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# SYNTHETIC FUELS DEVELOPMENT

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#### FF-4

#### SYNTHETIC FUELS DEVELOPMENT

Cadre Members: J. Cowles, AEC, J. P. Nichols, ORNL Overal Coordinator, Ail Coai Programs: J. C. Bresee, AEC Liquefaction Coordinating Laboratory Representative: D. E. Ferguson, ORNL -NOTICE-This report was prepared as an account of work sponsored by the United States Government, Neither the United States nor the United States Energy Research and Development Administration, nor any of Supporting Laboratory Representatives: G. Burnet, Ames their employees, nor any of their contractors, subcontractors, or their employees, makes any varranty, express or implied, or assumes any legal liability or esponsibility for the accuracy, completeness S. Lawroski, ANL M. Steinberg, BNL or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights. Gasification Coordinating Laboratory Representatives: W. Ramsey, LLL D. Stephens, LLL Supporting Laboratory Representatives: R. Salmon, ORNL K. Mudge, PNL L. Booth, LASL S. Lawroski, ANL Page 4-1 Summary. . . . 4-1 Introduction . . . . . . . . . • • . . . 4 - 2Objectives . . . . • • . • . . . • . 4 - 4. . Scope. . . ٠ • . . • • ٠ . . . 4-4 Present & Proposed Five-Year Programs for Synthetic Fuels 4-7 Process Development . . . . . . . . . . 4 - 7Liquefaction Process Development. . . . . . 4-16 High Etu Gas Process Development. . . . . . 4-19 Recommendations. . . . . . . . . . . . . . . • . 4 - 20AEC Laboratory Contributions. . . . -. • • . • 4-21 Program Balance . . . . . . . . . . . . . . . 4-21 . Manegement. . . . . . • . . . • . 4-22 . Miscellaneous . . . . . . . . . • . • . . 4-22 Comments by Laboratory Representatives . • . . • . . . . 4A-1 .

#### IV. SYNTHETIC FUELS DEVELORMENT

#### SUMMARY

The objective of a Synthetic Fuels Development Office of the Coal Programs Division will be to develop a broad base of process chemistry and engineering that will provide a continuing source of methods for process improvements and new process concepts. The types of projects to be managed through this office will consist of (1) laboratory and bench scale work to develop process chemistry, (2) work at the process development scale (up to 10 tons coal/day) to provide engineering concepts and data for process evaluation and scaleup, and (3) supporting work in such areas as materials and component equipment. The projected FY 1975 funding for this office is shown in the following table:

	Transferred From:	FY 1975 Obligations (\$ millions)
Liquefaction Process Development	OCR USBM	30.6 <b>26.</b> 7
High Btu Gas Process Development	OCR USBM AEC	17.7 13.9 <u>3.3</u>
Total		62 5

#### INTRODUCTION

Petroleum and associated gas, fortunately, often occur together in nature. At the well head, simple disengagement techniques can separate fixed portions of natural gas and crude oil. The oil can then be refined to a wide variety of liquid fuels.

When coal is converted into gas and liquid phases, the proportions can be controlled by plant design. The goal of a plant for fuels production from coal is not just producing gas or liquids from coal, but producing gas and liquids in the most desirable proportions. Thus, we have proposed high BTU gasification and liquefaction of coal be combined in one ERDA office in this report. Each gasification and liquefaction process produces

a characteristic mix of solid, liquid, and gaseous fuels in the initial reaction stages. This mix must be converted by subsequent refining operations to the most profitable product mix. The optimum flowsheet for these operations often appears to be an integrated combination of current gasil...ation and liquefaction flowsheets. Liquefaction processes in particular, by judicious alteration of controllable process parameters, can produce widely varying proportions of char, heavy fuel oils, distillate fuels, and gaseous products. We believe the best coal gasification and liquefaction fuel planning car be accomplished by a single management group responsible for setting overall project priorities in the light of the national need for coal derived fuels. The option of combining the best sets of unit gasification and liquefaction operations to be responsive to national needs should make this group much more flexible than two separate liquefaction and gasification groups.

The primary focal point for coal liquefaction and gasification in the Federal Government has been and continues to be the Office of Coal Research (OCR), although the Bureau of Mines (USEM) plays a substantial role. NSF and AEC also have coal synthetic fuel projects. In 1975 OCR will fund over half the government R&D in liquefaction and gasification and perhaps 90% of the pilot plant and larger projects in these areas. Interestingly, OCR has just undergone a reorganization which included transferring some key USEM personnel into OCK. This might reflect implementation of a plan to draw together all coal R&D under one group in the Department of the Interior. The new OCR organization is shown in Fig. 4.1.

#### OBJECTIVES

The objective of the Coal Conversion R&D Program is to develop - on an accelerated and cost effective basis - a base of technology that will permit the rapid commercialization of coal conversion processes that are technically and economically viable, are environmentally acceptable, and represent a wise utilization of national resources. In achieving this objective, a development process is being conducted that evolves through the stages of (1) laboratory and bench scale, (2) process development scale, (3) pilot and prototype plant scale, and (4) demonstration and pioneer plant scale.



#### SCOPE

The scope of this section is restricted to the first two steges of commercialization, the latter two being dealt with in a subsequent section. Here we deal with coal conversion programs on the process development scale and below, including related applied research work delineated by problems anticipated in the various considered processes.

#### OVERVIEW

For purposes of discussion and a broad overview the following definitions are advanced to define these various scales of development in terms of capacity and capital cost.

Table 4.1 Scales of Process Development

Capacity (tons coal/day)	Capital Cost (\$ millions)		
<1	<1		
110	4-10		
50-80	15-30		
200-1,000	50-80		
5,000-10,000	200-500		
20,000-30,000	400-800		
50,000	800-1,500		
	Capacity (tons coal/day) <1 1-10 50-80 200-1,000 5,000-10,000 20,000-30,000 50,000		

The function of laboratory and bench scale units is primarily to establish the scientific feasibility of a process. The work normally is conducted in batch or semicontinuous units that represent only the key steps of a potential process.

The function of a Process Development Unit (PDU) is to establish the engineering feasibility of a process. The PDU normally provides the first opportunity to evaluate engineering aspects such as component designs, materials of construction, and adaptability for scaleup.

Pilot and prototype plants have the objectives of determining the engineering viability of the process and providing data for scaleup. In general, a prototype scale plant has the advantage that it needs to be scaled up by only a factor of 10 to 20 to the first trains of commercial plants and may utilize the smallest sizes of commercial equipment of the type that would be utilized in a commercial plant.

Demonstration plants and pioneer plants have the objective of establishing the economic viability of a process. The demonstration plants normally consist of a first process train that later may be replicated to provide for full commercial scale. A pioneer plant is the first commercial, multi-train plant of a given type.

In general, a strategy for process development (especially for liquefaction processes, which are currently in an early stage of development) that evolves through the stages of (1) bench, (2) process development, (3) prototype plant, and  $(\frac{1}{2})$  pioneer plant is recommended. The following presents specific recommended strategies within each scale of operation.

Laboratory, Bench. and PDU Scale. - In view of the risks of failure in establishing coal conversion technology, the complexity of conversion processes (e.g., effects of many types of coal), and the very high costs of pilot, prototype, demonstration and pioneer plants, we believe that it is fundamentally sound policy and inexpensive insurance to provide for (1) a broad and deep base of supporting technology in process chemistry, process engineering, and environmental effects, (2) support of many alternative and competing concepts through the PDU scale, and (3) multiple, independent PDUs for each of the prime candidate processes.

The bench scale unit and the PDU are the basic tools of process development. They must continue to operate even during the operation of larger plants to provide a testing ground for process variations. Duplication of effort at the bench and PDU scale is not necessarily wasteful. We need to diversify our process development base; we should have a large number of bench and PDUs throughout the country. Different qualified investigators do not "duplicate" each other's work; their efforts inevitably differ in small but often vitally significant details.

<u>Prototype Scale</u>. - A prototype scale plant is generally recommended as the next scale of development after a PDU based upon the following considerations:

(1) It represents a technically feasible scaleup by a factor of about 100 with a relatively acceptable cost of failure.

- (2) Such a plant (nominally a factor of 10 larger than a pilot plant) normally is large enough to utilize the types of equipment (vessels, pumps, filters, compressors, heat exchangers, etc.) that would be employed in a commercial plant and would require a scaleup to single trains of full-sized commercial plants by factors of only 10 to 20.
- (3) As opposed to the direct scaleup from pilot plant size to the first train of a commercial unit (e.g., a typical scaleup from 50 to 5000 tons coal/day), the prototype plant provides for (a) considerably less technological risk, (b) lower capital investment (e.g., \$60-80 million vs \$200-500 million), (c) faster completion (perhaps 30 months vs 36 months), and (d) greater facility for rapidly making necessary process and equipment modifications.

<u>Demonstration/Pioneer Scale.</u> - Normally, sufficient data would be available from prototype scale operation to enable the construction of pioneer commercial plants which would be constructed with a very high percentage of private funds.

The FDU, prototype, pioneer plant strategy outlined above is the strategy that has been chosen by Hydrocarbon Research Inc. for development of their H-Coal process and, also, is the announced strategy of EXXON for development of their extraction-carbonization process. One proposal for development of the solvent refined coal process is based upon an existing 6-ton/day (Wilsonville, Alabama) and a 900-tou/day prototype plant that would be the forerunner of a pioneer plant.

#### PRESENT AND PROPOSED FIVE-YEAR PROGRAMS FOR SYNTHETIC FUELS PROCESS DEVELOPMENT

Funding obligations for FY 1974 and the period FY 1975-FY 1979 for proposed synthetic fuel development projects that would be transferred from the OCR, USEM, and AEC are shown in Table 4.2. The following sections will provide brief descriptions of the individual projects in liquefaction and high-Btu gasification.

#### Liquefaction Process Development

#### 1. Direct Hydrogenation Processes

#### FY 1974 \$6.71 million FY 1975 \$19.8

This activity involves the continued development of processes for the catalytic hydrogenation of coal in coal-oil slurries. It has the objectives of providing basic support for the H-Coal process and continuing the development of fixed bed processes such as those currently under development by the Bureau of Mines (Synthoil) and Gulf Oil.

This is the highest priority liquefaction activity and embodies projects involving supporting technology and process development units of various sizes. Hydrogen, at elevated pressure and temperature, is added directly to coal to liquefy and desulfurize it, first to provide a clean heavy fuel oil and then to upgrade the fuel. Various catalysts and various reactor styles are being evaluated. Processes for power plant fuel oil will be emphasized in the classification, as well as ancillary projects to also refine the product into gasoline and diesel fuel, and to use some product oil and waste solids to make the needed hydrogen from steam. These processes use a fixed or ebullated catalyst beds in the reactor with turbulent flow of hydrogen and coal slurry to maintain operability. New throw-away catalysts will be evaluated in a Berguis-type reactor wherein catalyst is slurried with the coal. A few unusual catalytic systems are also included. Use of nescent hydrogen as obtained from carbon monoxide and water will be evaluated for coal liquefaction.

The process development work being performed by Gulf Oil involves fixed-bed catalytic hydrogenation of coal in a coal-oil slurry. Key features of this process are: (1) use of a simple, unique reactor design

		Funding Colligations by Fincal Year (§ millions)							
	•	Actency	1974	1975	1976	1977	1978	1979	1975-1979
iquel.	action Process Development		4						
1.	Direct Hydrogenation Processes	och Usixa	4.79 <sup>7</sup> 1.95	4.0 15.3	16.0 19.9	38.0 10.9	21.0 10.5	6.0 12.0	85.0 68.1
2.	Extraction-Hydrogenation Processes	OCR	0.8	3.0	1.5	-	-	•	4.5
3.	Pyrolysis Processes	OCR	1.5	7.6	9.0	8.0	• 5.0	. •	29.6
4.	Fischer-Tropsch Variations	och Usini	2.0 0.13	7.8 . i.0	6.0 2.5		1.5	- 1.5	13.8 16.0
5.	New Processes	ocr USBN	1.01	6.5	14.0 1.5	7.7 2.0	7,5 1,3	7.1 1.8	1:2.8 6.6
6.	Engineering Support	OCR	. 0.9	2,2	8.7	4.8	3.0	3.0	21.7
7.	Process Development Support	och Usirm	1.7	4.5 7.6	4,5 <u>9.0</u>	11.0 9.0	4.0 9.0	7.0	21.0 41.5
Total	Total .		14.75	63.0	91.6	90.9	6::.8	· 42.4	350.7
ligh I	atu das Process Pevelopment					•			
1.	Steam-Iron Process	oca	4.67	5.0	3.5	-	•	-	9.5
2.	Liquid Phase Methanation	oca	0, 3%	L.O	0.5	-	- '	-	4.9
3.	Self-Agglomerating Process	0CH	0.07	1.0	1.5	-	-	-	2.5
4,	Hydrane Process	12233	0.35	5.0	7.5	10.5	3.5	3.5	58.0
.5.	lydrogen Generation	0.5421	0.05	1.6	3.0	8.9	3.0	3.0	13.1
6.	Stirred & Slagging Fixed Seda	03%4	-	1.2	6.0	8.5	9.0	6.0	30.7
7.	New Frocesses	ocr UBN1	-	0.7		2.0	ī. 3	<b>i</b> .8	0.7 6.1
8.	Process Development Unggort	ojr Voivi	5.40	5.0 7.0	20.0 8.0	10.0 8.5	10.0 9.0	10.0 5.0	55.0 37.5
9.	In Situ Coal Gasification	VEC UJIN <mark>D</mark>	1.50	3.5 <u>7.0</u>	8.1 <u>5.0</u>	12.ú <u>10.0</u>	14,8 <u>3.0</u>	14.7 <u>2.0</u>	53.7 <u>22.0</u>
	Total .		13.1	35.0	64.1	64.ú	53.6	46.0	264.3
lotal	Process Development Program		27.89	<u>99.0</u>	155.7	155.5	115.4	89.4	615.0

Table 4.2 Survey of Proposed IN 1975-FY 197) Authorizations for Sub-Wilot Plant Scale, Process Development Projects in Synthetic Puels that Nould be Transferred to EREA Under NR 11510<sup>11</sup>

· Based upon FY 1975 budget data provided to Interagency Conl Conversion Tank Force on Nay 9 and May 16, 1974. <sup>b</sup>US3N In Situ Gasification Program presently oriented toward production of low htp gas.

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that allows processing of the coal slurry without plugging of the bed of pelleted catalyst, and (2) use of a commercial catalyst for desulfurization of heavy petroleum oil. The expected primary product is a No. 6 fuel oil. Process development should be completed by FY 1975, including design of the pilot plant, with construction to start in FY 1976.

A PDU unit will be constructed to scale up the USEM Synthoil process (turbulent catalytic reactor) capable of processing 6 to 10 tons of coal per day. The Synthoil process has proved in the laboratory that high sulfur and ash coal can be converted into a premium quality fuel oil that can be burned in power and industrial plants without exceeding pollution emission standards. Fuel tests on Synthoil products will be demonstrated and research conducted to optimize conversion of selected fractions to gasoline and to hydrogen. Direct hydrogenation R&D will also include scaleup of the Costeam process. The Costeam process converts coal to clean liquid fuel by reacting it with carbon monoxide and steam. In addition, a process will be develope. for direct catalytic hydrogenation of coal using inexpensive catalysts.

#### 2. Extraction-Hydrogenation Processes

#### FY 1974 \$0.8 million FY 1975 \$3.0 million

This activity will continue to support the development of extraction processes such as Solvent Refined Coal (SRC), Consol Synthetic Fuel (CSF), and a similar process under development by EXXCN. During solvent extraction, hydrogen is transferred from the solvent to coal yielding a product that is fluid at elevated temperatures where it can be de-ashed to remove pyritic sulfur as well as the other inorganic solids. Pressures and temperatures are advantageously lower, but removal of organic sulfur is not as effective as direct hydrogenation, requiring a second stage of hydrogenation for high organic sulfur coals.

One objective of this activity will be to investigate the technical feasibility and economic potential of the conversion of coal to distillate fuels by <u>extraction and thermal cracking in a delayed coker</u>. The key feature of this process is the use of a delayed coker for carrying out most of the operations including removal of the char and ash. The char and ash would be removed in the delayed coker by conventional hydraulic drilling. If successful, this process could be readily scaled up with a high degree of confidence because the equipment and the related technology are already

#### 3. Pyrolysis Processes

#### FY 1974 \$1.50 million FY 1975 \$7.60

This activity will involve continued development of alternative processes for production of clean coke and clean liquid fuels by fluidizedbed pyrolysis under a controlled atmosphere of hydrogen.

<u>Clean Coke Process</u>. - Bench-scale research by U. S. Steel has been completed and the process development work will be completed about the end of FY 1974. The process development unit is being operated to establish the feasibility of the operation, and studies are being made of process water streams to avoid water pollution and corrosion of plant equipment. The project will be ready for pilot plant in FY 1975. Costs are being shared by industry cosponsorship.

The objective of this project is to enable use of high-sulfur coals for coke production, and to avoid pollution in the coking process. It will also improve the economics of OCR's project for solvent refined low-sulfur, low-ash coal. The clean coke process combines low-temperature carbonization and hydrogenation of coal. The process emphasizes the production of coke and chemical products, but production of liquid fuels can readily be included if desired. The amount of hydrogen required for the chemical products is about one-half that required for liquid fuels. The process requires no external hydrogen supply, and is essentially non-polluting.

<u>Hydrocarbonization-Hydroliquefaction Process</u>. - Development of a liquid fuels version of the clean coke process and preliminary design of a small (10 ton/day) pilot plant will begin. Late in FY 1974 committents for equipment items for the pilot plant will be made. Utilizing technology developed on Project COED and the clean coke process, the Oak Ridge National Laboratory will construct a process development unit for low tem erature carbonization at about 10 atmospheric pressure. Concurrently, an engineering evaluation of the process will be completed for large-scale use of eastern coal to manufacture clean liquids, solids, and gases for use in eastern power plants.

#### 4. Fischer-Tropsch Processes

FY 1974 \$2.10 million FY 1975 \$11.80

Development work will be carried out on the production of liquids from coal by the catalytic conversion of CO and H<sub>2</sub> produced by gasification of coal. This work will include the production of synthetic oil by the Fischer-Tropsch process, and the production of alcohol fuels, including methanol.

#### 5. <u>New Processes</u>

FY 1974 \$1.0 million FY 1975 \$6.50

Molten Salt Processes. - This project is oriented to produce lead-free gasoline of 90 octane or higher from coal. Basic tests show that through the use of zinc chloride as a bydrocracking catalyst the objectives can be achieved. It is intended to carry this project further to provide its feasibility for commercial application. This process has been satisfactorily demonstrated at small scale. Major problem encountered was the recovery of the catalyst.

<u>Flash Hydrogenation</u>. - This project will investigate a liquefaction process which provides for rapid and direct contact of gaseous hydrogen with solid coal at high temperature. Very short contact time for hydrogenation, followed by rapid cooling of the products, is known to minimize repolymerization of the products and favor the production in high yield of light oils (benzene, toluene, and xylene) together with some hydrocarbon gases. A rotating fluidized-bed reactor has a high capacity for coal conversion at short controlled contact time favorable for high yields of liquid products while heating finely divided coal to reaction temperature rapidly with hydrogen gas. Catalytic or noncatalytic processing is compatible with this reaction system.

<u>Chemicals from Coal</u>. - A project by Dow Chemical Company to study the feasibility of using coal liquefaction products for the manufacture of chemicals is proposed which will provide a basis for capital and cost estimates for a commercial chemicals plant. This work will define the economics, estimated preliminary capital requirements, and operating costs for a commercial plant which would use coal liquefaction products for the production of useful alignatic and aromatic compounds. Product samples from several coal liquefaction processes will be subjected to process studies, analytical inspections, and otherwise subjected to laboratory evaluations for characterization. Utilizing data obtained from these studies, inspections, and process evaluations, preliminary process flowsheets, and materials balances will be prepared. Capital and cost estimates will be generated for a commercial chemicals plant.

A small program to produce olefins by fast contact and recycle through a multiple stage combustor/reactor is in the planning stage. Preliminary design and evaluation of reactor chemistry, rates, and yields will be accomplished this fiscal year.

<u>Methanol Liquid Catalysts.</u> - This project has the objective of developing a process for conversion of methanol to high octane gasoline in a catalytic process, using a new catalyst. This process may have the potential for earlier commercial application than other coal liquefaction processes, since the remaining research and development required might be completed in one to two years.

<u>NUCC (Nuclear-Coal) Process</u>. - This nuclear-coal conversion process has high yields of oil and gas with less ecological impact. In this process one-half to two-thirds of the process coal is replaced with nuclear energy, reducing the ash and carbon dioxide. It may be competitive by 1985. Phase I for two years is for demonstrating unit selection, technical and economic verification, development of a detailed program plant, and to define the probability of success.

#### 6. Engineering Support Projects FY 1974 \$0.90 million FY 1975 \$2.20

Liquefaction Data Book. - Handbooks are essential aids to engineers; however, handbooks must present data in condensed forms and with little critical evaluation. They are thus prone to be outdated and are not sufficiently comprehensive for the task at hand. This has led the large engineering firms to develop data books of technical and design information for the specific design and evaluation efforts of interest to them. This is accomplished by groups of experts who evaluate all available fundamental data on a subject, correlate them within the most advanced framework, and then present their findings in the most convenient forms for design engineers to use. Designers are thus freed from the tasks of searching for information; errors of interpretation, extrapolation, and interpolation are minimized; uniformity of design bases is ensured; and greater accuracy of design is achieved. This project would initiate and continue the compilation of a data book for use by the many organizers participating in the national program for conversion of coal to clean fuels. The first year or two will be used to cover existing data. A continuing effort is necessary to keep up with new data and to replace obsolete data.

Keat Exchanger. - The synthesis of fuels from coal, both liquid and gas, is dependent on the addition of large quantities of energy supplied at high temperature for the endothermic reactions of char and steam and the devolatilization of char. In conventional processes, this energy is supplied by the combustion of oxygen with the char, by electric energy, or by an intermediate heat carrier which releases either latent heat or chemical energy. Process chemistry is often adversely affected by the method to supply this energy. Heat pipes can transfer large amounts of thermal energy over their length and through a partition with virtually no temperature drop, and at high heat-transfer efficiency. Therefore, they allow the isolation of the heat source from the heat sink; in this way, heat from an energy source can be transferred from the source and released in areas where a careful control of the process composition is important. The attractive heat transfer properties of the heat pipes are the result of a closed system in which a transfer fluid is continually vaporized, absorbing large quantities of heat at the heat input end, and is condensed, releasing that heat at the output end. Because of the energy transfer capabilities of heat pipes, the evaporator end can be heated by thermal energy in a system that is physically and thermally isolated from the condenser end of the heat pipe where the energy release occurs.

<u>Coal Liquid Pipelining</u>. - After coal has been converted from a solid state to a gaseous or liquid state the products must be transported to the point where they will be used. There are problems which must be solved concerning this transportation. These consist of ecology, viscosity, pumping requirements, friction losses, the volatile nature of some of the by-products, etc. This program will entail engineering and economic studies directed to solving these problems. The end product will be an engineering data book which will be available to designers of future projects.

<u>Characterization of Liquid Fuels from Coal</u>. - It is necessary to determine the physical and chemical characteristics of samples prepared under different conditions of temperature and pressure and flow rate. The products must also be characterized in terms of refining properties and suitability for upgrading into quality liquid fuels. The work of this project is to develop the required analytical methods for accomplishing these purposes. The objective is to enable the selection of the type of conversion process and operating conditions that would provide liquids from coal having the most favorable refining characteristics. The Bureau of Mines facility at Bartlesville, Oklahoma, will be used for this purpose.

#### 7. Process Development Support

#### FY 1974 \$1.70 million FY 1975 \$4.50

Specific Purpose Coal Preparation. - This project is generating new knowledge as to the impact of coal composition, coal mining, and coal preparation on the practical aspects of gasification and liquefaction. Gasification and liquefaction processes will be made more attractive economically, and more trouble-free operationally, if the raw material fed into the system can be made uniform in character and near optimum in composition. Current research objectives include: (a) establishment of the relationship between a coal's composition and its behavior and yield in conversion processes, (b) development of commercially feasible methods for controlling the composition of raw coal to provide process feedstocks which can be uniformly maintained at the desired quality, (c) development of mining and preparation techniques which will increase productivity without decreasing the quality of the product and which will provide for reclamation of the mined land and provide for the useful disposal of refuse materials, and (d) development of fully identified and representative samples of the nation's coal seams for experimental use in defining the utilization potential of various coal types.

Lignite Resource Development. - This project is directel toward development of processes for upgrading lignite into fuel gas, liquid fuels, chemicals, and related products. This requires RAD to determine technologically obtainable economic yields of useful products from lignite, and four types of research are contemplated: (1) methods of discolving lignite in the presence of hydrogen, carbon monoxide, and mixtures of carbon monoxide and steam, (2) carbonization studies to determine yields, and methods of upgrading products, (3) hydrogenation-distillation studies on extract using various catalysts, and (4) studies in continuous low-pressure hydrogenation of lignite under a range of operating conditions. The products will be determined, using typical identification techniques, and typical results will be analyzed for possible development of an integrated process development and possible pilot plant application.

- Research utilizing western U.S. coals in a direct first-stage hydrogenation process to yield synthetic crude oil, solid char fuel and fuel gases is the main objective of this project. Certain properties of this raw synthetic crude (such as low hydrogen content, high oxygen content, sulfur content, etc.) would prohibit feeding this material into a conventional oil refinery. A second stage, therefore, involves hydrotreating to yield a synthetic crude oil which could serve as a feedstock for a modern conventional refinery. Current investigations are also being conducted relative to the production of high quality gasoline in substantial yields from western coals.

Desulfurization. - This ongoing project is a laboratory-scale investigation of process variables which affect the degree of desulfurizing of solvent refined coal and other liquids. (This will be a supporting effort to coal liquefaction projects. Desulfurizing is a very significant part of those processes.) Various coals are being tested for these purposes. The contractor is investigating the independent variables affecting the degree of sulfur removal by hydrogen treating while in solution - to identify those variables significant enough to warrant more complete study. About 25% of their study may be completed in sufficient detail to develop mathematical or graphic model of the effects of the significant variables. The results will be used to develop the process design of a coal desulfurization process in sufficient detail to estimate its economic feasibility. This project supports OCR's project for solvent refining of coal.

#### High Btu Gas Process Development

#### 1. Steam-Iron Process

FY-1074	\$4.67	million
FY-15.	\$6.00	

This activity will continue the development of an alternative process for the production of a hydrogen-steam mixture for use in the Hygas hydrogasifier. In this process, hydrogen is produced by alternately reducing a hot bed of iron oxide with producer gas manufactured from coal or coke, and then decomposing steam by reaction with the reduced iron, thereby reoxidizing the iron and producing hydrogen. The specific objective is the design, construction, and operation of a pilot facility to obtain process and design data for scaleup.

#### 2. Liquid Phase Methanation Process

#### FY-1974 \$0.34 million FY-1975 \$4.00

The Liquid Fhase Methanation Process is being developed for the production of high-Btu gas from coal-derived gases. The process is capable of almost completely converting gas streams containing high concentrations of carbon monoxide into methane in a single pass through the reactor.

In this process, inert liquid is pumped upward through the reactor at a velocity sufficient to both fluidize the catalyst and remove the reaction heat. The low-Btu feed gas is also passed concurrently upward through the reactor where it is converted to a high-concentration methane stream in the presence of the catalyst. The exothermic reaction heat is taken up by the liquid as sensible heat and by vaporization. The overhead product gases are condensed to remove the product water and to recover any vaporized liquid for recycle. The main flow is circulatory through a heat exchanger for temperature control.

Future work will be aimed at selecting a preferred reaction liquid and cativest to use in a more detailed reaction-variable study. A larger scale process-development unit is under construction.

#### 3. Self-Agglomeration Process

#### FY-1974 \$0.67 million FY-1975 \$1.00

This project is for development of a coal gasification fluidized-bed coal burner to provide the heat for steam gasification of coal for producing a "make" the intended for subsequent conversion to pipeline gas. A major feature of the process is the method of combustion, which applies the "selfagglomerating" fluidized-bed technique for burning coal with simultaneous pelletization of the ash during the combustion.

In addition to providing a pelletized heat-transfer medium to the gasifier, the self-agglomerating fluidized-bed burner is effective for collecting the ash contained in the incoming fuel. Thus, the fuel can be burned to yield a combustion gas essentially free of flyash. This particulatefree hot combustion gas can then be expanded in an open-cycle gas turbine for recovery of kinetic energy.

A preliminary design of the pilot plant is being prepared. The objective is to obtain operating data sufficient for technical and economic assessment of the process applied to commercial production of pipeline gas. (The present scope of the program does not include conversion of the gasifier product, for example, through methanation.)

#### 4. Hydrane Process

#### FY-1974 \$0.36 million FY-1975 \$3.0

This hydrogasification process handles all types of coals, requires no pretreatment of the coal to destroy its caking properties, and produces a high methane yield in the gasifier. A laboratory unit is now operating, and a large scale PDU will be designed, constructed, and operated.

#### 5. Hydrogen Generation

#### FY-1974 \$3.06 million FY-1975 \$1.6

Improved methods are needed for the generation of process hydrogen at reasonable cost. This is of great importance to hydrogenification processes. This program will include laboratory, bench scale, and pilot plant efforts.

#### 6. Stirred and Slagging Fixed Beds FY-1975 \$1.2 million

The Bureau of Mines has processed American caking coals with a fixedbed gasifier through the use of a stirrer in the gasifier. Past efforts were aimed at producing low-Btu gas. This project will develop the stirred-fixed bed to produce high-Btu gas, by introducing and testing design changes in the existing gasifier. Scale up to a larger unit is planned. Also, beginning in FY-1976, the USMB will reinitiate the development of a slagging fixed bed gasifier at the Grand Forks Energy Research Center.

#### 7. New Processes

FY-1974 \$0.00 million FY-1975 \$0.7

Novel gasifiers will be tested, including modifications of the Lurgi gasifier, to allow commercial gasification plants to be developed earlier.

#### 8. Process Development Support

FY-1974 \$5.40 million FY-1975 \$13.0

The support projects are for equipment development, materials development, and process development, found to be necessary during the execution of the pilot plant program.

Equipment Development. - Production of high-Btu gas from coal will involve large scale (from 16,000 to 100,000 tons per day) feeding of coal. The coal must be reduced to--and controlled at--closely graded particle sizes. It must then be fed into reactors operating at high pressures and temperatures (1500 psi and 3000°F). Particulate matter must be separated from product gases. Char and ash must be discharged from the reactor. Equipment such as grinders, pumps, valves, separators, instruments, etc., to meet all these requirements of capacity and conditions do not exist. Concepts must be formed; the most nearly suitable equipment must be modified, new equipment must be designed and all these tested and developed before large plants can hope to be operated.

<u>Materials Development</u>. - The new processes being developed for the generation of high-Etu gas from coal utilize combinations of temperatures, pressures, and operating conditions that create considerably more stringent requirements for materials of construction than are required in other sectors of chemical product manufacturing. In order to specify the materials that can survive these conditions for the lifetime of the plant with a high degree of reliability, it is necessary to know and understand the behavior and resistance of many candid te metals and ceramics under actual use conditions. A construction materials test and development program has been initiated to subject a wide range of materials to actual conditions (temperature, pressure, gaseous components) existing in each of the coal gasification pilot plants now being or to be operated. The response of these materials (signs of corrosion, weight loss, embrittlement) to their environment will be recorded, correlated, and will become the basis for materials specification.

<u>Process Development</u>. - A program has been initiated to study, operate, evaluate, and compare various techniques that can be used to manufacture hydrogen in large volumes. Processes now under investigation include the electrothermal gasification of char, the steam-iron process, and the steamoxygen process. As other candidate processes become known and appear suitable for similar evaluation, they will be included in this process development program. The data generated will be used to establish which process for the manufacture of hydrogen should be included as part of a commercial coal conversion plant.

#### 9. In-Situ Ccal Gasification

#### FY-1974 \$1.60 million FY-1975 \$5.3

The Bureau of Mines and the AEC propose studies of the <u>in situ</u> gasification of coal. Laboratory and field work will be conducted leading to large scale demonstration projects before 1980. The AEC concept envisions the in situ production of high Btu gas in deep coal deposits (500 - 3000 ft depth) through the introduction of steam-oxygen mixtures to a burning coal region that has been artificially fractured with chemical explosives. The USEM program is aimed at the production of low Btu gas in shallower coal seems (0-500 ft depth) using the natural permeability of the coal seam to free reactant gas and remove product gas.

#### RECOMMENDATIONS

The requested 1975 funds for coal liquefaction and high-Btu gasification are well over twice the 1974 federal funding level and there is a high probability that most, if not all, of these funds will be appropriated. Thus, there is a sufficiency of projects and concepts being considered for FY-1975 funding. The major need is not for additional money.

A key area of concern in these greatly accelerated programs is the adequacy of the technical base for launching multiple pilot and demonstration scale plants. Of further concern is the small number of people sufficiently experienced to expand this limited technical base. We believe the whole question of maintaining a proper balance of priorities between large empirically oriented plants and the supporting R/D necessary to keep these plants on stream is critical.

Also critical is the question of which processes to accelerate, and when and how fast, i.e., an overall cost-benefit analysis of the national coal R/D options available. The results of this analysis could help set the tone of the long range supporting R/D activities and the allocation of the few coal knowledgeable people available to most efficiently pursue coal R/D. In brief, the most important need in coal liquefaction and gasification is establishment of a good mechanism for picking the optimum R/D route.

The OCR report written to support the FY-1975 budget proposals is a good start in the direction of a more balanced and orderly national coal R/D program. However, there appears to be considerable room for improving project selection criteria and quantifying priorities between various R/D options. We believe this expansion of top level objective program evaluation is a prime requirement of the coal synthetic fuels program.

#### AEC Laboratory Contributions

The AEC Headquarters' staff has been concerned for over a year with the problem of optionally applying the considerable talents of the national laboratories to the critical area of coal R/D. There are a number of high visibility areas where the laboratories could participate in coal R/D, evidenced by the LLL proposed in situ gasification program and the proposals to OCR from ORNL, ANL, ENL, and LASL.

Last summer a request was sent to the national laboratories asking them to evaluate their own potential for doing coal related R/D. The request was made to provide information for George Hill, then Director of the Office of Coal Research, so that he would be cognizant of the national laboratories' capabilities during the national interagency energy planning activities conducted last fell. Since these self-evaluations probably contain the most comprehensive list of 193 coal R/D capabilities, a compendium of these evaluations is incluing in Appendix 4A.

#### Program Balance

- Overall program has been brought into apparently reasonable balance within the broad areas. With current information we cannot provide a better balance between gasification, liquefaction, and combustion than existing inhouse government/industry ad hoc panels now serving in DOI.
- A more sophisticated systems approach will be needed now to guide management in relating funding to often conflicting demands such as those of commercial economics, coal supply, product marketability, capital availability, international policies, national economics, and national security.

#### Management

- 1. AEC, and DOI to a lesser extent, must press Congress for more reasonable management situations if we are to experience results comparable to those of other national commitments, (Manhattan Project, etc.), where local autonomous control by technical management allowed the necessary flexibility of effort. Currently, there is overcontrol in timing and funding. Lesson from industry: sink or swim in individual projects will encourage individual productivity.
- Allow management some flexibility to fund emergency technology, i.e., have sufficiency of discretionary dollars.
- 3. Other institutional bottlenecks should be relieved: liberalize patents, and cost sharing rules, forget simplistic 1/2, 1/3 cost sharing rules, and develop rational criteria.

#### Miscellaneous

- 1. Large R&D centers give flexibility, provide for hifts of effort between projects and conserve resources and manpower. The present empirical programmatic philisophy may be less fruitful then maintaining broad research base. We submit that the national labs should be an integral part of the government fossil program.
- 2. OCR preoccupation with boiler fuels should be reexamined; distillates are a bigger national problem than oil to electricity.
- 3. Low Bt.: gas and methanol are fruitful areas for R&D. We suggest that the national laboratories should set the following priorities: lst - Supporting Technology - bits and pieces of larger projects. 2nd - Liquefaction - small scale projects still available. 3rd - Pressurized Combustion - small scale projects still available. 4th - New power systems, MHD, etc. - room for bench scale work. 5th - Low Etu Gas 6th - High Etu
  - 7th Pioneer plants

#### COMMENTS BY LABORATORY REPRESENTATIVES

The following are comments on the Synthetic Fuels Program that were contributed by representatives of the coordinating and supporting laboratories.

 We believe that a major deficiency of the present program is that it does not provide for the creation of a permanent centralized coal process research and development facility. We mean, for example, a facility

comparable to those owned by the major oil companies. Such facilities have the advantages of (1) continuity of personnel, resulting in thorough familiarity with all aspects of petroleum (or coal) process research and development, (2) strong supporting facilities (analytical, chemistry, product testing and evaluation, process design, engineering design, instrumentation, etc.), (3) centralized organization, so that when a problem is encountered (process, materials, catalyst, etc.), the entire resources of the laboratory can be mobilized, if necessary, to attack the problem without delay, and (4) flexibility, so that process design, process conditions, catalyst formulation, etc., can be modified, when necessary, in accordance with day-to-day results, rather than being locked in according to a predetermined course. Such facilities typically have a large number of cench-scale and process development units, and typically are working on a number of processes simultaneously. Typically also, the people working with the bench-scale and process development units have a close working relationship with the process and mechanical engineers who design the pilot plants and demonstration units. Throughout the development phase, these engineers carry out design studies and economic studies related to process optimization and full-scale application. The same engineers are frequently involved in the startup of demonstration plants.

In contrast to this approach, the present program is aimed at the development of specific processes, and pays virtually no attention to the need for developing permanent large-scale research and development facilities.

. We feel that this subprogram is internally reasonably in balance. The point has been made that there is real need for many diversified, relatively small (bench and process development unit) scale efforts on coal conversion. Levels of effort proposed by ORC and NSF would seem to be adequate to support this effort. If a process innovation leads to reducing the cost of synthetic natural gas as little as 1¢ per million Btu, and that innovation is applied to twelve 250 MAcf/day plants, then consumers would be saved \$10 MM per year.

- 3. We have been unable to identify any important missing tasks within this subprogram. However, there are many areas vital to the commercialization of coal gasification that are outside the scope of this subprogram. For example, critical shortages in process engineering, mining engineering, mining and construction manpower are anticipated. The Project Independence Report on Synthetic Fuels from Coal contains much more information on these issues.
- 4. Numerical modeling is highly useful when closely tied to an experimental program. Our consensus was, however, that it could not be used to skip any of the steps in the bench-PDU-pilot plant-demonstration plant sequence.
- 5. It has been strongly suggested that new pilot plants should be sited where they can make use of a technology and labor base. It is inefficient to have pilot plants so remotely located that, for example, maintenance personnel can be used only at that plant. Also, nearby availability of feed-stocks and markets is not important for pilot plant location.
- 6. We should plan to build on the strength of the national laboratories in basic and applied research. It would seem at every opportunity we should include provision for a research input in our planning. In the case of coal liquefaction, our laboratories have more in common with the Bureau of Mines than with OCR. Perhaps we should be suggesting the merging of the Bureau with the national laboratories under a single administration, to provide the research plus fundamental development component we need. Otherwise, we may find the Bureau and laboratories in competition.
- 7. We should also recognize the urgent need for manpower development in mining, engineering and other energy-related disciplines. Special attention needs to be given to the education and training of the people in these categories who will be needed in increasing numbers. We have in mind a separately recognized program which would provide university grants and matching funds for training, equipment, facilities, etc. The pattern to be followed would be similar to that found in the past in the Division of Nuclear Energy and Training and in NSF.

#### APPENDIX 4A

Summaries of Capabilities for Coal Related Research and Development at:

Argonne National Laboratory (ANL) Brookhaven National Laboratory (BNL) Oak Ridge National Laboratory (CRNL) Lawrence Livermore Laboratory (LLL) Los Alamos Scientific Laboratory (LASL) Sandia Laboratories

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STATEMENT OF THE ARGONNE NATIONAL LABORATORY'S COAL R&D CAPABILITIES

The brief statement below indicates areas of work that are relevant and that as an integrated effort would provide an excellent in-house capability in support of the expanded coal-use development program. An integrated program containing the following elements,

- chemistry programs in the areas of process steps, impurity effects, catalysis, advanced analytical techniques, and new process concepts,
- chemical engineering programs on unit operations and process steps,
- materials studies in the areas such as corrosion, mechanical properties, failure evaluation and system studies, and
- 4. component development programs containing components design and evaluation,

provides a very powerful tool to program management. In addition to the specific areas mentioned, all of these groups would provide an in-house capability not only for performing research but also au in-house capability that could be used in technical evaluation and program planning. A complementary area where Argonne has done work is the problem of strip mine land management and reclamation. We believe our efforts in this area could be expanded significantly. The Argonne effort on noxious pollutants supported by DBER completes the resources of Argonne.

Programs	Dates	Total program costs to date thousands of dollars	Supported	Remarks
Dispersion from tall stacks.	1969-73	1,000	AEC	Advanced capability of modeling plume dispersion
Oxidation of SO <sub>2</sub> in coal and oil fired plumes	1969-73	1,000	АЕС-ЕРА	SO <sub>2</sub> oxidizers less rapidly in coal fired plumes
Oxidation of acid mine drainage	1969-70	100	AEC-FWPCA	Several reports on ozonation and limestone treatment were published
Development of pumpable bolts for coal mine roof support	1970-73	1,500	Bu.Mines	Prototype machine being developed by industrial co., DNL is sub- contractor
Studies of coal-nuclear MHD cycles	1969-71	25	AEC	Several publications in literatur

4A-2

Recent Brookhaven National Laboratory Programs and Capabilities Pertinent to Coal Research

The Department of Applied Science conducts programs in:

- 1) Basic chemistry research including work in polymer chemistry, organic chemistry, surface chemistry, organic crystallography and fast reaction kinetics.
- Materials research including superconducting materials, high temperature materials, and concrete-polymer materials.
- 3) Cryogenics research including superconducting magnet design and construction.
- 4) Process chemistry including separations chemistry, polymer synthesis.
- 5) Mechanical engineering including stress analysis, structural design and reactor design.
- 6) Chemical engineering including heat transfer and reaction vessel design.

The Department in conjunction with Burns and Roe, Inc., has submitted a proposal to OCR on a process fo converting coal to liquid hydrocarbon products in a rotating fluidized bed reactor. Capabilities of the Oak Ridge National Laboratory for .Research and Development on Coal-Related Activities

The Oak Ridge National Laboratory has recently proposed research and development programs on a number of coal-related activities. The largest of these consists of a comprehensive program aimed at the development and early demonstration of practical, economic processes on a commercial scale for converting coal to clean liquid fuels for the transportation sector. Smaller efforts are being proposed on the development of advanced underground mining techniques and on the commercial-scale demonstration of the production of methanol from coal.

The detailed formulation and execution of these activities would be carried out primarily by a group of chemical engineers in ORNL's Chemical Technology Division who are experienced in process development and demonstration. Over a period of 25 years, the Division has taken a number of major processes of world-wide importance from basic chemical studies through the successful design and operation of large, complex demonstration plants. The Division's capability for successfully developing processes through all stages of research and development including the design and operation of multimillion-dollar demonstration plants is believed to be particularly relevant to the proposed coal-related activities. The Division is experienced in the development of equipment and control methods for individual unit operations important to coal conversion processes. The Division is also edept at making in-depth studies and projections in areas such as energy supply and demand, and is familiar with modern techniques for cost-benefit analysis. A notable example is a recent study for defining alternatives whereby a large, coal-fired steam plant could meet proposed Indiana emission standards. Although the Division's activities are related primarily to aqueous-based processes for the nuclear industry, a considerable amount of experience also exists on high temperature, non-aqueous processing and in diverse areas such as bioengineering and the development of high-speed automated analytical techniques for biomedical and environmental applications.

The Oak Ridge National Laboratory has an unparalleled collection of supportive disciplines and facilities that will be indispensable in the successful execution of the proposed activities. These include chemists, metallurgists, ceramicists, biologists, and an integrated central design organization. In particular, the Analytical Chemical Division is experienced at developing techniques for monitoring and analyzing various streams and materials associated with process development. There are important environmental considerations associated with the proposed activities, and the Laboratory has experience in assessing the impact of large, complex operations on the environment.

Finally, the Laboratory has strong ties with the Tennessee Valley Authority and with the University of Tennessee whose central operations are located conveniently to the Laboratory. The Laboratory is also located near important coal-mining operations in the Appalachian region.

#### LLL CAPABILITIES FOR COAL AND OIL SHALE RESEARCH AND DEVELOPMENT

LLL is uniquely qualified to conduct complete research processes from concept through inhouse laboratory research to full scale field execution and demonstration. LLL has extensive laboratory and experimental field testing facilities well supported by one of the largest scientific laboratories in the nation. The laboratory is specifically qualified in the area of natural resource development based on long experience in conceptualizing and executing programs on the massive breakage of rock materials. Specific abilities to do research and development on coal conversion processes and or oil shale, including those developed during our recent feasibility study on in situ coal gasification are:

- High pressure, high temperature equipment for studies of coal and char gasification reactions with steam, hydrogen and carbon dioxide.
- Extensive analytical capabilities for coal gased chemicals including mass spectrometry and on-line chromatography facilities.
- 3. Complete thermal and physical property measuring techniques plus the differential scanning, calorimetry specifically designed to measure water content in natural material, and

equipment do to simultaneous DTA, TGA, and RGA measurements.

- 4. Calculational thermodynamic capabilities include computerized calculations of equilibrium multi-phase compositions in a code to model simultaneous heat and mass transfer, chemical kinetics, and a fluid flow in 1, 2 and 3 dimension is being written for in situ gasification of coal.
- 5. A high pressure rock mechanic laboratory for measurement of mechanic properties and equation of state of natural materials.
- 6. Complete metallurgical labs and extensive experience in problems of hydrogen embrittlement and hydrogen containment of high pressures.
- 7. A background in computerized control of lab experiment and automatic data acquisition systems. This experience is being applied to a large scale adiabatic reactor experiment to simulate in situ gas and shale retorting processes.
- 8. Extensive radiochemical and tritium handling facilities providing the capability for chemical tracer studies with hydrogen and carbon isotopes at very low radioactivity levels.
- 9. Experience in economic assessment of in situ processes related to coal.
- 10. The laboratory Biomedical Division and Environmental Group can perform extensive and environmental impact evaluations.

#### CAPABILITIES OF THE LOS ALAMOS SCIENTIFIC LABORATORY IN COAL UTILIZATION RESEARCH AND DEVELOPMENT

Many of the technical problems which have been traditionally solved at the LASL have involved difficult and unique materials developments and materials handling involving remote operations, experimental diagnostics in extremely adverse environments, and design for high temperature - high stress applications involving nickel base alloys, refractory metals, ceramics, and refractories.

The LASL is a self-contained R&D organization having on-site an architectorial and engineering staff, a standards lab, a large technical library, one of the largest computer facilities in the nation, shops, graphic arts and purchasing departments which support twelve technical divisions (Physics, chemistry, theoretical physics, etc.).

Specific Areas of Possible LASL Research and Development in Coal Utilization

- Physical-chemical measurements (thermochemistry, electrochemistry, physical and mechanical properties.)
- 2. Heat pipe cooling of methanators
- 3. Thermal decomposition/electrolytic recombination topping systems for production of electrical energy from coal.
- 4. Fuel cells indirectly powered by coal
- 5. Production of stable isotope as tracers
- 6. Numerical fluid flow simulation
- 7. Fate in the environment of sulfur
- 8. Fate in the environment of trace elements (mercury, lead, arsenic, selenium and radionuclides)

9. Hydrofracturing techniques to aid in-situ extraction

10. Study of the formation and agglomeration of fines in coal combustion

11. In-Situ gasification using suction flow

12. High temperature gas loop heat exchanger test facility

13. Broad-based research support for the Four Corners power plant.

- 1. Physico-chemical measurements
  - a) Thermochemistry (particularly with respect to the oxides, carbides of refractory metals) enthalpies of formation, heat of vaporization and reactions, specific heat measurements.
  - b) electrochemistry

molten salt electrolytes

c) Physical and mechanical property measurements examples: thermal conductivity, elastic constraints, surface characteristics, crystal structure, high pressure studies, etc. Capabilities of Sandia Laboratories in Coal Related Projects

Sandia has a strong background in both basic materials R&D and physical testing of materials. On the basic research side it is more physics than chemistry oriented. They have also been involved in field programs to study environmental pathways of pollutants.

Sandia is currently performing a feasibility study of in situ coal gasification utilizing high explosive fractured coal seams. The study is based on sophisticated computer codes for response of materials to shock waves. Similar codes are being used to study optimum water jet drilling techniques with possible coal mining applications.

Another potential Sandia project involves recovery cf methane for fuel use from certain Eastern coal mines. If any of these feasibility studies appear promising Sandia hopes to participate in joint industry government projects in these areas.

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