COMBINED CO_2-O_2 UNDERGROUND PYROLYSIS-GASIFICATION FOR SOUTHWESTERN COALS

N. E. Vanderborgh, E. M. Wewerka, W. J. Trela, J. M. Williams, Jr. and G. R. B. Elliott

Abstract

Although vast quantities of subbituminous coals are located in the Southwestern U. S., severe technical problems exist in utilizing this resource. Much of these coals occur at depths where surface mining is not feasible. Even if this were not the case, a combination of limited water availability and environmental controls suggests that rapid expansion of coal utilization is not feasible. New technologies must be developed to exploit the known, vast coal resources in this region.

Details will be given on one such proposed process that involves a preliminary pyrolysis step followed by CO_2-O_2 gasification. This process is designed to yield both a hydrocarbon and a medium -Btu gas product stream. Arid, southwestern subbituminous coals appear well suited for an initial hydrocarbon removal step. Studies suggest that CO_2 is an adequate reagent to efficiently remove some 35% of the available carbon by reductive pyrolysis. The resulting semi-char has been shown to maintain adquate reactivity for eventual gasification.

Preliminary laboratory and engineering analyses for this underground coal conversion technology are described.

COMBINED CO_2-O_2 UNDERGROUND PYROLYSIS-GASIFICATION FOR SOUTHWESTERN COALS

Large enough coal resources have been identified in the Southwestern regions of the United States to permit planning for increases in coal utilization to meet the significant energy needs of the Western states. (1,2) This reemphasis upon coal is timely as proven reserves of both oil and gas are declining. (3) Yet, marked increases in coal utilization are limited by a variety of environmental and technological factors.

Only a small fraction, probably less than 5%, of the known coal resource can be extracted using surface mining technology. (4) Identified seams show dip, falling rapidly out of surface mining reach. Lenticular, thin and multiple seams of considerable depth suggest that underground mining may not be attractive.

Increased coal utilization using the remaining surface-minable coal may also be difficult to realize. (5) Opposition to increased strip mining is appearing. Stack cleaning of combustion gases from these high ash, low surfur materials has proven difficult. There remain serious questions about the advisability of increased combustion facilities. Any significant increase in coal utilization within the arid Southwestern region will also be limited by water availability. (Although limited brackish aquifers have been identified, it is not obvious that this water can be readily consumed for industrial processing.)

These factors, a combination of technological and environmental concerns, have already slowed a series of projects that were designed to increase coal utilization in the Southwestern United States. (5) It is becoming ever more obvious that the unique mixture of regional conditions leads to problems that are not readily addressed with existing technology. There is only a limited quantity of coal, using surface mining, that can be extracted and that coal is not easily used in combustion facilities. The concept presented in this paper is one approach that appears to have promise in expanding coal production in the Southwestern United States without moving into difficult and possibly restraining factors.

"Chemically mining" coal, underground conversion to gaseous or liquid fuels, is hardly a new idea (6). The thought of utilizing coal without the coincident societial and environmental costs of conventional mining has intrigued mankind for decades. (7,8) Partial underground combustion,"underground coal gasification," has been actively explored in the Soviet Union during the last fifty years (7). After some disappointing results in Alabama during the 50's, underground coal gasification programs are again active in the United States. (9) The Laramie Energy Research Center currently is demonstrating underground coal gasification in thick, subbituminous coals in Wyoming. (10) Lawrence Livermore Laboratory is exploring a concept of oxygen-steam gasification, again in the coal fields of Wyoming. (11) The Morgantown Energy Research Center has begun studies exploring gasification in Eastern bituminous coals. (12) Initial commercial tests are being evaluated using lignite beds in Texas. Concurrently, tests are also planned in Canada and in Belgium. However, none of these other experimental programs are turned to address the particular technical problems that now exist in the Southwestern region of the United States.

TWO STAGE PYROLYSIS-GASIFICATION

The underground coal extraction process proposed here is shown schematically in Figure 1. On the lower left-hand-side, gasification occurs in a previously treated, underground coal seam. This reaction, fed by an oxygen rich feed and moderated with CO_2, produces a continuous supply of a low-Btu gas. Carbon dioxide replaces the more conventional steam injection. This gas stream exits from the underground generator at elevated temperatures. Anticipated levels of sulfur, nitrogen and particulate matter in this gas dictate that gas cleaning is essential prior to utilization. This process is more easily accomplished at lower temperatures. Consequently, the gas is first fed through a heat exchanger and then into a gas cleaning operation. In the cleaning process, the gas is also stripped of CO_2 and sulfur leaving a combustible gas for utilization. In order to maintain reasonable sizes of the cleaning equipment, the gas is first pressurized. Exiting gas steams are also at pressure and might be transported some distance prior to final usage. Utilization of this cleaned product gas should cause no more environmental degradation than methane combustion.

Carbon dioxide, extracted during the gas cleaning process, is heated using the sensible heat from the gasification process and is then used for two purposes. Part of the gas is fed back into the gasifier for regenerative control. The remainder is fed into another, adjacent coal seam. This process, a hot gas preconditioning step, dries and partially pryolyses the virgin coal leaving a seam of open and uniform porosity with controlled reactivity. Pyrolysis products, moisture, liquid and gaseous hydrocarbons, are brought to the surface, collected, and shipped to a hydrocarbon processing plant. Following adequate reductive pyrolysis (with hot CO_2), the gas stream is changed to a mixture containing oxygen and the hot coal bed is gasified. Another, adjacent seam, following manifolding and seam opening, is then used for the site of the next pyrolysis section.

The overall process is designed to initially convert coal, using hot gas flows, into a stream of liquid hydrocarbons that could partially suppliment existing sources of petroleum feed stocks. During this process the mass transfer properties and the chemical reactivities of the coal seam are modified leaving a highly porous bed for subsequent gasification. This type of process is designed for the high volatile content, subbituminous coals found in the Southwestern United States.

DRYING AND PYROLYSIS OF SOUTHWESTERN COALS

Coals located in the Southwest are typically of subbituminous rank. (4) However, unlike the majority of such low-rank coals, these seams contain only a modest amount of water. Ten per cent moisture, is perhaps an average value although analyses of core segments can show values near 3%. (13) Previous underground coal gasification shows that water plays a key role in underground processing for this compounds acts as a major chemical reactant that can easily degrade gas quality. (14) Water is also an important heat transfer agent. Equally important, however, is the fact that water is a key factor in seam permeability. Consequently, moisture control underground must be a major consideration in any underground gasification process.

Permeability of underground coal seams is a difficult parameter to accurately determine. It appears that many of these subbituminous coals naturally show low permeabilities in the range of 0.1 mD. Removing moisture from these coals enchances the permeability by at least three orders of magnitude. (15) This behavior is readily understandable if one considers coal a solid, perhaps a hardened gel, with various-sized interconnecting pores and capillaries. We assume that the majority of these pores are less than 50 nm ($5x\ 10^{-8}$ cm) (16) and that these pores give coal a molecular-sieve property. Certain molecules appear to be able to penetrate the coal structure while other, not necessarily larger ones, are excluded. Temperature plays an important role in gas transport within pore structures of this type.

Moisture in pore structures of these coals effectively fills pores, due to the tetrahedral bonding capabilities of water, in a three-dimensional manner efficiently closing the material to mass transport. (17) (These low rank coals, due to their high heteroatom content, typically show hydrophillic behavior.) Removing the moisture effectively requires an agent that opens pore structure. Carbon dioxide is effective in doing this for like water, CO_2 also firmly adsorbs onto coal surfaces but unlike water, CO_2 is not capable of filling pore interiors. Rather one can assume that once a monolayer of this gas has adsorbed, the interior of the pore remains open. Consequently, the first important reason for moisture removal is to gain enchanced permeability both to move liquids and to cut down on pumping work requirements during the gasification process.

Moisture also degrades the gasification process. First steam formation lowers process temperatures increasing CO_2 production. (14) Secondly, the reaction of carbon monoxide with moisture is not advantageous for then CO is converted into a mixture of gases (H_2 and CO_2) with approximately the same total heat content but twice the volume. Lastly, moisture in the reduction zone should cool that zone, decreasing the effective residence time for CO production. All of these reasons suggest that one will be far ahead if water is first removed from the gasification process prior to CO generation. Such water removal seems feasible in Southwestern subbituminous coals by hot gas treatment.

Hot gas drying-pyrolysis requires the transport of significant quantities of gases - first calculations suggest that approximately one liter of hot gas is needed to pyrolyze one gram of coal. Moreover, that 1 gram of coal (volume approximately 0.65 cm^3) is converted to gaseous products with a volume, at STP, of 250 cm^3. Moving these quantities requires that the seam have reasonable flow characteristics. It seems unlikely that virgin coal will show high enough permeability to readily move sufficiently large quantities of gases. Consequently it may be necessary to develop seam opening techniques such as long range explosively driven penetrators, electrolinking, directed chemical leaching, etc. Evidence suggests that initial opening is feasible. (18) Moreover, carbon dioxide appears especially suitable for additional seam opening. This gas exhibits a high thermal conductivity and a low gas viscosity suggesting that for a primary heat transfer agent, CO_2 can be delivered with minimum pumping costs. Once a segment of the seam has been pyrolyzed to enhance flow parameters, then additional flows can be maintained.

However, even if enchanced permeabilities can be obtained, seam heating using hot gas flows cannot be done rapidly without large pumping costs, large quantities of heat and CO_2. Since gasification is a slow process, and the pyrolysis step is tied to that gasification, then pyrolysis must also be done slowly. One can assume that the heating process may take several months to accomplish. Should coal not be an efficient insulator, heat losses would prove prohibitive. However, unlike convective heat transport which will be promoted during the drying process, conductive heat transport is inefficient in coal. Thermal conductivities near 0.1 W/MK are well known. (19) In the absence of convection, heated coal will remain at high temperatures for long periods of time. Such data is shown in Figure 2. These data show the thermal waves measured at three different distances (five, ten and 20 feet) from a 950° F wall as a function of time. Heat, under these conditions, will be contained for years in a coal volume and will remain there until convective processes extract it. This heat will be available to increase process temperatures during the gasification step. Since necessary temperatures need be near 1000° C, this is an important energy contribution.

GASIFICATION OF DRIED SOUTHWESTERN COALS

Following the pyrolysis step, some 35% of the initial mass of the coal (as received) might be removed in a hydrogen-rich fraction. Laboratory studies suggest that the remaining semi-char will exhibit low and interconnected permeability. (13) Other studies show that the reactivity of this hydrogen-depleted material with oxygen is decreased somewhat from that over the virgin material; however this decrease (reaction rates are slower by less than a factor of two) should still leave sufficient reactivity for the gasification process. (20)

Gasification on a hydrogen-depleted char leads, primarily, to a stream of CO. The utilization of this gas presents many possibilities especially so since utilization can be in a controlled industrial atmosphere with little concurrent health hazards. There seem to be no reason to convert this product gas to other materials, e.g., methane, methanol, etc., although certainly these options exist.

ENGINEERING ANALYSIS OF THE COMBINED PYROLYSIS-GASIFICATION PROCESS

Initial engineering analyses have been completed on this underground coal gasification process. This combined-cycle process is somewhat similar to other above-ground facilities. For instance, there is good similarity between this underground process and a low-Btu gas generator/gas cleanup/ electricity generator system. However, in the present case, the majority of the processes occur underground. Underground processing offers some distinct advantages-residence times can be extended without changing costs - as well as some distinct advantages - pumping work can well be excessive and control can be difficult - over conventional coal processing facilities.

The results for these studies are based on the engineering flow diagram shown in Figure 3. This figure shows two separate, interconnected processes, the gasification path, heat removal, pressurization, gas cleanup and then utilization. (The cost projections are scaled to supply a 1,000 MW_e plant.) Waste heat and carbon dioxide are stripped from the gasification process to run the lower cycle, the hot gas pyrolysis step.

Projected annual consumption and production figures are listed in Table 1 (Again, these are projected for a 1,000 MW_e plant;hydrocarbon projections are taken from laboratory pyrolysis data.). One can see that such an operation would consume some 4 x 10^6 tons of coal annually and produce electricity, low molecular weight hydrocarbons, higher molecular weight hydrocarbons (pyrolyzed liquids), sulfur and pressure-volume work. (This latter results from pressurization of the output stream. The work equivalent contained in this gas volume is 50 MW. Most probably this work would be expended in operating plant utilities.)

There are two different resource recovery aspects to consider. The first is the fraction of the total coal contained in the seam that is recovered; the second is the cost of recovering a particular quantity of energy contained in a segment of the coal. Obviously, both of these recovery considerations are important. It is unrealistic to think that underground coal processing will ever show recovery efficiencies that approach 100%. However, these data do suggest that underground pyrolysis-gasification can lead to a favorable financial and energy return.

Resource recovery considerations are interrelated to a wide variety of different topics. For instance, subsidence in previous underground coal gasification has been a major problem. Pipes sever, ruining expensive manifolding installations. Cracks open, allowing excessive gas escapages. Above ground buildings tilt, causing structural damage. It appears that the optimum system for underground coal pyrolysis-gasification may well leave enough coal underground to minimize subsidence. (This would not be unlike current room-and-pillar techniques of underground mining, a technique used for exactly the same reason.) These and other site specific questions need to be answered before one can accurately define the economics of resource recovery. What is now clear, however, is that without careful control of the underground process, the optimization of resource recovery may not be possible.

Another important consideration is the cost of the produced fuel gas. Preliminary data are shown for this projection in Table 2. Costs projections were taken from similar studies estimating costs for above ground surface gasification to produce a low-Btu gas. (21) The operating and maintenance cost charge nothing for the cost of the fuel; $12.3 million is the anticipated annual field development charge. Bottom line projections must be altered to include taxes-royalty on the coal consumed. These projections show that each $1.00 ton or royalty increases the cost of the final product by about 10¢/10^6 BTU. Thus a $3.50/ton of coal consumed increases the final fuel costs to $1.56/$10^6$ Btu. This projected favorable cost should remain stable over the lifetime of the plant.

The data lend to several interesting conclusions. Foremost of these is that the field development charges are not an important factor. (These projections were calculated for a 7.6 m (25 foot) seam located 152 m (500 ft.) below the surface using 7.6 m (25 ft.) well spacing. Charges for increased field development are, however, most influenced by changes in the pumping work. Large amounts of energy are expended in moving gases underground. Costs are reflected in the high annual capital charges. Increases in underground permeability gained, for instance, in decreasing well spacing may be a wise investment. Second, the difficult technology is, to a large extent, below ground. Gas handling and cleaning systems, if one accepts the eventuality of low temperature gas cleaning, are all off-the-shelf systems. This is also true for electricity generating systems - conventional gas turbines can be incorporated without concern for high temperature corrosion and abrasion. These data also suggest that gas leakage, concurrent subsidence, and water influx, factors that can uncontrollably change the underground chemistry and rheology are the things that really most influence the economics of the process. Therefore, the program at Los Alamos Scientific Laboratory has been designed to carefully investigate these several aspects of the underground process.

TECHNOLOGY DEVELOPMENT PROGRAM

Studies are now exploring the processes of concurrent heat and mass transfer through Southwestern subbituminous coals. Central to these initial studies is the problem of moisture removal and permeability modification by hot gas treatment. Laboratory data will be obtained on representative coal blocks to identify necessary kinetic parameters of concurrent heat and mass transfer. These data are essential for mathematical modeling of these underground processes. These modeling activities will predict flow profiles and lead to suggest manifolding techniques.

Studies also will explore the interrelated problems of resource recovery and environmental impact. An appreciation now exists that environmental degradation due to gas leakages, to various subsidences and to groundwater contamination is interrelated with resource recovery. The economic trade-offs between a "contained" underground operation and recovery efficiency are also under study.

These laboratory and analytical studies will serve as input into planning for a series of controlled field tests using Southwestern subbituminous coals. These tests will operate on a segment of coal somewhat close to the surface, perhaps near 15 m (50 feet) deep. First a defined region of the coal will be isolated from the rest of the seam. One feasible way to perform this isolation is to construct a concrete-pier wall from the surface, through the seam and part way into the underburden. A representation of a section of this containment wall is shown in Figure 4. Concrete piles will be laid in a rectangular pattern defining a coal section 15 m x 15 m (50 ft x 50 ft). Feed and control pipes will then be inserted into the coal. Important here are pipes placed behind the wall, "water cooling" pipes, to maintain the integrity of the wall and assure that the fire can't move out of the contained section. Undoubtedly, some sort of subsidence control will be essential.

Although this experimental arrangement has some similarity to laboratory block tests, several important features suggest that this approach is necessary to learn which really takes place underground. One needs to work with virgin coal, with underground moisture and gas content intact. Yet one needs to be assured of defined mass balances. The thrust behind these studies is to obtain definitive answers about flows, chemical and heat balances and resource recovery. Two separate tests are planned. The first of these will study the drying and pryolysis of a coal section. Following seam opening, directed flows of hot gases will pass through a coal section. Pyrolysis will be carried on for a fixed period and then quenched. Detailed postmortem analyses will be made on the block. A second test will again study the drying and pyrolysis of a second block; however, this test will be carried through a gasification stage prior to the final post mortem. These field tests will provide data for subsequent planning and large-scale commercialization.

ACKNOWLEDGEMENT

The authors wish to thank F. P. Schilling, G. A. Baca, J. W. Barnes and G. E. Cort for their helpful discussions and assistance.

REFERENCES

(1) F. E. Kottlowski, E. C. Beaumont and J. W. Shomaker, "New Mexico" in 1975 Keystone Coal Industry Manual, McGraw-Hill, Inc., New York, p. 585, 1975.

(2) C. D. Kolstad, "The 1975 Energy Production System in the States of the Rocky Mountain Region", Los Alamos Scientific Laboratory, LA-6624, December 1976.

(3) E. C. Arnold, et al., "New Mexico's Energy Resources '75", Bulletin 107, New Mexico Bulletin 107, New Mexico Bureau of Mines and Mineral Resources, Socorro, NM 1976.

(4) anon, "Outlook for Coal: Bright, but with Problems", Chem. Eng. News., 55, Feb. 14, 1977.

(5) J. W. Shomaker, et al., "Strippable Low-Sulfur Coal Resources of the San Juan Basin in New Mexico and Colorado," Memoir 25, New Mexico Institute of Mining and Technology, State Bureau of Mines and Mineral Resources, Socorro, NM, 1971.

(6) G. R. B. Elliott, "Chemically Mining Coal," Mining Engineering, Sept., 1973.

(7) J. L. Elder, "Underground Gasification of Coal", in Chemistry of Coal Utilization, H. H. Lowry, Editor, John Wilen and Sons, Inc., New York 1963, pp. 1023-1041.

(8) National Coal Board, The Underground Gasification of Coal, Sir Isaac Pitman and Sons, Ltd. 1964.

(9) R. Raimondi, P. L. Terwiliger, and L. A. Wilson, Jr., "A Field Test of Underground Combustion of Coal," J. Petroleum Technol., Jan. 1975, pp. 35-41.

(10) G. C. Cambell, C. F. Brandenburg and R. M. Boyd, "Preliminary Evaluation of Underground Coal Gasification at Hanna, Wyo., Bureau of Mines, Laramie, Wyo. BM-TPR-82, Oct., 1974.

(11) G. H. Higgins, "A New Concept for in-situ Coal Gasification," Lawrence Livermore Laboratory, Rept UCRL-51217, Rev. 1, Sept. 27, 1972.

(12) W. K. Overbey, Jr., C. A. Komar and J. Pasini, III, "Underground Combustion of Coal: Proposed Concept for Direction Control of the Combustion Zone," paper presented at the Annual Meeting of the Geological Society of America, Dallas, TX, Nov. 11-14, 1975.

(13) H. E. Nuttall, W. G. Stoddart and W. J. Chen, "Reactivity and Kinetics of Sub-bituminous New Mexico Coal," 51th Annual Conference of S.P.E., New Orleans, Oct. 3-6, 1976. Paper No. SPE 6154.

(14) N. Z. Brushtein and V. S. Za'grebalnaya, "The Influence of Moisture on the Process of Underground Gasification," Podzemmaya Gasifikatsiya Uglei, 3, 33 (1957).

(15) R. N. Pitkin, I. V. Eremin and K. I. Cheredkova, "Permeability of Yuzhno-Abinsk Coal From Kuzbass", Trudy Inst. Goryuckikh Iskopaemukh, Akad. Nauk S.S.S.R., Otdel. Tekh. Nauk, 7, 85 (1957).

(16) J. Thomas, Jr., H. H. Damberger, "Internal Surface Area, Moisture Content, and Porosity of Illinois Coals: Variations with Coal Rank," Illinois State Geological Survey, Circular 493, 1976.

(17) P. Cinche, H. Marsh and S. Pregermain, "Absorption of Carbon Dioxide, Methanol and Water Vapor on Cokes-Determination of Micropore Volume," Fuel, 46, 341, 1967.

(18) W. J. Carter, Los Alamos Scientific Laboratory, Los Alamos, NM, private communication.

(19) I. L. Faberov, E. S. Avdonina, and N. P. Turevskaya, "Effects of Preheating on the Thermal Conductivity of Blocks of Moscow Coal and of Oil Shale," Trudy. Inst. Goryuchikh, Akad. Nauk S.S.S.R., Otdel Tekh. Nauk., 7, 94 (1951).

(20) N. E. Vanderborgh, Los Alamos Scientific Laboratory, unpublished results.

(21) Second Supplement to the Application of El Paso Natural Gas Co. for a Certificate of Public Convenience and Necessity, FPC Docket No. CP 73-131, October 8, 1973.

TABLE 1: RESOURCE FLOW AND PRODUCTION PROJECTIONS FOR TWO-STAGE PYROLYSIS GASIFICATION UNDERGROUND COAL UTILIZATION FACILITY

A. INPUT STREAMS (ANNUAL RATES)

Coal Consumed . . . . 3.51×10^6 tons[a]

Air Consumed . . . .8.03×10^9 scf

Water Consumed . . . . .negligible net input

B. OUTPUT STREAMS (ANNUAL QUANTITIES)

Electricity	8.77×10^9 KWh	\$0.03/KWh	\$ 2.63×10^8
Low MW HC's[b]	1.08×10^{13} Btu	\$1.50/10^6Btu	\$ 1.62×10^7
High MW HC's[c]	1.26×10^6 Bar.	\$ 10/barrel	\$ 1.26×10^7
Raw Sulfur	9.70×10^7 lbs.	\$ 50/ton	\$ 2.43×10^6
PV Work	4.38×10^8 KWh	\$0.03/KWh	\$ 1.31×10^7
		Annual Value	\$ 3.07×10^8
		Per Ton Coal	\$88.55

[a] Assuming a 25' coal seam, annual area of seam addressed is (20/f acres) where f = fraction recovered, i.e., if half of the coal is consumed, then coal under 40 (20/0.5) acres would be consumed.

[b] Low molecular weight hydrocarbons, mainly C_1 - C_4 hydrocarbons, gases

[c] High molecular weight hydrocarbons, mainly C_5 - C_9 hydrocarbons, liquids

Other potential products, especially ammonia, are not listed.

TABLE 2: LASL TWO STAGE CO_2-O_2 UNDERGROUND COAL EXTRACTION-ECONOMIC PROJECTIONS

	ANNUAL COST (K$)	BASIS
OPERATION AND MAINTENANCE		
Coal Feedstock	0	
Field Development[a]	12,300	25 ft. well spacing
Plant Operation	10,440	
Administration	5,760	
Misc. Taxes	4,160	1.1% of plant investment
	$32,680	
DEPRECIATION	17,860	4.7% of plant investment
CAPITAL CHARGES	57,000	15% of plant investment
BYPRODUCT REVENUES	(11,000)	estimated market value
TOTAL ESTIMATED COSTS	$96,540	
ESTIMATED COSTS PER 10^6 BTU	$1.21	
ESTIMATED COSTS INCLUDING ROYALTY OF $3.50/ton coal	$1.56	

Costs calculated assuming mid 1975 completion of construction.

Costs do not include capital or operating costs for electricity generation or hydrocarbon separation facilities. Potential revenues from hydrocarbons are likewise neglected.

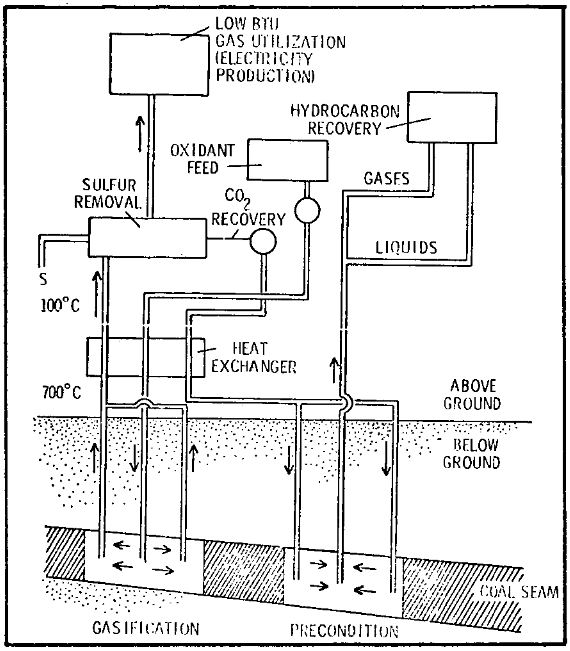

Figure 1: Two Stage Underground Gasification and Pyrolysis Facility For Utilization of Deeply Lying Southwestern Subbituminous Coals.

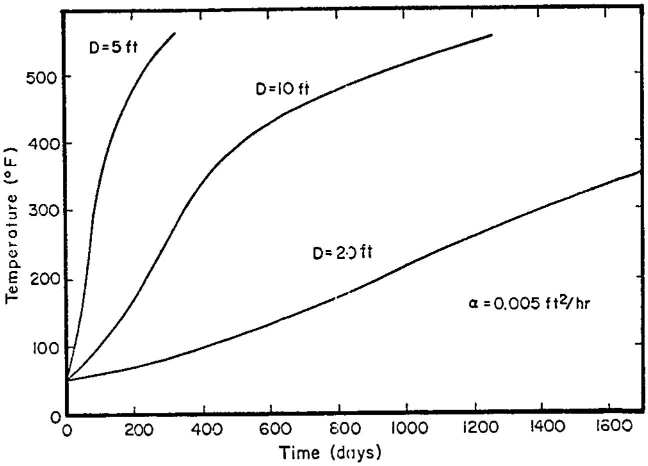

Figure 2: Thermal Waves Resulting from 950°F Wall 5, 10 and 20 feet In Coal with α = 0.005 ft^2/hr. Conduction Only.

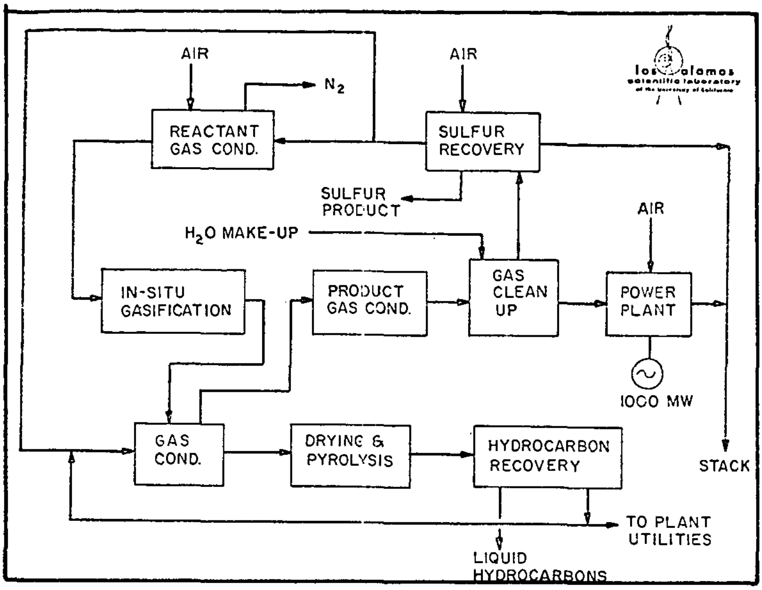

Figure 3: Flow Diagram of Above Ground and Below Ground Processing Facilities for Two-Stage CO_2-O_2 Process. (All process steps except in in-situ gasification and drying and pyrolysis are above ground)

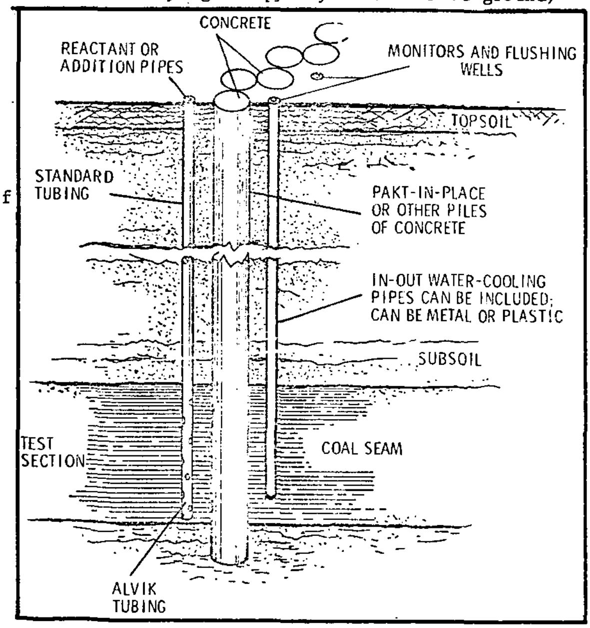

Figure 4:

Cross Section of Mass Control Wall for Underground Experiments