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UNDERGROUND THERMAL GENERATION OF HYDROCARBONS FROM DRY, SOUTHWESTERN COALS

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ABSTRACT

The LASL underground coal conversion concept produces intermediate-BTU fuel gas for nearby industries such as "minemouth" electric power plants, plus major byproducts in the form of liquid and gaseous hydrocarbons for feedstocks to chemical plants e.g., substitute natural gas (SNG) producers. The concept involves controlling the water influx and drying the coal, generating hydrocarbons, by pyrolysis and finally gasifying the residual char with O_2/CO_2 or air/CO_2 mixtures to produce industrial fuel gases. Underground conversion can be frustrated by uncontrolled water in the coal bed. Moisture can (a) prevent combustion, (b) preclude fuel gas formation by lowering reaction zone temperatures and creating kinetic problems, (c) ruin product gas quality by dropping temperatures into a thermodynamically unsatisfactory regime, (d) degrade an initially satisfactory fuel gas by consuming carbon monoxide, (e) waste large amounts of heat, and (f) isolate reaction zones so that the processing will bypass blocks of coal. Large amounts of electric power (or other) production along with vast supplies of hydrocarbon fuels are produced by this process.

OIL AND GAS FIELDS IN AMERICA ARE consumed twice as fast as they are discovered. This fact well describes the Nation's energy situation. Although alternative energy sources including solar and nuclear will make contributions to the national energy mix, neither can supply those hydrocarbons needed for transportation, space heating, and chemical synthesis. Instead, the Nation's vast coal resources must be directed to fill this need.

Difficulties restrict greatly increased coal production using current technology. Specifically, capital, labor, and environmental limitations exist throughout the coal fuel cycle. Underground coal processing, i.e., the utilization of coal in place to produce clean fuels, could surmount these limits.

One such underground coal conversion process (schematically presented in Fig. 1) is being evaluated by the Los Alamos Scientific Laboratory (LASL) (1). Coal, 250 to 1,000 feet underground, is first heated with hot gas injection supplied through manifolding emplaced in the seam. The heating produces moisture and hydrocarbons leaving in place a hot, porous semi-char. This char is gasified while still hot by a heat CO_2/O_2 gas stream, thereby producing a fuel gas containing little nitrogen and moisture impurities and mostly carbon monoxide.

This fuel gas is cooled to allow cleaning with proven technology to remove sulfur compounds and to recover carbon dioxide for use in pyrolysis and control. The fuel gas now is in the intermediate-Btu range of around 275 Btu/scf.

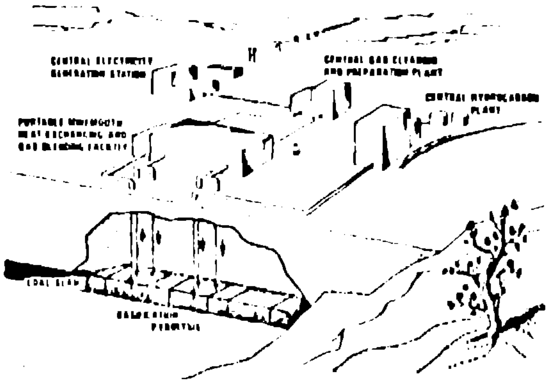

Fig. 1-LASL concept in underground coal conversion.

This gas cooling just mentioned decouples heat production and electricity generation from the "minemouth" operations. This decoupling permits good control of the fuel-gas quality both through the gas cleanup and through blending the CO fuel with a higher-Btu fuel gas from the pyrolysis system to deliver a constant Btu-content product. Sensible heat which is stripped from the hot producer gas is used to drive the first-stage pyrolysis in another section of the coal.

Two separate classes of products are generated by this type of coal conversion system: First, hydrocarbon gases and liquids are produced for use elsewhere, and, second, a highly uniform gas product of intermediate-Btu content is produced for more local consumption, e.g., at an electric power station essentially at the mine mouth.

EXISTING PROCEDURES FOR UNDERGROUND COAL CONVERSION

Existing schemes for underground coal conversion typically utilize drilling, linkage of vertical wells by reverse combustion or deviated drilling, and then extraction by forward combustion. (2) During forward combustion the coal is dried, pyrolyzed, and gasified at similar times but at different places in the processing front. (3) This behavior is shown in Fig. 2. Here a directed flow channel labeled "char channel" links two vertical wells. Oxidative fuel gases (air- or oxygen-containing mixtures) are pumped into the left well, and they

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*Numbers in parentheses designate References at end of paper

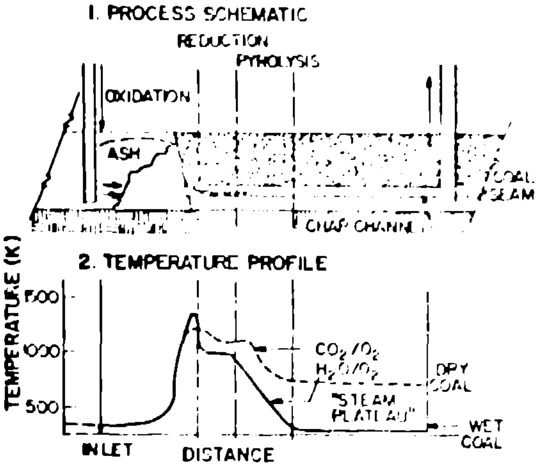


Fig. 2-Temperature profiles along the centerline of the reaction path for a coal seam undergoing underground conversion.

react with the coal seam to produce a low- or intermediate-Btu gas which is delivered from the right well.

Many workers have noted that the process can be divided into several regions. (4) Initially, in the oxygen-rich zone, oxidative processes form CO_2 and H_2O. Next, these hot gases transfer into the reductive region where worthwhile fuel gases, H_2 and CO, are produced. The fuel gases then lose heat to coal, generating pyrolysis products. Finally, the pyrolysis gas/H_2/CO mixture vaporizes moisture.

(The preceding steps are idealized, of course. The successful formation of major fractions of carbon monoxide depends strongly on the temperature. CO_2 and H_2 will not react with solid carbon to produce useful fractions of CO and H_2 unless the temperature is at least 1000K. Furthermore, undesirable side reactions can occur. These matters are discussed in this paper.)

Two probable kinds of centerline temperature profiles through the reactive zone are shown in the lower part of Fig. 2. Inlet temperature are controlled by residual ash temperatures and gas feed temperatures. Gases entering the reaction zone reach temperatures in excess of 1300K at the oxidation zone maximum. Endothermic reductive processes are driven by heat from the oxidative section. Finally, the product gases interact with the unreacted coal seam. If the coal seam contains liquid water, the products are generated at some "steam plateau" temperature, the temperature of pressurized equilibrium steam and water controlled by the pressure of the outlet gas stream.

The two profiles show temperatures for reactant feeds O_2/H_2O and O_2/CO_2. (We assume that the necessity to minimize the work of gas compression will preclude the utilization of air. Cleaning large quantities of nitrogen, prior to combustion, is prohibitively expensive.) The carbon dioxide-oxygen temperature is different for two reasons. First, the higher thermal diffusivity of carbon dioxide spreads peak temperatures in the energetic oxidation region. Second, due to the slower CO_2-char kinetics than H_2O-char kinetics, reactions with CO_2 require longer residence time. However, kinetic differences do not degrade yields because high temperatures are maintained for longer distances if the reactions are slowed. Endothermic reductive reactions occur with CO_2 and H_2O in either case. With carbon dioxide the reductive zone is extended. Such an extension clearly is not detrimental. Actually it may be beneficial through increased thermal-stress cracking in the underground region.

WATER IN UNDERGROUND COAL CONVERSION

Many seams, not only those of lower rank, are productive aquifers. We suspect that little of this high-volume transport occurs in distributed porous flow. Rather, flow occurs in fracture systems. Additional coal porosity is known to exist as either macro-pores (mercury porosimetry) or micro-pores (helium porosimetry). Generally it is thought that smaller pore structures have molecular-sized dimensions, 10^{-9} m. (5)

Energy of Water Removal: It has been well documented that underground coal gasification in seams with high moisture content is technically difficult. Part of the problem becomes apparent from data in Table 1. As seam moisture increases, appreciable fractions of the total energy content of the coal, here 10^4 Btu/lb on combustion, must be expended for moisture evaporation and for heating the vapor to desired process temperatures. Clearly, a moisture content (or a rate of moisture influx) will eventually be reached for which underground conversion is impossible.

Table 1 - Heat content and energy consumption for water removal, per cubic meter of coal.

H_2O %	Mass gm $\times10^{-6}$	Total Heat Btu[a] $\times10^{-6}$	Water Heat Btu[b] $\times10^{-6}$	Energy Cost %
5	1.50	31.5	0.26	0.83
10	1.50	29.8	0.52	1.74
20	1.40	24.7	0.97	3.93
30	1.35	20.9	1.40	6.72
40	1.35	17.6	1.87	10.50
50	1.30	14.3	2.25	15.70
60	1.25	11.0	2.60	23.60

[a] Calculated assuming coal has a heat content of 10^4 Btu/lb (dry basis).
[b] Calculated assuming seam is originally at 5°C and moisture is heated to 500°C.

The Gasification Reaction: Fig. 3 indicates the thermodynamic maximum Btu per scf which can be attained at various temperatures for different burn conditions. The upper curves are for the equilibrium heating values for pure H_2O or CO_2 reacting with solid carbon. Note that the heating values drop off sharply in the range 800 to 500°C. The fuel gases are hardly worth producing if temperatures are not above 600°C.

For air-burned materials (i.e., $H_2O + N_2$ and $CO_2 + N_2$) the curve is similar, but heating values

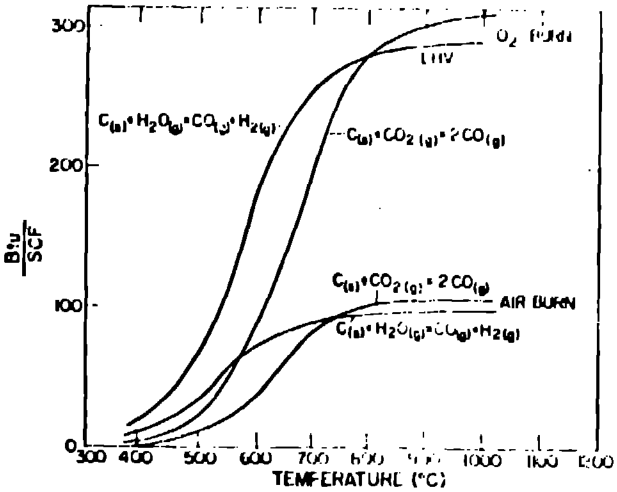


Fig. 3-Equilibrium heating values per scf for carbon/oxygen and carbon/oxygen/hydrogen mixtures.

per scf are lower because of the dilution by inert nitrogen.

The problem may be more complex than is indicated. While the calculations indicate the best fuel gases that can be produced, the kinetics are more unfavorable. Solid carbon must be in the neighborhood of 900°C before fast reactions are certain.

Thus, if water influx is too great, or if too much water is initially present in the coal, coal may never reach process temperatures suitable for underground conversion.

Degradation of the Product Through Reaction with Water: The water gas reaction between hot carbon and steam to produce a fuel gas

$$C_{(solid)} + H_2O_{(gas)} = CO_{(gas)} + H_{2(gas)} \qquad (1)$$

depends on the fact that the reaction

$$2CO_{(gas)} = C_{(solid)} + CO_{2(gas)} \qquad (2)$$

does not proceed. The similar water-gas shift reaction does take place readily. Here CO is destroyed by reaction with water vapor

$$CO_{(gas)} + H_2O_{(gas)} = CO_{2(gas)} + H_{2(gas)} \qquad (3)$$

While H_2 has the same approximate heat content, it is more difficult to remove CO_2 from H_2 than to remove H_2O from CO. Moreover, volumes are increased with no increase in the total heating value. Water vaporization (as was discussed earlier) takes up heat which is never usefully recovered.

Effects of Water on Coal Permeability: A dominant role of moisture on underground conversion appears in changing the permeability of coal.

The permeability through wet coal under simulated *in situ* conditions is near 0.01 md. Removal of stress on coal results in the formation of microfracture systems that leads to measured permeability values near 10 md. (6) Removal of moisture from such coals increases permeability to a similar extent. It is becoming ever more clear that moisture-stress-permeability effects are interrelated in these coal systems.

Moisture is strongly adsorbed within coal pores. Typically, especially with low rank coals, various hydrophillic groups occur on the surface of these channels. Due to the tetragonal nature of bonding in water molecules, coal pores are effectively filled by moisture, forming impermeable material.

CO_2 is adsorbed even more strongly than water onto coal surfaces. Furthermore, CO_2 is a linear molecule which, unlike tetragonal-bonding H_2O, cannot fill pores in the coal. Thus CO_2 creates a water-free and relatively open pore structure.

These CO_2 properties of selective adsorption, low viscosity and high heat capacity, make this gas the drying agent of choice.

Our experiments have clearly pointed out that the permeability increases produced upon drying are locally reversible. However, the original bulk patterns of flow throughout the coal will certainly not be reproduced. Instead, the patterns of gas flow and water deposition will locally alter the flow and create blockages in the paths, halting further gas flow. Areas will be effectively isolated from convective heat flow.

This redeposition of moisture has severe implications for underground processing. For instance, should moisture invade a processing zone, liquid water can condense into coal pores and isolate regions which ideally should be reacting to a heat and mass flow. This results in an instability that bypasses water-rich zones wasting coal.

Any water-rich region is subject to evaporative cooling and recondensation underground due to fluctuations in the temperature or pressure of gases moving past it. (6) At the site of steam condensation the deposition of water into the surface cracks will block off that specific region to reactive flow. As corollary, both the loss of heat and the loss of mass flow can result in bypassed coal as the coal gasification front moves through a region of the coal field: thus efficiency demands the removal of water during the coal processing reactions.

Steam injection, which has been proposed for underground gasification, (2) can actually introduce water and aggravate existing heat flow paths. We propose to inject O_2/CO_2 to circumvent this problem.

WALLING BY HYDRAULIC FRACTURE TECHNIQUES

Although our laboratory experiments show that coal-block drying is technically feasible, field success will require that almost all water invasion from neighboring zones is restricted. Underground processing in this way requires a dry seam, below 10% moisture. And moisture must be excluded. Southwestern hydrology identifies shallow coal seams as aquifers that connect surface runoff with deeply lying sandstone strata. Recharge is periodic and depends upon recent climatic conditions. It will be necessary to develop technology that will restrict moisture flow into processing regions. One proposed method for accomplishing this is shown in Fig. 4.

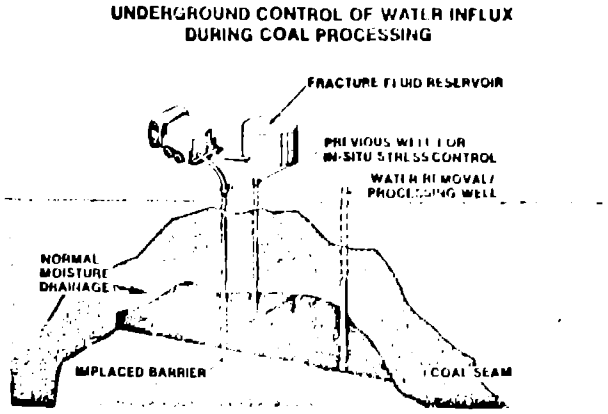


Fig. 4-Wall preparation through hydraulic fracture with water-impermeable fluids.

Hydraulic fracture would be utilized to construct underground barriers. Initially, stress modification techniques would be utilized to redirect *in situ* stresses in a way that hydraulic fracture would run perpendicular to seam dip. (Vertical fractures are assumed here because of the large overburden of deeply lying formations. Should the coal fracture horizontally, other techniques would be required).

Fracturing is done with a fluid which will polymerize (or set) after a requisite time. The crack then will be filled with material that impedes moisture intrusion into the processing zone. Other possibilities exist including increasing stress with special grouts to impede water flow. These ideas are, as of now, only conceptual. It is obvious that successful underground coal conversion technology will require ways to "fix" water invasion. Much work remains to be done in this area.

RESOURCE UTILIZATION PROCESS EFFICIENCY

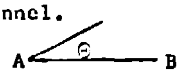
Underground coal conversion alters coal technology from a mining process to an operation more like petroleum production. And, operation economics are similar to oil-field economics. The main concern is pay-out, i.e., the product value compared to the cost of production. Although, from an environmental standpoint, one is concerned about "resource utilization efficiency", the technology economics do not depend upon high total recovery efficiency (such as the high efficiencies now obtainable with long-wall underground mining). Rather, there must be successful extraction of energy from a particular coal section. This is entirely analagous to oil production. Improved recovery is attempted using secondary and tertiary techniques usually well after primary production and associated economics are completed.

Drying underground coal permits divergence of the process path from the center-directed channel. Such spreading is critical to the economics of the operation. If we consider a typical situation, Table 2, this becomes apparent. Calculations show completely consumed coal tonnages using a two-spot pattern spaced 50 m apart in a 2-m thick seam. (Such spacing might be optimistic.) The "spreading angle" describes the angle which the processing-zone moves into the coal mass. This is the angle between that interface and the well-to-well axis. Should little spreading occur, say 5°, even with this geometry, only 361 tons of coal can be processed. It is hard to see how manifolding costs could be recovered here in anything but a fairly shallow seam. However, if the spreading angle can be increased to an extent that the process zone leaves the borehole at an 30° angle, then 2,400 tons of coal can be processed through the same manifold. This factor-of-7 increase appears to us to be the critical parameter in process economics. Regardless of the seam thickness, unless marked divergence both horizontally and vertically occurs, little energy can be recovered from any well arrangement. Moisture removal appears to be the big factor in obtaining large spreading angles.

Table 2 - Conversion of coal as a function of process spreading angle

Spreading Angle $^{\circ a}$	Total Volume^b m^3	Total Mass of Coal tons^c
5	218	361
10	441	727
20	910	1501
30	1443	2381

^{a}Spreading angle, degrees, between divergence of process zone and central borehole-to-borehole channel.



^{b}Vertical wells spaced 50 m apart in a 2-m thick seam.

^{c}Calculated assuming density of 1.5 gm/cm^3.

PYROLYSIS PRODUCT YIELDS

Coal pyrolysis has been extensively studied. (7) Interpretation of results is complicated by the fact that processes of cooling (evaporation or endothermic pyrolysis) compete with heat input. Heat transfer becomes dominated by convective processes and such processes are interrelated with heat transfer since product gases are produced as functions of temperature. The situation demands close attention to heating rates and particle size. One can readily speculate that convective mass transport will be influenced by *in situ* stress. Consequently, assuming that yields measured in the usual laboratory conditions reflect similar underground yields is questionable. Nevertheless, that assumption is a start.

Typical subbituminous pyrolysis gas data are shown in Table 3. These data were collected on powdered coal (30 mesh) in a cylindrical geometry. Data given represent 350°C values. From thermogravimetric data, we know that approximately 25% of the original coal mass (as received) is volatilized at this temperature.

A straightforward calculation leads to the fact that thermal processing produces 8.2 x 10^3 scf per ton of subbituminous coal. Assuming that this product has a heating value of 500 Btu/scf, this low molecular-weight gas is equivalent to 4.09 x 10^6 Btu/ton, and assuming that projected gas prices are reliable, this byproduct gas will contribute approximately $10/ton of coal consumed. A 1,000 MW_e power generating facility requires approximately 6 x 10^6 tons coal/year. Production of the power would, using the LASL scheme, cogenerate 2.4 x 10^{13} Btu/year of low molecular-weight gas. Such a gas could be readily upgraded to methane.

This annual production needs to be compared to proposed surface gasification processes designed to produce 250 MM scf/day. Annual production of that plant would be 9 x 10^{13} Btu. Thus, underground production scaled to 1000 MW_e amounts to approximately 25% of that of a large, surface gasification facility.

From a resource base, production of coal pyrolysis gas appears very promising. From each square mile of a 6-ft seam a total of 3.7 x 10^{13} Btu could be produced. Methane drainage (assuming 1.3 mg/g of coal) using state of the art fracture technology at ambient temperature could produce only 1.4% as much, 5 x 10^{11} Btu/mile^2. The San Juan Basin alone contains approximately 2 x 10^4 mile^2. Thus, some 500 quads of low molecular weight hydrocarbon gas could be produced from that basin alone by pyrolysis as a byproduct of coal gasification of just one of the several seams.

Table 3 - Composition of pyrolysis gases from Fruitland coal.

Component	Volume %	gm/mol of pyrolysis gas
H_2	22	0.44
CH_4	20	3.20
CO_2	21	9.34
CO	11	3.08
C_2's[a]	5	1.30
C_3's[b]	4	1.60
H_2O	17	3.06
		21.9 gm/mol

Products collected at 350°C following transfer through a tar-mist trap at 100°C. Heating rate approximately 3°C per minute.

[a] Assuming acetylene, C_2H_2.

[b] Assuming propyne, C_3H_4.

CHAR GASIFICATION WITH CO_2 MODERATION

The LASL concept ends with a CO_2/O_2 gasification of char produced by initial hot gas pyrolysis. During the pyrolysis reaction a hydrogen enriched fraction (see Table 3) is stripped from the coal leaving a carbon rich residue, char. It is true that all coal is deficient in hydrogen and that liquefication of coal requires an outside hydrogen source. However, processing in this way segregates the hydrogen, removing about 50% of the hydrogen (not considering hydrogen in H_2O vapor production) while removing about 25% of the carbon. Consequently, the pyrolysis products are hydrogen enriched and can serve as a petrochemical source with no additional hydrogen source.

The remaining char must be processed with good efficiency. The intriguing possibility in this concept is that, because of this staged production, better yields will be realized during gasification. Consequently, not only is the hydrocarbon production "free" but one can predict (a) that the total energy recovery is higher due to this staged approach, and (b) that more uniform products will result.

Because moisture and volatiles have been removed from the char, char combustion will be different from that of unaltered coal. The combustion temperature will be higher and the burn will be more uniform. Less heat input is required for the already hot and dry char. CO_2, CO, and H_2O at perhaps 1000 C will replace hydrocarbon pyrolysis products as the flowing heat transfer agents. Due to the lack of pyrolysis products and the removal of the "flame hopping" combustion spread mechanism, radiation will be the significant heat transfer step during char gasification. Cooling effects of drying coal will be avoided and because of the high char temperatures, a higher CO/CO_2 ratio will be obtained.

Since steam formation in this previously dried process zone does not lower temperatures of the product gases, approximately 25% of the total remaining energy in the char is delivered as sensible heat. This fraction is removed and utilized for drying and pyrolysis of another, nearby coal section.

Expected product composition from this step is listed in Table 4. The other column is field results from native coal, gasified with air injection. There is a considerable volume reduction using O_2/CO_2 on char. This is an important consideration.

Table 4 - Gasification product stream composition.

	Volume Per Cent	
Component	Air-UCG[a]	O_2/CO_2-Staged UCG[b]
N_2	38.1	0
H_2O	30.4	5.0
CO_2	11.0	27.0
CO	6.1	54.0
H_2	11.9	13.0
Tar	1.4	0.5
CH_4	1.1	0.5
	100.0	100.0
Scf/10^3	13.6	4.26

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a. Field Experience, taken from (8)

b. Calculated values taken from conservative estimates of laboratory char gasification data. (9)

Economic assessments clearly point to the process penalties for moving high volume of gases. Note that air-injection results in a process stream composed of 68% nitrogen and water. Moving these inert gases (and disposing of the water) is highly energetic. Likewise, all of these gases must be thoroughly cleaned prior to combustion. Obviously, cleaning a minimum quantity of gas is necessary.

SULFUR REMOVAL BY PYROLYSIS

As mentioned previously, hydrogen is segregated during pyrolysis, removing a hydrogen-enriched product stream. Thermal treatment is known to preferentially remove organic sulfur from coal as well. The end result is that decreased gas cleaning may be required following gasification. The economic consequences of this are unclear. In the extreme, air could replace oxygen for gasification. For, if product gases are devoid of sulfur compounds, gas cleaning might be unnecessary. And if product gas need not be cleaned, then there are sound reasons to utilize air injection on the dry, hot char. This possibility also needs evaluation.

COMBINED HEAT AND MASS TRANSFER IN LABORATORY COAL EXPERIMENTS

Experiments underway correlate laboratory measurements of heat transfer during heat injection studies with a finite element, combined heat-and-mass transfer computer code. Coal blocks are encased in cements to control mass transfer. A single hole is then drilled through the block and hot gases are used to inject heat through that passage. (This simulates the underground situation, Fig. 2). Details of these studies have been presented previously. (1, 6)

Typical data from these studies are given in Fig. 5. The coal block was initially heated within a precise thermostat to 100°C, the "oven temp". During this heating, a limited quantity of moisture was exhausted from the central passage. The introduction of hot CO_2 (injected at a temperature of 139°C and a flow of 400 cm^3/min) resulted in higher inlet side temperatures and lower outlet side temperatures (Fig. 5). CO_2 caused water evaporation, lowering the temperatures in the interior of the block below 100°C for considerable periods of time. Finally, after some 60% of the "moisture content" of the coal was removed, temperatures in specific regions of the block increased in a way that indicated open permeability for convective heat transport. These data can be modeled with considerable success if we assume that moisture removal changes permeability of discrete regions within the block.

COMBINED UNDERGROUND PYROLYSIS GASIFICATION

Engineering analysis of thermal generation of hydrocarbons from coals has been completed. As the flow chart in Fig. 6 shows, the greatest majority of processing occurs above ground. Of the many unit operations, only two, coal gasification and underground coal pyrolysis, occur below the surface. Yet these two dominate the entire system. Economics demands that the energy content of production wells be dense. If this is not so, underground coal gasification can involve prohibitive pumping costs, therefore an unworkable technology. It is essential that the underground section is well understood and that an open, permeable bed is prepared. Thermal treatment of subbituminous coals is an attractive possibility to accomplish this.

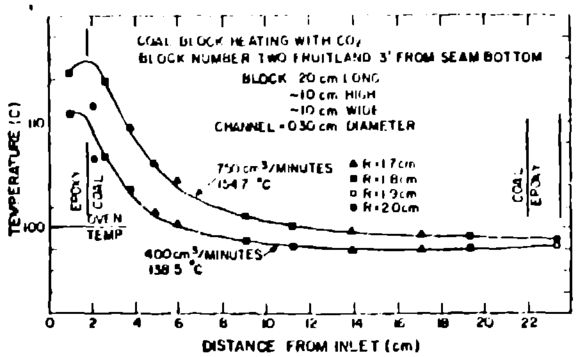

Fig. 5-Coal-block heating with hot CO_2

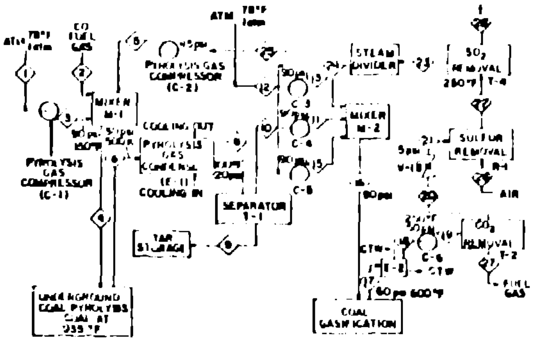

Fig. 6-LASL *In Situ* Coal Gasification Process Simulation

SUMMARY

Efficient underground coal processing requires that moisture is removed from addressed coal seam sections. In the absence of seam moisture, product gases exhaust at elevated temperatures. Sensible heat from this product stream is most sensibly used for drying and pyrolysis of an adjacent seam section. Underground conversion done in this way leads to significant supplies of hydrocarbon gases for future energy needs.

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