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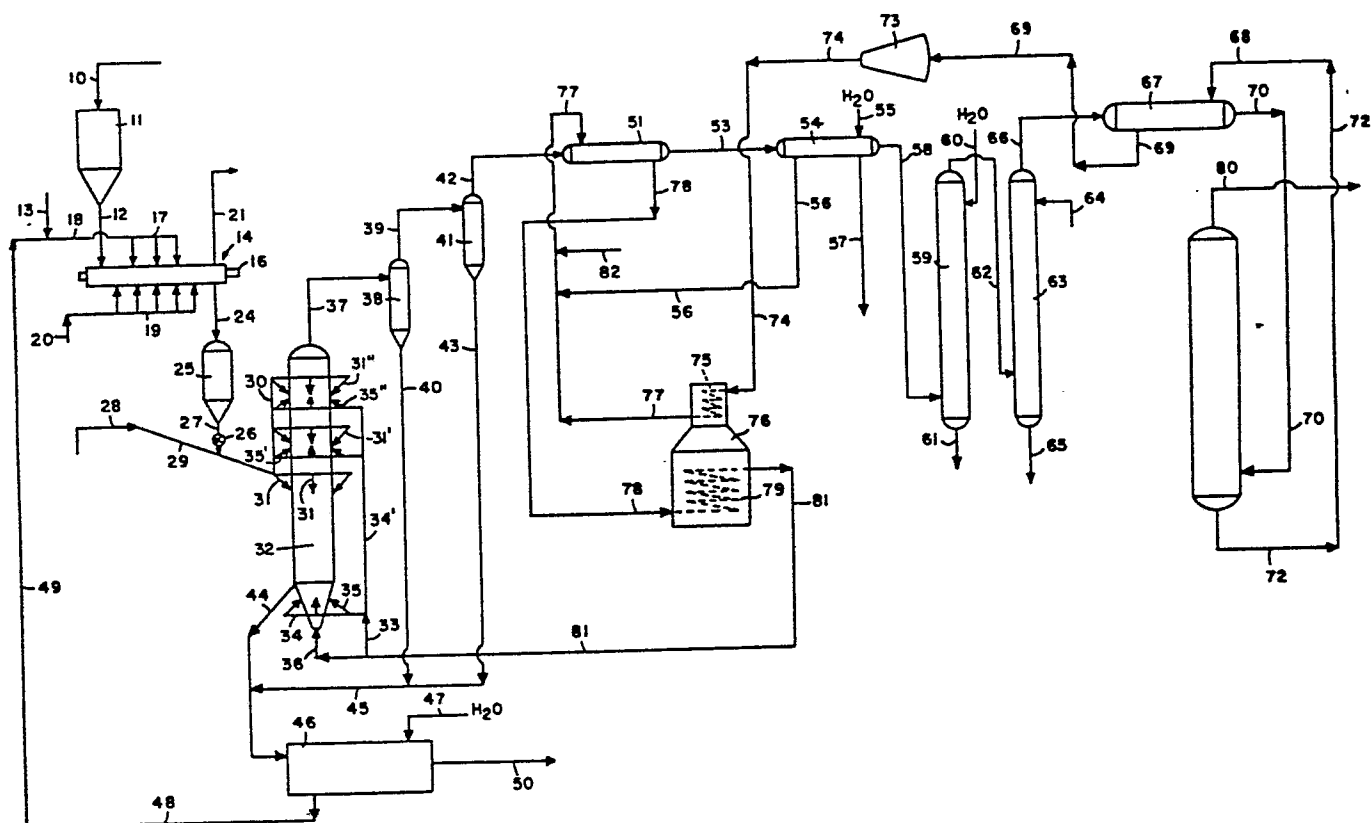
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54 **Coal conversion process.**

57 This concerns a gasification process for solid carbonaceous materials in a fluidised bed in the presence of steam, added hydrogen, carbon monoxide and a catalyst. The reactor volume is reduced by elevating either the point or points at which the solid carbonaceous feed is introduced, the point or points wherein hydrogen and carbon monoxide are introduced or the point or points at which both the solid carbonaceous feed and the carbon monoxide and hydrogen are introduced. When the points of feed introduction 31'-31" and the points of carbon monoxide and hydrogen introduction 35'-35" are elevated, the reduction in volume is greater than would have been predicted from a relocation of either of these points separately.

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"Coal Conversion Process"

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2 This invention relates to an improved process
3 for converting solid carbonaceous materials. More par-
4 ticularly, this invention relates to a process for
5 gasifying solid carbonaceous materials.
6 Before the turn of the century it was known
7 that hydrocarbon gases and liquids, tars and chemicals
8 could be obtained not only from petroleum, but from coal
9 and other carbonaceous liquids solids. Very early
10 processes employed destructive distillation, coal being
11 transformed into gases and petroleum-like liquid products.
12 Primary emphasis in many of these processes is on gasifi-
13 cation of the coal with the objective of improving
14 processes for the production of water gas, producer gas,
15 or hydrogen, as opposed to the production of coal liquids.
16 For the past several decades, due to disallocations of
17 supplies, there have been reoccurring periods of interest
18 in the gasification of coal to produce fuel gases, first
19 primarily in Europe; and then, in this country. The art
20 reflects the various periods of interest in terms of peaks
21 defined by large numbers of patents, and literature.
22 Presently existing and projected shortages of natural gas
23 in this country have sparked a renewed and very keen
24 interest in the gasification of coal, and it appears that
25 this will be a long-range trend. Consequently, intensive
26 research and development efforts are now underway to
27 produce synthetic high-BTU, intermediate-BTU and synthesis
28 fuel gases for commercial usages.

29 It was early recognized that some mineral and
30 trace inorganic constituents naturally present in some
31 coal could exert favorable catalytic influences in gasi-
32 fication reactions vis-a-vis thermal reactions, and a
33 variety of catalytic materials have been added to coal to
34 alter the natural chemistry inherent in various of the
35 early coal gasification processes. The thrust of present
36 research is to develop processes for the production of
37 synthetic high-BTU gases with far higher efficiencies than

1 was possible in the classical European, or early Euro-
2 American processes. There are, however, inherent chemical
3 kinetic limitations in coal gasification processes which
4 have defied solution, and these problems are yet unsolved.
5 Yet, solution is essential, and there remains a strong
6 interest in providing better coal gasification processes,
7 or catalysts for use in catalytic coal gasification
8 processes.

9 In a coal gasification process, i.e., one whose
10 object is to produce a high BTU gas, an intermediate BTU
11 gas or a synthesis fuel gas; steam or a similar reagent
12 and particulate coal are fed to a gasifier at elevated
13 temperature and pressure and converted to a synthesis gas,
14 or gaseous mixture of high methane content, which contains
15 significant amounts of carbon monoxide and hydrogen.
16 Recently, it has been proposed to separate the methane
17 from the carbon monoxide and hydrogen in a catalytic
18 process and to then recycle the carbon monoxide and
19 hydrogen to improve thermal efficiency where a high BTU
20 gas is desired or recycle a portion of the entire stream
21 when an intermediate BTU gas is desired. Generally, the
22 methane in the recycle stream may be reformed to carbon
23 monoxide and hydrogen prior to the recycling step. More-
24 over, the entire hydrocarbon gas may be reformed when
25 synthesis gas is the desired product. Processes of this
26 type are described in U. S. Patents Nos. 4,094,650 and
27 4,118,204. Practical objectives, however, require thermal
28 efficiencies coupled with a reduction of reactor size.

29 More recently, it has been discovered that the
30 recycling of hydrogen to the reactor retards the steam
31 gasification of coal and similar liquid and solid carbona-
32 ceous materials thereby increasing the size of the gasifi-
33 cation reactor. The need for an improved gasification
34 process exhibiting the same thermal efficiency but re-
35 quiring a smaller gasification reactor is, therefore,
36 believed to be readily apparent.

1
2 It has now, surprisingly, been discovered that
3 the foregoing disadvantages of the prior art gasification
4 processes may be overcome with the method of the present
5 invention and an improved gasification process provided
6 thereby. It is, therefore, an object of this invention
7 to provide an improved method for gasifying carbonaceous
8 materials. It is another object of this invention to
9 provide such an improved process wherein the thermal
10 efficiency normally associated with the introduction of
11 carbon monoxide and hydrogen directly to the gasification
12 reactor is realized. It is still another object of this
13 invention to provide such an improved process wherein a
14 smaller gasification reactor is required. These and
15 other objects and advantages will become apparent from
16 the description set forth hereinafter.

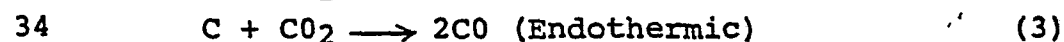
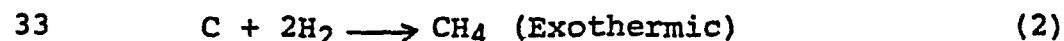
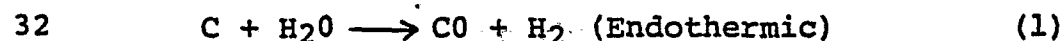
17 In accordance with this invention, the foregoing
18 and other objects and advantages are accomplished by gasi-
19 fying a carbonaceous material in a fluid bed at elevated
20 temperatures and pressures such that either the carbona-
21 ceous feed, added hydrogen and carbon monoxide or all
22 three are introduced into the gasification reactor at a
23 point generally above the bottom of the bed but suffi-
24 ciently below the top thereof to permit substantial
25 equilibration of the gas phase and to avoid tar break-
26 through from the bed. Surprisingly, thermal efficiency
27 is maintained by introducing added carbon monoxide and
28 hydrogen into the gasification reactor and into the fluid
29 bed at an elevated point or points. As pointed out more
30 fully hereinafter, raising the point or points of intro-
31 duction of the carbonaceous feed and the carbon monoxide-
32 hydrogen mixture results in a synergistic reduction in
33 gasification reactor size.

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1
2 As indicated, supra, the present invention
3 relates to an improved process for gasifying carbonaceous
4 materials. Thermal efficiency is enhanced by introducing
5 a mixture of carbon monoxide and hydrogen into a fluid bed
6 comprising the carbonaceous material at varying degrees
7 of gasification. The gasifier reactor size required is
8 reduced by elevating the point or points at which either
9 the carbon monoxide-hydrogen mixture or the carbonaceous
10 feed is introduced to the fluid bed. Maximum reduction
11 in the gasifier reactor size required is realized when
12 both the carbon monoxide-hydrogen mixture and the carbon-
13 aceous feed are introduced into the fluid bed at a point
14 or points above the bottom of the fluid bed and suffi-
15 ciently below the top to permit substantial equilibration
16 of the gas phase and to avoid tar break through from the
17 bed.

18 In general, the process of this invention may be
19 used to gasify any carbonaceous material that will fluidize
20 in a gas stream. The process is, therefore, particularly
21 suited to the gasification of solid carbonaceous materials
22 such as coal, coal char, metallurgical coke, petroleum
23 coke, charcoal, activated carbon and the like. In some
24 cases, inert carriers having carbon deposited on the
25 surfaces thereof may also be gasified in the process of
26 this invention.

27 As indicated previously, the gasification of coal
28 and similar carbonaceous materials normally produces a
29 synthesis gas composed primarily of hydrogen and carbon
30 monoxide. The principal reactions which take place in
31 such a system include the following:



36 The reaction kinetics during conventional gasification
37 operations are such that the product gas normally contains

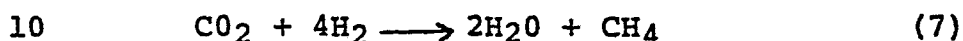
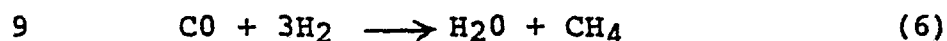
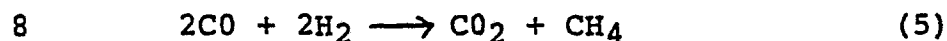
1 varying amounts of methane. In steam gasification, the
2 methane which is present occurs primarily as a result of
3 devolatilization of the coal. The direct hydrogenation of
4 carbon in accordance with equation (2) above is known to
5 be very slow as compared to the endothermic reactions of
6 steam and carbon dioxide with carbon as set forth in
7 equations (1) and (3). The products of conventional steam
8 gasification operations are thus primarily hydrogen and
9 carbon monoxide and such operations are highly endothermic.
10 As pointed out earlier, it has been proposed that this
11 endothermicity be reduced by carrying out the operation
12 in the presence of hydrogen to promote the exothermic
13 carbon-hydrogen reaction of equation (2) but this normally
14 requires a substantially higher reaction temperature than
15 is needed for the steam-carbon reaction. Moreover, it is
16 now known that hydrogen inhibits the reaction of steam
17 with carbon to produce carbon monoxide and hydrogen. As
18 a result, gasifier reactors of a larger size than would
19 otherwise be required are required when hydrogen and/or
20 carbon monoxide-hydrogen mixtures are introduced at or
21 near the bottom of the gasifier.

22 In general, the gasifier reactor size is reduced
23 when the feed point or points are raised above the bed
24 since the inhibiting effect of the devolatilization gases
25 is limited to that portion of the bed at and above the
26 point of feed introduction. In this regard, it should be
27 noted that the devolatilization will occur rapidly after
28 feed introduction and the gases will flow upwardly. The
29 solid particles, on the other hand, will be distributed
30 throughout the entire fluid bed since such a bed approaches
31 a perfectly mixed characterization. Ideally, the feed
32 point or points will be located at the highest elevation
33 possible without tar breakthrough from the bed.

34 Similarly, the gasifier reactor size is reduced
35 when the point or points at which carbon monoxide and
36 hydrogen are introduced is raised in the reactor. In
37 this regard, it should be noted that the introduction of

1 carbon monoxide and hydrogen into the gasifier will
2 improve thermal efficiency in gasification processes of
3 the type described in U. S. Patents No. 4,094,650 and
4 No. 4,118,204.

5 In addition to the gas phase
6 reaction illustrated by equation (4) above, the following
7 gas phase reactions occur:



11 All of these gas phase reactions are exothermic and, when
12 an amount of carbon monoxide and hydrogen in excess of the
13 equilibrium amount is introduced, heat is produced.

14 Ideally, then, the point or points of introduction will
15 be at the highest elevation which will permit the gas
16 phase to substantially reach equilibrium at the top of
17 the fluid bed. In this way, the heat generated will be
18 distributed throughout the fluid bed by the well mixed
19 solid particles comprising the bed.

20 In general, commercial scale gasification fluid
21 beds will range in height from about 60 to about 125 feet
22 and both the carbonaceous feed and carbon monoxide and
23 hydrogen feed to the bed will, independently and generally,
24 be introduced at a point or points along the bed and within
25 a range of distances ranging from about 10% of the total
26 bed height below the top of the bed to about 60% of the
27 total bed height from the top. Such disposition will, of
28 course, result in a maximum size reduction or a minimum bed
29 height for any given gasification operation. It is,
30 however, within the scope of this invention to position
31 either the carbonaceous feed or the carbon monoxide and
32 hydrogen introduction within the specified range with the
33 other being introduced at or near the bottom of the bed
34 in a manner consistent with the prior art.

35 It is believed that the present invention will
36 be better understood by reference to the appended drawing.
37 Referring to the drawing, then, the process illustrated is

1 one for the production of a chemical synthesis gas by the
2 gasification of carbonaceous material such as coal, lig-
3 nite, coal char, coke or similar carbonaceous material
4 with steam at an elevated temperature in the presence
5 of a catalyst.

6 In the embodiment illustrated, a particularly
7 preferred catalyst is prepared by impregnating the feed
8 solids with a solution of an alkali metal compound or
9 mixture of such compounds and thereafter heating the
10 impregnated material to a temperature sufficient to pro-
11 duce an interaction between the alkali metal and the
12 carbon present. Generally, the solid feed material will
13 be finely divided to a particle size suitable for fluidi-
14 zation and a particle size of about 8 mesh or smaller on
15 the U. S. Sieve Series Scale is particularly suitable. In
16 the embodiment illustrated, the feed is passed into line
17 10 from a feed preparation plant or storage facility that
18 is not shown in the drawing. The solids introduced into
19 line 10 are fed into a hopper or similar vessel 11 from
20 which they are passed through line 12 into feed preparation
21 zone 14. This zone contains a screw conveyor or similar
22 device, not shown in the drawing, that is powered by a
23 motor 16, a series of spray nozzles or similar devices 17
24 for the spraying of an alkali metal-containing solution
25 supplied through line 18 onto the solids as they are moved
26 through the preparation zone by the conveyor, and a similar
27 set of nozzles or the like 19 for the introduction of a hot
28 dry gas, supplied through line 20, which serves to heat
29 the impregnated solids and drive off the moisture. A
30 mixture of water vapor and gas is withdrawn from zone 14
31 through line 21 and passed to a condenser, not shown, from
32 which water may be recovered for use as makeup or the like.
33 The majority of the alkali metal-containing solution is
34 recycled through line 49 from the alkali metal recovery
35 portion of the process, which is described hereafter. Any
36 makeup alkali metal solution required may be introduced
37 into line 18 via line 13.

1 In general, sufficient alkali metal-containing
2 solution is introduced into preparation zone 14 to provide
3 from about 1 to about 50 weight percent of an alkali metal
4 compound or mixture of such compounds on the coal or other
5 carbonaceous solids. From about 5 to about 30 percent is
6 preferred. The dried impregnated solid particles prepared
7 in zone 14 are withdrawn through line 24 and passed to a
8 closed hopper or similar vessel 25 from which they are
9 discharged through a star wheel feeder or equivalent device
10 26 in line 27 at an elevated pressure sufficient to permit
11 their entrainment into a stream of high pressure steam,
12 inert gas or other carrier gas introduced into line 29 via
13 line 28. Moreover, it is within the scope of this inven-
14 tion to use all or a portion of the carbon monoxide and
15 hydrogen as a carrier gas. The carrier gas and entrained
16 solids are passed through line 29 into manifold 30 and
17 introduced into the gasifier 32 through any one or more of
18 a plurality of feed points 31, 31' and 31''. As previously
19 indicated, the uppermost or highest feed point will be at
20 least about 10% of the total fluid bed height below the
21 top of the fluid bed and the lowest feed point will be no
22 more than 60% of the total bed height below the top of the
23 fluid bed. In lieu of or in addition to hopper 25 and
24 star wheel feeder 26, the feed system may employ parallel
25 lock hoppers, pressurized hoppers, aerated standpipes
26 operated in series, or other apparatus to raise the input
27 feed solids stream to the required pressure level.

28 In general, the gasifier 32 will be operated at
29 a pressure between about 100 and 1500 psia, preferably at
30 a pressure within the range of about 200 and 800 psia for
31 any desired product distribution. The carrier gas may be
32 preheated to a temperature in excess of about 300°F, but
33 below the initial softening point of the coal or other
34 feed material employed. Feed particles may be suspended
35 in the carrier gas in a concentration between about 0.2
36 and about 5.0 pounds of solid feed material per pound of
37 carrier gas. The optimum ratio for a particular system

1 will depend in part upon the particle size and density,
2 the molecular weight of the gas employed, the temperature
3 of the solid feed material and the input gas stream, the
4 amount of alkali metal compound employed and other factors.
5 In general, ratios between about 0.5 and about 4.0 pounds
6 of solid feed material per pound of carrier gas are pre-
7 ferred.

8 Gasifier 32 contains a fluidized bed of carbon-
9 aceous solids extending upward within the vessel above an
10 internal grid or similar distribution device not shown in
11 the drawing. The bed is maintained in the fluidized state
12 by means of steam introduced through bottom inlet 36. The
13 bed may be partially maintained with carbon monoxide and
14 hydrogen introduced through line 33, manifold 34 and peri-
15 pherally spaced injection lines and nozzles 35, when a part
16 or all of carbon monoxide and hydrogen are introduced at
17 the bottom of the bed. As previously indicated, however,
18 hydrogen retards the desirable gasification reactions and
19 is, therefore, preferably introduced into the fluid bed at
20 a higher elevation. When this is done, the carbon monoxide
21 and hydrogen may be introduced through any one or more of
22 a plurality of injection points 35'-35'' which are supplied
23 by manifold 33' and which are independently positioned in
24 the same portion of the fluid bed as the carbonaceous feed
25 points 31, 31' and 31''. Alternatively, all or a part of
26 the carbon monoxide and hydrogen could be introduced into
27 the fluid bed with the carbonaceous feed, as previously
28 indicated, through line 28.

29 The particular injection system shown in the
30 drawing is not critical and hence other methods for in-
31 jecting the steam, hydrogen and carbon monoxide may be
32 employed. In some instances, for example, it may be pre-
33 ferred to introduce the gases through multiple nozzles to
34 obtain more uniform distribution of the injected fluid
35 and reduce the possibility of channeling and related
36 problems. The space velocity of the rising gases within
37 the fluidized bed will normally be between about 100 and

1 about 3000 actual volumes of steam, hydrogen and carbon
2 monoxide per hour per volume of fluidized solids.

3 With the fluidized bed in gasifier 32, the
4 carbonaceous solids are subjected to a temperature within
5 the range between about 1000°F and about 1500°F, preferably
6 between about 1200°F and 1400°F. At such a temperature the
7 carbon-alkali metal catalyst will equilibrate the gas
8 phase reactions occurring during gasification to produce
9 additional methane and at the same time supply substantial
10 amounts of additional exothermic heat in situ.

11 Due to the gas phase equilibrium conditions
12 existing as a result of the carbon-alkali metal catalyst
13 and due to the presence of added hydrogen and carbon mono-
14 oxide, there will be a net heat production. Moreover,
15 competing reactions, that in the absence of the catalyst
16 and the added hydrogen and carbon monoxide would ordinarily
17 tend to produce additional hydrogen and carbon monoxide,
18 are suppressed. The heat produced tends to balance the
19 endothermic heat consumed by the reaction of the steam
20 with carbon and as the amount of carbon monoxide and
21 hydrogen added increases, an overall thermoneutral reaction
22 can be approached. When the gasifier is basically in heat
23 balance, the heat required to preheat the feed to the
24 reaction temperature and compensate for heat losses from
25 the gasifier is supplied for the most part by excess heat
26 in the gases introduced into the gasifier through line 36.
27 In the absence of the exothermic heat provided by the
28 catalyzed gas phase reactions, these gases would have to
29 be heated to substantially higher temperatures than those
30 normally employed here.

31 The gas leaving the fluidized bed in gasifier 32
32 passes through the upper section of the gasifier, which
33 serves as a disengagement zone where the particles too
34 heavy to be entrained by the gas leaving the vessel are
35 returned to the bed. If desired, this disengagement zone
36 may include one or more cyclone separators or the like for
37 removing relatively large particles from the gas. The gas

1 withdrawn from the upper part of the gasifier through line
2 37 will normally contain an equilibrium mixture at reaction
3 temperature and pressure of methane, carbon dioxide,
4 hydrogen, carbon monoxide and unreacted steam. Hydrogen
5 sulfide, ammonia and other contaminants formed from sulfur
6 and nitrogen contained in the feed material may also be
7 present in this gas and entrained fines may also be
8 present.

9 As is well known, basically the same gaseous
10 effluent will be produced in the gasifier when steam is
11 used to effect the gasification. As is also well known,
12 the ultimate gaseous product depends upon the further
13 processing to which this effluent is subjected. As a
14 result, the improvement of this invention is equally
15 applicable to any catalytic process wherein a carbonaceous
16 material is gasified in the presence of steam.

17 In the embodiment illustrated, the effluent gas
18 is introduced into cyclone separator or similar device 38
19 for removal of the larger fines. The overhead gas then
20 passes through line 39 into a second separator 41 where
21 smaller particles are removed. The gas from which the
22 solids have been separated is taken overhead from separator
23 41 through line 42 and the fines are discharged downward
24 through dip legs 40 and 43. These fines may be returned
25 to the gasifier or passed to the alkali metal recovery
26 portion of the process.

27 In the system shown in the drawing, a stream of
28 high ash content char particles is withdrawn through line
29 44 from gasifier 32 in order to control the ash content of
30 the system and permit the recovery and recycle of alkali
31 metal constituents of the catalyst. The solids in line 44,
32 which may be combined with fines recovered from the gasi-
33 fier overhead gas through dip legs 40 and 43 and line 45,
34 are passed to alkali metal recovery unit 46. The recovery
35 unit will normally comprise a multistage countercurrent
36 leaching system in which the high ash content particles
37 are countercurrently contacted with water introduced

1 through line 47. An aqueous solution of alkali metal
2 compounds is withdrawn from the unit through line 48 and
3 recycled through lines 49 and 18 to feed preparation zone
4 14. Ash residues from which soluble alkali metal compounds
5 have been leached are withdrawn from the recovery unit
6 through line 50 and may be disposed of as land fill or
7 further treated to recover added alkali metal constituents.

8 The gas leaving separator 41 is passed through
9 line 42 to gas-gas heat exchanger 51 where it is cooled by
10 indirect heat exchange with a gaseous mixture of methane
11 and steam introduced through line 77. The cooled gas is
12 then passed through line 53 into waste heat boiler 54
13 where it is further cooled by indirect heat exchange with
14 water introduced through line 55. Sufficient heat is
15 transferred from the gas to the water to convert it into
16 steam, which is withdrawn through line 56. During this
17 cooling step, unreacted steam in the gas from exchanger
18 51 is condensed out and withdrawn as condensate through
19 line 57. The cool gas leaving waste heat boiler 54
20 through line 58 is passed to water scrubber 59. Here the
21 gas stream passes upward through the scrubber where it
22 comes in contact with water injected into the top of the
23 scrubber through line 60. The water absorbs ammonia and
24 a portion of the hydrogen sulfide in the gas stream and is
25 withdrawn from the bottom of the scrubber through line 61
26 and passed to downstream units for further processing.
27 The water scrubbed gas stream is withdrawn from the
28 scrubber through line 62 and is now ready for treatment
29 to remove bulk amounts of hydrogen sulfide and other acid
30 gases.

31 The gas stream is passed from water scrubber 59
32 through line 62 into the bottom of solvent scrubber 63.
33 Here the gas passes upward through the contacting zone in
34 the scrubber where it comes in contact with a downflowing
35 stream of solvent such as monoethanolamine, diethanolamine,
36 a solution of sodium salts of amino acids, methanol, hot
37 potassium carbonate or the like introduced into the upper

1 part of the solvent scrubber through line 64. If desired,
2 the solvent scrubber may be provided with spray nozzles,
3 perforated plates, bubble cap plates, packing or other
4 means for promoting intimate contact between the gas and
5 the solvent. As the gas rises through the contacting zone,
6 hydrogen sulfide, carbon dioxide and other acid gases are
7 absorbed by the solvent, which leaves the scrubber through
8 line 65. The spent solvent containing carbon dioxide,
9 hydrogen sulfide and other contaminants is passed through
10 line 65 to a stripper, not shown in the drawing, where it
11 is contacted with steam or other stripping gas to remove
12 the absorbed contaminants and thereby regenerate the
13 solvent. The regenerated solvent may then be reused by
14 injecting it back into the top of the scrubber via line 64.

15 A clean gas containing essentially methane,
16 hydrogen, and carbon monoxide in amounts substantially
17 equivalent to the equilibrium quantities of those gases
18 in the raw product gas withdrawn from gasifier 32 through
19 line 37 is withdrawn overhead from the solvent scrubber
20 via line 66. The methane content of the gas will normally
21 range between about 20 and about 60 mole percent and the
22 gas will be of an intermediate BTU heating value, normally
23 containing between about 400 and about 750 BTUs per
24 standard cubic foot.

25 As will be readily apparent, this intermediate
26 BTU gas could be withdrawn as a product. When this is
27 done a portion of the product could be separated and then
28 subjected to steam reforming to produce the carbon monoxide
29 and hydrogen required for improved thermal efficiency.
30 Alternatively, the carbon monoxide and hydrogen could be
31 provided from any of the sources therefor known in the
32 prior art.

33 The intermediate BTU gas withdrawn overhead from
34 solvent scrubber 63 through line 66 is introduced into
35 heat transfer unit 67 where it passes in indirect heat
36 exchange with liquid methane introduced through line 68.
37 The methane vaporizes within the heat transfer unit and is

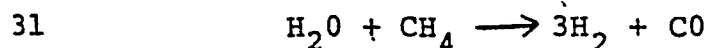
1 discharged as the intermediate BTU gas, which is primarily
2 composed of methane, hydrogen and carbon monoxide, is
3 cooled to a low temperature approaching that required for
4 liquefaction of the methane contained in the gas, after
5 which the chilled gas is passed through line 70 into
6 cryogenic unit 71. Here the gas is further cooled by
7 conventional means until the temperature reaches a value
8 sufficiently low to liquefy the methane under the pressure
9 conditions existing in the unit. Compressors and other
10 auxiliaries associated with the cryogenic unit are not
11 shown. The amount of pressure required for the liquefac-
12 tion step will depend in part upon the pressure at which
13 the gasifier is operated and the pressure losses which are
14 incurred in the various portions of the system. A sub-
15 stantially pure stream of liquefied methane is taken off
16 through line 72 and may be withdrawn as product. In the
17 In the embodiment illustrated, however, the methane is
18 passed through line 68 into heat transfer unit 67 as
19 described earlier. Hydrogen and carbon monoxide are
20 withdrawn overhead from cryogenic unit 71 through line 80
21 and recovered as a chemical synthesis product gas.
22 Normally, the cryogenic unit is operated and designed in
23 such a manner that less than about 10 mole percent of
24 methane, preferably less than about 5 mole percent,
25 remains in the product gas removed through line 80. Thus,
26 the chemical synthesis gas produced in the process is one
27 of extremely high purity and therefore has many industrial
28 applications.

29 As previously indicated, the methane could be
30 withdrawn as product and the carbon monoxide and hydrogen
31 separated in the cryogenic separator returned to the
32 gasifier to facilitate thermal efficiency. In the embodi-
33 ment illustrated, however, the recycle methane gas removed
34 from heat transfer unit 67 and through line 69 is passed
35 to compressor 73 where its pressure is increased to a value
36 from about 25 psi to about 150 psi above the operating
37 pressure in gasifier 32. The pressurized gas is withdrawn

1 from compressor 73 through line 74 and passed through
2 tubes 75 located in the convection section of steam
3 reforming furnace 76. Here, the high pressure gas picks
4 up heat via indirect heat exchange with the hot flue gases
5 generated in the furnace. The methane gas is removed from
6 the tubes 75 through line 77 and mixed with steam, which
7 is generated in waste heat boiler 54 and injected into
8 line 77 via line 56. The mixture of methane gas and
9 steam is then passed through line 77 into gas-gas heat
10 exchanger 51 where it is heated by indirect heat exchange
11 with the raw product gas removed from separator 41. The
12 heated mixture is removed from exchanger 51 and passed
13 through line 78 to steam reforming furnace 76.

14 The preheated mixture of steam and methane gas
15 in line 78 is introduced into the internal tubes 79 of
16 the steam reforming furnace where the methane and steam
17 react with one another in the presence of a conventional
18 steam reforming catalyst. The catalyst will normally
19 consist of metallic constituents supported on an inert
20 carrier. The metallic constituent will normally be
21 selected from Group VI-A and the iron group of the
22 Periodic Table and may be chromium, molybdenum, tungsten,
23 nickel, iron, and cobalt, and may include small amounts of
24 potassium carbonate or a similar compound as a promoter.
25 Suitable inert carriers include silica, alumina, silica-
26 alumina, zeolites, and the like.

27 The reforming furnace is operated under condi-
28 tions such that the methane in the feed gas will react
29 with steam in the tubes 79 to produce hydrogen and carbon
30 monoxide according to the following equation:



32 The temperature in the reforming furnace will normally be
33 maintained between about 1200°F and about 1800°F, prefer-
34 ably between about 100°F and about 300°F above the
35 temperature in gasifier 32. The pressure will range
36 between about 10 and about 30 psi above the pressure in
37 the gasifier. The mole ratio of steam to methane

1 introduced into the reactor will range between about 2:1
2 and about 15:1, preferably between about 3:1 and about
3 7:1. The reforming furnace may be fired by a portion of
4 the methane gas removed from heat transfer unit 67 via
5 line 69, a portion of the intermediate BTU gas removed
6 from solvent scrubber 63 through line 66, or a similar
7 fuel gas.

8 The gaseous effluent stream from the steam
9 reforming furnace, which will normally be a mixture con-
10 sisting primarily of hydrogen, carbon monoxide, and un-
11 reacted steam, is passed, preferably without substantial
12 cooling, through lines 81 to manifolds 33 and/or 34' and
13 ultimately into gasifier 32. This stream will be the
14 primary source of the hydrogen, carbon monoxide and steam
15 required in the gasifier. In a preferred embodiment,
16 therefore, it is desirable that the reforming furnace
17 effluent contain sufficient carbon monoxide and hydrogen
18 to provide the desired thermal balance.

19 As pointed out previously, substantial quantities
20 of exothermic heat are released in the gasifier as a
21 result of the reaction of hydrogen with carbon oxides and
22 the reaction of carbon monoxide with steam. Thus, the
23 carbon monoxide and hydrogen in the reformer effluent
24 stream comprises a substantial portion of the heat input
25 into the gasifier. To supply the desired amounts of
26 hydrogen and carbon monoxide in the effluent, sufficient
27 methane should normally be present in the feed to the
28 reforming furnace so that enough carbon monoxide and
29 hydrogen is produced by steam reforming the methane to
30 compensate for the amount of hydrogen and carbon monoxide
31 removed in the chemical synthesis product gas withdrawn
32 from the process overhead of cryogenic unit 71 through
33 line 80.

34
35 In a preferred embodiment of the present inven-
36 tion, coal will be gasified with steam in the presence of
37 an alkali metal catalyst and at a temperature within the

1 range from about 1200 to about 1400°F and at a pressure
2 within the range from about 200 to about 600 psia. The
3 gasification will be accomplished in a fluid bed having a
4 bed height within the range from about 60 to about 130 feet.
5 The fluid bed will be maintained with steam introduced at
6 the bottom of the gasification vessel and distributed
7 through a suitable grid. The coal feed will be introduced
8 into the fluid bed at one or more points located within the
9 range from about 20 percent of the total height below the
10 top of the bed to about 50 percent of the total height
11 below the bed. The catalytic process will be operated so
12 as to produce a substitute natural gas and substantially
13 all of the carbon monoxide and hydrogen contained in the
14 gaseous effluent from the gasifier will be recovered and
15 recycled to the gasification vessel. The recycled carbon
16 monoxide and hydrogen will be introduced into the fluid
17 bed at one or more points positioned along the fluid bed
18 within from about 20 percent of the total height from the
19 top of the bed to about 50 percent of the total height
20 from the top of the bed.

21 In the preferred embodiment, the amount of
22 carbon monoxide and hydrogen recycled will be equal to the
23 amount of carbon monoxide and hydrogen which would be
24 produced as a result of the steam gasification of the coal
25 if no carbon monoxide and hydrogen were introduced and when
26 sufficient nominal holding time is provided to permit
27 equilibration of the gaseous effluent from the gasifier.
28 Also in the preferred embodiment, the exact point or points
29 of the coal feed introduction will be optimized as a
30 function of the activity of the coal to steam gasification.

31 Having thus broadly described the invention and
32 set forth a preferred embodiment thereof, it is believed
33 that the invention will be even better understood by
34 reference to the following Example.

35

36

37

1 EXAMPLE

2 In this example, a series of steam gasifications
3 was completed over a range of gasification temperatures
4 of 1275°F and pressures of 5000 psia and at a steam to
5 coal ratio of 1.58. From these tests, a kinetic model
6 was developed and from this model, it has been predicted
7 that the optimum fluid bed volume can be reduced by 11
8 percent by raising the coal feed point to a height within
9 the range from about 10% below the top of the bed to about
10 60 percent of the total height below the top of the bed.
11 It has also been predicted from this model that the reactor
12 volume can be reduced by 27 percent if an equilibrium
13 mixture of carbon monoxide and hydrogen is introduced at a
14 point or points located at a point below the top of the
15 bed by an amount equal to about 10 percent of the total
16 height. It has further been predicted that the total
17 volume can be reduced by 42 percent if both the feed
18 point and the carbon monoxide and hydrogen point or
19 points of introduction are both relocated. Based on
20 predictions from the results obtained by relocating
21 each feed point separately, it was anticipated that only
22 a 35 percent reduction would have been realized by re-
23 locating both feed points simultaneously.

24 From the foregoing, it is believed readily
25 apparent that elevation of either the feed point or the
26 carbon monoxide and hydrogen introduction point will
27 result in a significant reduction in reactor size or
28 required bed height. It is also believed readily apparent
29 that if both of these feed points are elevated a signifi-
30 cant and synergistic reduction in total bed height is
31 realized.

32 While the present invention has been described
33 and illustrated by reference to a particular embodiment
34 thereof, it will be appreciated by those of ordinary skill
35 in the art that the same lends itself to variations not
36 necessarily illustrated herein.

CLAIMS

1. A gasification process wherein a solid carbonaceous material is gasified in a fluidized bed in the presence of steam, added hydrogen, CO and a catalyst and wherein either the solid carbonaceous material or the added hydrogen and carbon monoxide or all three are introduced into the fluid bed at a point or points along the bed and within a range of distances ranging from about 10 percent of the total bed height below the top of the bed to about 60 percent of the total bed height from the top.

2. A process according to claim 1 wherein an alkali metal catalyst is present during gasification.

3. A process according to either of claims 1 and 2 wherein the amount of hydrogen and CO added to the fluid bed is sufficient to provide the desired thermal balance.

4. A process according to any one of the preceding claims wherein the amount of carbon monoxide and hydrogen added to the fluid bed is equal to the amount of carbon monoxide and hydrogen which would be produced during the steam gasification of the solid carbonaceous material if no carbon monoxide and hydrogen were introduced and when sufficient nominal holding time is provided to permit equilibration of the gaseous effluent from the gasifier.

5. A process according to any one of the preceding claims wherein the added hydrogen and carbon monoxide is recovered from the gaseous effluent from the gasification reactor.

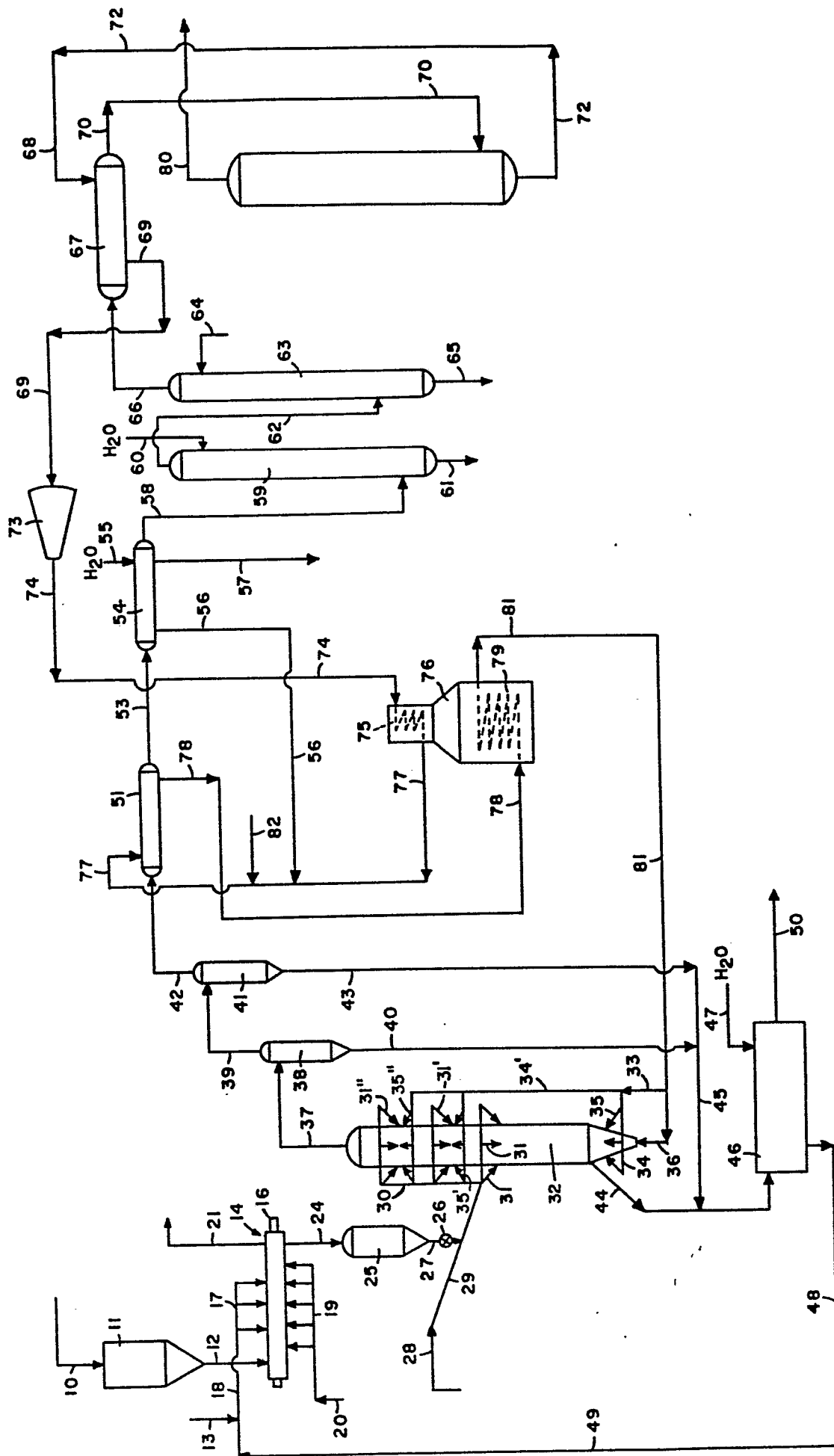
6. A process according to any one of the preceding claims wherein substantially all of the hydrogen and carbon monoxide contained in the gasification reactor effluent is separated and recycled to the fluid bed.

7. A process according to any one of the preceding claims wherein the added hydrogen and carbon monoxide is obtained by reforming at least a portion of the hydrocarbon contained in the gasification reactor effluent.

8. A process according to any one of the preceding claims wherein all of the hydrocarbon gas contained in the gasification reactor effluent is separated, reformed and recycled to the fluid bed as a mixture of hydrogen and carbon monoxide.

9. A process according to any one of the preceding claims wherein the carbonaceous material is introduced at one or more points positioned along the fluid bed within a distance from about 20 percent of the total height below the top of the fluid bed to about 50 percent of the total height below the top of the fluid bed.

10. A process according to any one of the preceding claims wherein said carbonaceous material and the hydrogen and carbon monoxide are separately and independently introduced into said fluid bed at said point or points along the bed.





European Patent
Office

EUROPEAN SEARCH REPORT

0044123

Application number

EP 81 30 0493

DOCUMENTS CONSIDERED TO BE RELEVANT			CLASSIFICATION OF THE APPLICATION (Int. Cl.)
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	
	<p><u>US - A - 4 200 495 (LISS)</u></p> <p>* Column 7, lines 31-47; column 8, lines 1-69; column 14, lines 2-60 *</p> <p>--</p> <p><u>US - A - 4 211 669 (EAKMAN)</u></p> <p>* Columns 11-14; claims *</p> <p>--</p> <p><u>DE - A - 2 741 805 (RHEINISCHE BRAUNKOHLENWERKE)</u></p> <p>* Page 1, claim 4; page 2 claim 6; page 8, lines 13-19 *</p> <p>--</p> <p><u>US - A - 3 847 567 (KALINA)</u></p> <p><u>FR - A - 2 381 820 (EXXON)</u></p> <p>& <u>US - A - 4 118 204</u></p> <p>----</p>	<p>1,9,10</p> <p>1-8</p> <p>1</p>	<p>C 10 J 3/54</p> <p>TECHNICAL FIELDS SEARCHED (Int. Cl.)</p> <p>C 10 J 3/54 3/56</p> <p>CATEGORY OF CITED DOCUMENTS</p> <p>X: particularly relevant A: technological background O: non-written disclosure P: intermediate document T: theory or principle underlying the invention E: conflicting application D: document cited in the application L: citation for other reasons</p> <p>&: member of the same patent family corresponding document</p>
<p>The present search report has been drawn up for all claims</p>			
Place of search	Date of completion of the search	Examiner	
The Hague	15-10-1981	WENDLING	