), which was submitted to DOE on November 30, 1990. Although preparation of these reports consumed most of the time dedicated to this program during the first quarter, some technical progress was achieved, which is summarized in the remainder of this section.

The design of the ultra-fine catalyst generator began by defining system elements and defining specifications for the elements. The elements fall under two principal areas, namely: 1) a supercritical flow system; and 2) a post-expansion flow system for particle collection and coal material impregnation. Design efforts in the first quarter of this program focused on the

first area mentioned above. Specifications for the supercritical flow system were based on the assumption that very severe conditions will be needed to dissolve solid catalyst materials (e.g., Fe_2O_3 and FeS_2). It was determined, therefore, to design the supercritical flow system around conditions achievable when using water as the supercritical fluid. Based on the work done by Dean Matson at Battelle Northwest Laboratories regarding generation of ultra-fine quartz particles via supercritical expansion of water, upper bounds for the thermodynamic conditions for our system were established.

The most severe conditions will occur at the expansion nozzle, namely: 600 atm and 600°C. A nozzle throat diameter of nominally 50 μm will be used which will require a pump capable of at least 12 cm^3/min of water at 600 atm. The specifications for the autoclave dissolution element were defined to be 600 atm and 500°C based on what may be required to achieve good catalyst material dissolution. Based on the flow rate stated above, an autoclave volume of nominally 350 cm^3 will give autoclave residence times sufficiently long to achieve reasonable catalyst dissolution. The length of the transfer tube that delivers the supercritical solution from the autoclave to the expansion nozzle was determined to be nominally 2.5 m with an ID of 0.5 mm. This length was determined based on heat transfer considerations. This line will be heated resistively by passing a DC current through it. It will be heated for the purpose of increasing the temperature of the supercritical solution above the temperature dictated by the autoclave prior to expansion.

A plumbing schematic for the supercritical flow system was generated, together with a parts list containing part numbers, suppliers, specifications, and cost. These materials are currently being ordered. Based on conversations with Dean Matson, Battelle Northwest Laboratories, we have found that the severe conditions associated with supercritical water will leach iron from

materials of construction that come in contact with it. As a result, these critical elements will be made of high strength, low iron, superalloys, namely Hastelloy. We did not anticipate this during the proposal phase of this program, and since Hastelloy components cost 2-3 times that of stainless steel components, we expect the cost of system fabrication to be greater than that proposed.

Personnel safety is also an issue with this system. The autoclave will be operating under very severe conditions (600 atm and 500°C) and the effects of an explosion must be factored into the overall system design. Since some of the materials that will be passed through this system can have severe health effects (e.g., $\text{Fe}(\text{CO})_5$), good ventilation and good practices for handling these materials will be necessary. To minimize personnel harm from explosion, the entire system will be contained in an explosion-proof enclosure. This enclosure will have an adequate ventilation system. A fume hood will be installed near the apparatus for safely loading harmful substances into the autoclave or pumping system supply vessel. OSHA regulations dictate that an emergency eyewash/shower must be located near the fume hood. These support systems were not envisioned during the proposal phase of this program. TDS is helping to minimize the impact these systems will have on this program's budget by covering support system parts costs and associated installation costs.

Although we have experienced a slow start in this program, progress has been otherwise satisfactory. If TDS cannot recover lost time as the program continues, a no-cost extension will be requested. It is clear that program costs for design and fabrication will exceed those anticipated at the proposal phase. With assistance from TDS (such as that mentioned above regarding safety systems) and good management practices, the program goals should be achievable within budget.

2.0 TECHNICAL APPROACH

2.1 PROGRAM OBJECTIVES

In the absence of significant developments in new catalyst compounds, it is generally agreed upon that major improvements in the performance of the solubilization stage of coal liquefaction processes will be achieved through use of once-through, dispersed catalysts with emphasis placed on optimizing the effects of catalyst size, size uniformity, and dispersion. Most importantly, catalyst activity has been shown to increase significantly with decreasing particle size.⁽¹⁻³⁾ Experimental evidence suggests it is reasonable to expect significant improvements in coal solubilization by using catalyst particles with good size uniformity and mean size less than 400 Å. Methods or processes for generating these ultra-fine catalyst particles have not been defined and are needed. The current program responds to this need.

The overall objective of the present program is to evaluate the extent to which ultra-fine, unsupported catalyst particles enhance the performance of the solubilization stage of two-stage catalytic-coal liquefaction processes. The ultra-fine, unsupported catalyst particles will be generated in situ (in coal or coal liquids) by rapid expansion of a supercritical solution containing dissolved catalyst material. To achieve the overall objective of this program, a twelve-month experimental program, supported by analytical efforts, is being conducted by TDS.

The specific objectives of the present program include: 1) design and fabrication of a supercritical flow system for generation of ultra-fine particles relevant to coal solubilization; 2) demonstrate generation of ultra-fine catalyst particles; 3) optimize catalyst particle characteristics (size

and size distribution) by systematically varying process parameters; 4) impregnate coal and coal-liquid samples with ultra-fine particles; and 5) perform liquefaction tests on the impregnated coal samples to evaluate the activity of the ultra-fine, unsupported catalyst particles generated via the supercritical solution expansion concept. The analytical efforts applied during this program will mainly be related to test rig design and correlation of experimental data.

2.2 CONCEPT DESCRIPTION

The program being conducted by TDS represents a new and innovative approach for dispersed coal liquefaction. More specifically, this program is designed to explore a novel concept for generating catalyst particles in a size range ($< 400 \text{ \AA}$) that should significantly enhance the performance of the solubilization stage of two-stage, catalytic-catalytic, coal liquefaction systems.

The concept forming the basis of TDS's approach to generation of ultra-fine catalyst particles involves the use of supercritical fluids (e.g., CO_2 , H_2O , etc.) and exploits two key characteristics of these fluids. Most iron-based catalyst compounds suitable for solubilizing coal have negligible solubility in organic solvents or water even at the thermodynamic conditions characteristic of the coal solubilization stage of liquefaction processes. When acted upon by a supercritical fluid, however, substances having otherwise negligible solubility exhibit modest solubility. A good example of this behavior was reported by Matson,⁽⁴⁾ where concentrations of quartz (SiO_2) of nominally one percent (molar) in supercritical water (400°C , 590 atm) were achieved. It is expected, yet unconfirmed presently, that the iron-compound catalysts suitable for coal solubilization will behave similarly. Thus, the first characteristic of supercritical fluids key to the viability of the proposed concept is their ability to dissolve materials that otherwise have negligible solubility.

The second key characteristic of supercritical fluids is that an extremely rapid density change can be achieved by expanding the supercritical solution through a pinhole orifice or capillary restrictor tube. When a supercritical fluid solvent saturated with an otherwise low solubility solute (e.g., iron-compound catalyst) is rapidly expanded through a pinhole orifice to atmospheric pressure, the drastic density decrease of the expanding supercritical fluid leads to highly nonequilibrium conditions that cause the solute to undergo extremely rapid supersaturation, followed by nucleation and particle growth. After a short distance from the orifice, background gas entrained in the underexpanded jet dilutes the jet causing particle growth to cease. The time over which a fluid element experiences conditions favoring nucleation and growth can be measured in microseconds. The short duration of these conditions leads to the formation of extremely small, nonvolatile solute particles. If the preand post-expansion conditions are selected properly, the particles formed will be suspended in a low density, dry gas. Thus, the second key characteristic of supercritical fluids is their ability to transition very rapidly from a dense gas to a low density gas in an expansion process, which provides conditions appropriate for ultra-fine particles formation.

The impregnation of coal and coal liquids with ultra-fine catalyst particles can be achieved by flowing the post-expansion, particle laden, low density gas through a fluidized bed containing coal material particles/droplets, a chamber containing a stream of free-falling coal material particles/droplets, or a spouted bed reactor containing coal material particles/droplets.

The process of particle formation by rapid expansion of fluid solutions was developed at Battelle's Pacific Northwest laboratories for the purpose of providing a new analytical technique for characterizing supercritical fluids.

Based on their work and TDS's work in coal-related technologies, it was recognized that the supercritical concept could be suitably modified and exploited for generation of ultra-fine catalyst particles for coal liquefaction processes.

2.3 PROGRAM APPROACH

As indicated in Section 2.1, the overall objective of this program is to evaluate the extent to which ultra-fine unsupported catalyst particles generated via the supercritical concept enhance the performance of the solubilization stage of two-stage catalytic-coal liquefaction processes. In order to meet this objective, an experimental program supported by analytical methods is being conducted. The specific objectives are: 1) design and build a supercritical flow system for generating ultra-fine catalyst particles; 2) demonstrate generation of ultra-fine catalyst particles; 3) optimize catalyst particle characteristics by systematically varying process parameters; 4) impregnate coal and coal-liquid samples with catalyst particles; and 5) perform liquefaction tests on impregnated coal and coal liquid samples to determine the activity of the ultra-fine, unsupported catalyst particles. Analytical efforts will be applied during this program, but will be related mainly to test rig design and correlation of experimental data.

The first phase of the program will be to design and fabricate a flexible test rig that can be used for investigating generation of ultra-fine catalyst particles by means of the supercritical concept. The design will allow independent control of process parameters and utilization of solid and liquid phase catalyst feedstock. It will have an expansion chamber in which catalyst particles can be collected. It will also provide apparatus for dispersing catalyst particles into coal materials. Temperature, pressure, and flow rates at critical points in the system will be monitored by suitable diagnostic

instruments. A sampling port will be installed in the supercritical fluid transfer line just prior to the expansion nozzle so samples can be extracted for composition analysis. During the design phase, an experimental test plan will be defined for the remainder of the program work.

The experimental tests to be conducted during the remaining phases of the program can be divided into three main areas: 1) solute/solvent optimization; 2) particle generation and optimization; and 3) catalyst performance evaluation. The purpose of the solute/solvent optimization studies is to determine process parameters (i.e., supercritical solvent type and pre-expansion temperature and pressure) which yield the greatest solubility for a given catalyst compound. The purpose of the particle generation and optimization studies is to systematically vary process parameters around those values defined during the solute/solvent optimization effort in order to achieve the desired catalyst particle characteristics, namely: mean sizes in the range 2 to 40 nm, with particle size distributions ± 20 percent of the mean value. The purpose of the catalyst performance evaluation studies is to impregnate a coal and a coal liquid with ultra-fine catalyst particles, determine their activity via liquefaction testing, and compare their performance to uncatalyzed coal liquefaction results and with current catalytic methods.

The work to be performed under this contract has been divided into tasks as shown in Figure 2.1. The original schedule for this program, including milestones, is presented in Figure 2.2. An updated schedule for this program is presented in Figure 2.3. A comparison of the two schedules shows that the completion milestone for Task 2 is delayed by approximately one to two months. The delay is due primarily to difficulties encountered by the Program Manager in freeing himself from existing commitments.

The basic design and assembly of the particle generator is taking place under Task 2. Although no overlap exists between Task 2 and Tasks 3 and 4,

Figure 2.1 Program Work Breakdown Structure.

DOE F1332.3
(11/84)

U.S. DEPARTMENT OF ENERGY
MILESTONE SCHEDULE PLAN

TITLE: Liquefaction of Coal REPORTING PERIOD: PLAN IDENTIFICATION NUMBER: DE-MC22-90PC90053

PARTICIPANT NAME AND ADDRESS:
Avco Research Laboratory
2385 Revere Beach Parkway
Everett, MA 02149

START DATE: 9/25/90

COMPLETION DATE: 9/24/91

ELEMENT CODE:	REPORTING ELEMENT:	DURATION:	FY 1991												FY 1992				PERCENT COMPLETE:				
			OCT	NOV	DEC	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC	JAN	FEB	MAR	Q.3	Q.4	PLAN
01.01	Work Plan	12-11	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II
01.02	Design	12-11	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II
01.03	Solute Optim.	12-11	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II
01.04	Catalyst Prep.	12-11	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II
01.05	Reports & Mgt.	12-11	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II	II

MILESTONE LEGEND:

- * = Start/Complete
- A = Submit Project Work Plan
- B = Sample Delivery to DOE
- C = DOE Results Delivered to ARL
- D = Final Report & Topical Report

STATUS LEGEND:

- "=" = Work To Go
- "f" = Progress to Date

PROJECT MANAGER SIGNATURE:

[Signature]

H.M. Appich

DATE: 10/19/90

Figure 2.2 Original Program Schedule.

"G053SCHD" COAL LIQUEFACTION

DOE F1332.3
(11/84)

U.S. DEPARTMENT OF ENERGY
MILESTONE SCHEDULE STATUS REPORT

Page 1 of 1

TITLE: Liquefaction of Coal

REPORTING PERIOD: 11/26/90 thru 12/30/90

IDENTIFICATION NUMBER: DE-AC22-90PC90053

PARTICIPANT NAME AND ADDRESS:
Avco Research Laboratory
2385 Revere Beach Parkway
Everett, MA 02149

START DATE: 9/25/90

COMPLETION DATE: 9/24/91

ELEMENT CODE:	REPORTING ELEMENT:	DURATION:	(Time Now)												PERCENT COMPLETE:										
			GFY 1991												GFY 1992										
			OCT	NOV	DEC	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC	JAN	FEB	MAR	Q.3	Q.4	PLAN	ACTUAL	
01.01	Work Plan	1*#(2)###A																					100	100	
(Project Work Plan submitted 11/30/90)																									
01.02	Design	(1)11*###== (2)=*																					88	14	
01.03	Solute Optim.																						0	0	
(1)11*###== (2)=*																									
01.04	Catalyst Prep.																						0	0	
01.05	Reports & Mgt.	1*#####																					22	24	
D																									

MILESTONE LEGEND: (Time Now)

- * = Start/Complete
- A = Submit Project Work Plan
- B = Sample Delivery to DOE
- C = DOE Results Delivered to ARL
- D = Final Report & Topical Report

STATUS LEGEND:

- "_" = Work To Go
- "#" = Progress to Date
- (1) = Originally Planned Start Date
- (2) = Originally Planned Completion Date

PROJECT MANAGER SIGNATURE:

H.M. Eppich
H.M. Eppich

DATE:

1/4/91

Figure 2.3 Updated Program Schedule.

which are Solute/Solvent Optimization and Catalyst Preparation and Evaluation, respectively, modification to the basic design and a final design for the particle generator will be conducted under Task 2, but will take place during the time frame of either Task 3 or 4. This is to say that the milestone delineated by the end of Task 2 corresponds to the completion of the fabrication of an untested particle generation system. The duration of Task 2 should be extended to overlap with the early stages of Task 3, but was not defined this way due to an oversight on TDS's part. Regardless, there will be feedback between the results of shakedown tests performed during Task 3 and system design evolution efforts to be performed, as needed, under Task 2.

3.0 TECHNICAL PROGRESS

3.1 TASK 1 - PROJECT WORK PLAN

OCTOBER 1990: This task was nearly completed during October 1990. The work remaining entails cleaning up the final draft and mailing copies to the appropriate DOE personnel. The preparation of this report was delayed because of prior commitments the Program Manager had to address before proceeding with this DOE program.

NOVEMBER 1990: This task was completed during November 1990. The Project Work Plan was submitted to DOE/PETC on November 30, 1990.

3.2 TASK 2 - EXPERIMENT DESIGN AND FABRICATION

OCTOBER 1990: The planned beginning of this task was the first month of the program. Due to prior commitments of the Program Manager, work to be conducted under this task was not initiated during the first reporting period. This work is presently underway, being performed in a dedicated/focused manner.

NOVEMBER 1990: Design of the Ultra-Fine Particle Generator commenced this month. The effort consisted of dividing the overall system into subsystem elements, identifying key issues associated with each element, and defining a design approach for critical elements. The subsystem elements are: supercritical flow system solvent/liquid-solute feed and pumping system, autoclave solvent/solid-solute mixing section, heated supercritical solution transfer line, temperature and pressure sensors, and HPLC sampling system; expansion nozzle geometries; and expansion chamber design, including coal material mixing sections and pumping system. The expansion nozzle was the first element to receive detailed analysis, since it will set an upper bound on system flow rate, which impacts solvent pump specifications. Arrangements

were initiated to visit faculty at MIT and the University of Lowell who are actively involved in supercritical chemical process development and have existing supercritical flow systems. The purpose of these visits will be to gain insight into the subtleties of generating supercritical solution flows and monitoring its physical properties. Such visits will expedite the supercritical flow system design process.

DECEMBER 1990: Design of the Ultra-Fine Particle Generator continued this month. The apparatus consists of two main elements, namely: 1) a supercritical flow system for generating catalyst particles; and 2) a post-expansion flow system for collecting catalyst particles and for impregnating coal or coal-liquids with catalyst particles. The design activities conducted during this reporting period focused on the design of the supercritical flow system aspect of the experimental apparatus. More specifically, a detailed flow system schematic was generated plus a detailed list of parts. The parts list includes materials of construction, manufacturers catalog numbers, and cost. The design of this flow system will deliver the required process conditions safely.

The flow system has two liquid pumping systems, one for the primary solvent (i.e., CO_2 or H_2O) and one for either liquid catalyst species ($\text{Fe}(\text{CO})_5$), pre-expansion dilution by primary solvent species, or solubilizing additives (i.e., H_2S or CS_2). These two supply legs will be capable of delivering a maximum of 140 cc/min liquid each at 600 atm. Pressure and thermocouple gauges will be installed on the high pressure side of these legs and the flow rate through each leg will be monitored on the low pressure side via armored flow meters. The supply legs will be fitted with pressure relief valves, which, in turn, will be fitted with flow sensors on the downstream side. The flow sensors will trip a shutdown sequence if the relief valves vent. The

shutdown sequence will include closing electric solenoid valves on solvent, solute, and/or additive bottles or containers, shutting down high pressure pumps, and cutting power to the autoclave heater.

The two supply legs will join just upstream of the autoclave reactor. A single line carrying the flow from both supply legs will enter the autoclave at its bottom. The autoclave will be seated in a sand bath heater. The sand bath heater will be capable of heating the autoclave to 500°C. The autoclave will be made of Hastelloy C-276 and designed to withstand 600 atm at 500°C. These conditions were defined based on the conditions desirable for supercritical water. The autoclave will be loaded in a packed bed manner with Inconel or Hastelloy beads separating layers of catalyst material. The supercritical solution will exit the autoclave at its top.

The supercritical solution, after exiting the autoclave, will enter a Hastelloy supercritical transfer line which will heat the solution prior to expansion to a maximum temperature of 600°C. The transfer line will be heated resistively by passing a DC current through the transfer tube. A metering valve will be installed in the downstream end of the transfer line followed by pressure and thermocouple gauges and the nozzle assembly. A laser drilled orifice in a thick disk (.05 to .1 in.) will be used for the nozzle initially, which should approximate the converging side of a Laval nozzle. This will be the first nozzle geometry to try and if it does not perform satisfactory the thin orifice plate or restrictor tube types will be tried. The laser drilled thick plate orifice nozzle is preferable due to its shape and structural integrity.

It is understood that the nozzle expansion process will affect particle size distribution. Some effort, therefore, was invested in developing a basic understanding of the aerothermal characteristics of a highly underexpanded

jet. TDS has a unique capability in this area. Our CFD personnel have a great deal of computational experience in this area due to their activities in predicting hypersonic reacting flows. Our quick look at the expansion flow for this process revealed that extremely high Mach numbers and low temperatures occur just upstream of the Mach disk. The Mach disk is actually a very strong normal shock which achieves pressure matching with the discharge plenum pressure. The Mach number of the flow downstream of the Mach disk is subsonic. The barrel shock surrounding the expansion region between the nozzle exit plane and the Mach disk achieves pressure matching at this boundary by sharp redirection of the momentum flux through the shock barrel boundary. Since the shock barrel is an oblique shock, the flow outside of the shock barrel is still highly supersonic. Thus, immediately downstream of the Mach disk, the core of the flow is subsonic, but is surrounded by a supersonic annular flow. Proceeding further downstream, viscous effects cause the core to become supersonic. Even further downstream, the flow field finally takes on characteristics of a self-similar, fully developed, decaying supersonic jet. The supersonic jet should decay to a flow of negligible momentum in of order 1000 orifice exit plane diameters (several inches). Because of the flow structure appearing in the Mach disk region (i.e., a subsonic core flow surrounded by a supersonic flow), fluid elements will experience different thermodynamic conditions for different periods of time. This may ultimately control particle size distributions. If this result is detected experimentally and prohibits achieving the desired particle size distributions, it will become necessary to design a diverging nozzle which gives uniform properties across the jet cross section.

3.3 TASK 3 - SOLUTE/SOLVENT OPTIMIZATION

No work has been performed under this task as of yet.

3.4 TASK 4 - CATALYST PREPARATION AND EVALUATION

No work has been performed under this task as of yet.

3.5 TASK 5 - REPORTING AND DOCUMENTATION

OCTOBER 1990: A Management Plan was prepared and submitted to DOE on October 25, 1990. A Milestone Schedule Plan (Form 1332.2) was prepared and submitted to DOE on October 22, 1990. A Cost Plan (Form 1332.7) was prepared and submitted to DOE on October 22, 1990. DOE Form 538 "Notice of R&D Project" was completed and submitted to DOE on October 25, 1990. Based on a discussion with our DOE Project Manager, Dr. Udaya S. Rao, a tentative date of January 8, 1990 was defined for the program kickoff meeting.

NOVEMBER 1990: By the end of November 1990, status, milestone, and cost management reports had been submitted for the first two reporting periods (October and November, 1990). Based on a telephone discussion with our DOE Project Manager, Dr. Udaya Rao, the tentative date for the DOE/TDS kickoff meeting was changed from January 8, 1991 to January 22, 1991.

DECEMBER 1990: By the end of December 1990, status, milestone, and cost management reports had been submitted for the first three reporting periods (October through December, 1990). Based on a telephone discussion with our DOE Project Manager, Dr. Udaya Rao, the date for the DOE/TDS kickoff meeting has been set for March 11, 1991. The first Quarterly Progress Report is in preparation and should be delivered to DOE/PETC soon.

4.0 CONCLUSIONS AND FUTURE WORK

4.1 CONCLUSIONS

Several system associated conclusions can be stated at this time. Based on discussions with Dean Matson, Battelle Northwest Laboratories, it has come to our attention that the severe thermodynamic conditions of supercritical water will cause leaching of iron from materials of construction that come in contact with it. To minimize this, Hastelloy, which is a low iron superalloy, will be used for those system components that are vulnerable, namely: the autoclave chamber; supercritical solution transfer line; certain valves and connecting fittings; and nozzle assembly. The cost of Hastelloy components is nominally 2-3 times that of stainless ones. We expect, therefore, that fabrication costs will exceed those envisioned when preparing the proposal for this work.

Since the system is being designed so that the severe conditions of supercritical water can be achieved (600 atm and 600°C), safety precautions must be implemented to protect personnel from explosion and dispersion of harmful substances. An explosion-proof enclosure will enclose the experiment. The enclosure will have a ventilation system that will prevent dispersion of harmful substances. A fume hood will be installed next to the experiment for loading harmful substances into supply vessels that will feed these substances into the supercritical fluid pumping system. An emergency eyewash/shower will be installed next to the fume hood, which is required by OSHA regulations. These precautions were not envisioned during the preparation of the proposal for this program and will add to the cost of this project. TDS will help minimize the impact of these costs by covering the costs for needed components and the installation costs for these subsystems.

4.2 FUTURE WORK

The scope of the work that will be performed in the next quarter is listed below.

- o Continue Task 2 effort (Experiment Design and Fabrication).
- o Prepare presentation for March 11, 1991 kickoff meeting.
- o Finish 1st Quarterly Report.

5.0 REFERENCES

1. Davidson, R.M., "Mineral Effects in Coal Conversion," Report No. IC\IS/TR22, International Energy Agency Coal Research, London, January 1983.
2. Gray, D. and Neuworth, M.B., "Disposable Catalysts in Coal Liquefaction: The Effect of Iron Sulfides," DOE Contract No. DEAC01-77ET10280, MITRE Corp., McLean, VA, September 1981.
3. Andres, M., Charcosset, H., Chiche, P., Davignon, L., Djega-Mariadassoa, Joly, J., and Pregermain, S., Fuel, 62, 69, 1983.
4. Matson, D.W., Norton, K.A., and Smith, R.D., Chemtech, 19, 480, 1989.

**DATE
FILMED
8/10/92**