

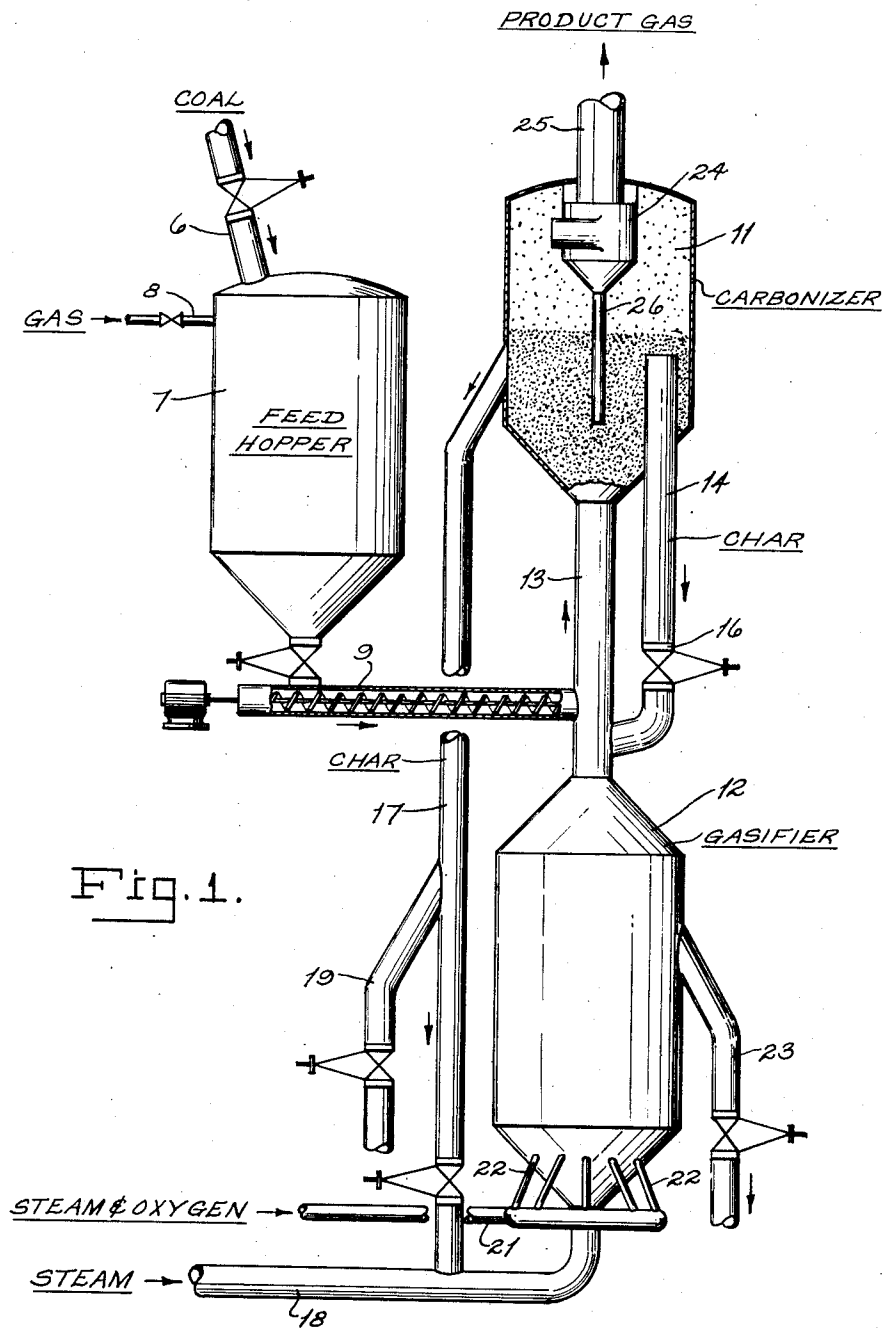
Dec. 15, 1953

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GASIFICATION OF CARBONACEOUS MATERIALS
CONTAINING VOLATILE CONSTITUENTS

2,662,816

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2 Sheets-Sheet 1



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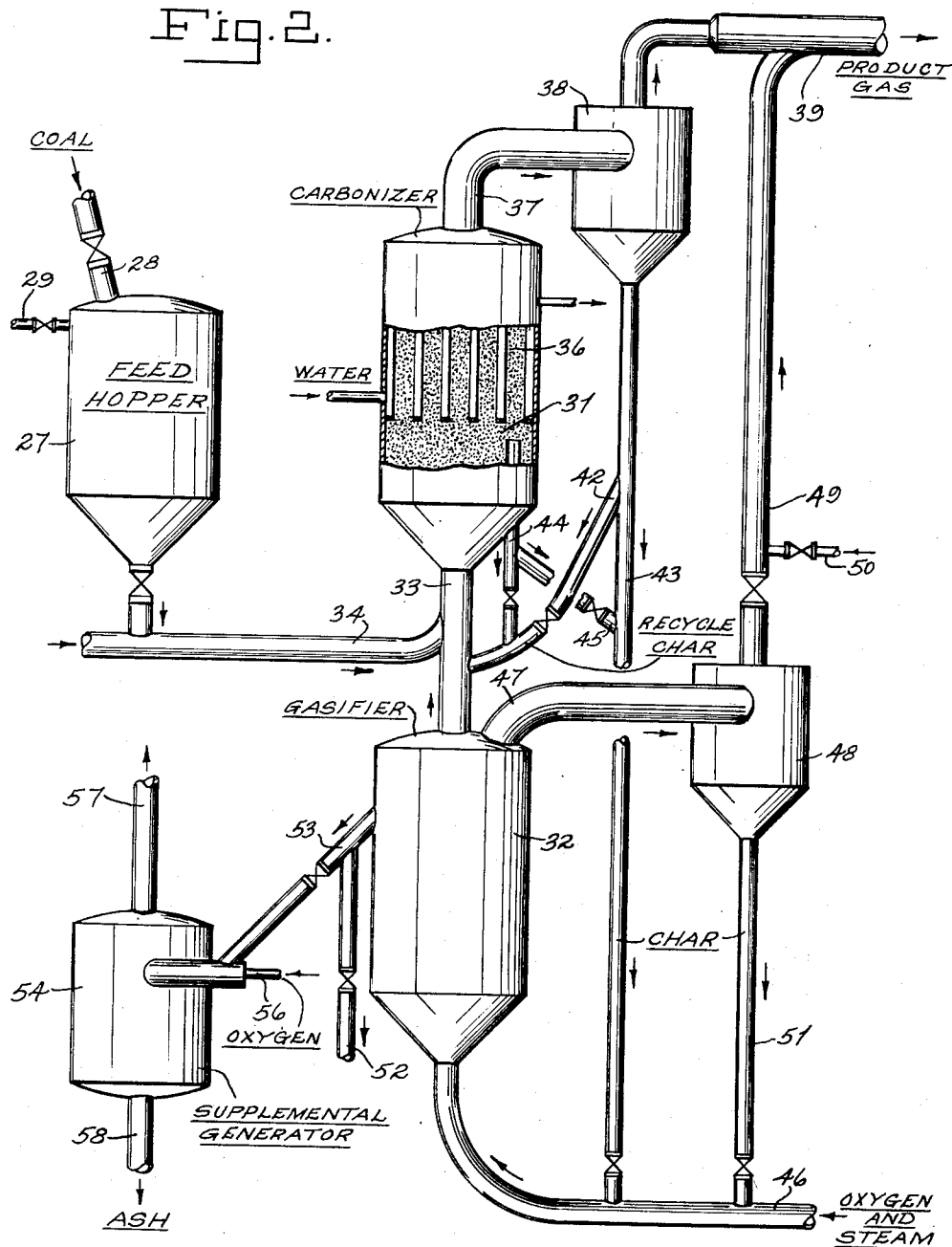
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Fig. 2.



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GASIFICATION OF CARBONACEOUS MATERIALS CONTAINING VOLATILE CONSTITUENTS

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This invention relates to the gasification of a solid carbonaceous material. The process of the invention is applicable to gasification of coal, lignite, oil shale and the like. It is particularly useful for treatment of those carbonaceous materials containing volatile constituents which tend to agglomerate on heating, particularly a coking coal.

The fluidized solids technique has been applied to processes for the gasification of carbonaceous materials. Pulverized coal for example, may be treated in a fluidized bed with hot inert gases to effect removal of volatile constituents or with an oxidizing gas for gasification to any desired extent. Some difficulty is experienced in fluidization of some of the softer coals due to their tendency to become plastic or tacky on heating causing agglomeration of the particles. Coal may be treated prior to gasification to prevent agglomeration. Such pretreatment may consist of heating the coal to drive off a portion of the volatile constituents therefrom or partial pre-oxidation of the coal with an oxygen-containing gas, nitric acid, or other oxidizing agent. An expedient which is used to some extent involves admixing the raw coal with char, ash, or sand to prevent agglomeration of the raw coal particles.

Gasification of carbonaceous materials containing volatile constituents may be accomplished by heating, by chemical combination, or by a combination of both. A somewhat elevated temperature is required for gasification, as is known in the art. The fluidized bed technique aids in maintaining the desired temperature conditions within rather close limits.

The process of the present invention is adaptable to handling of coking coals and the like without difficulty due to agglomerating tendency of the material and without preliminary treatment.

The process of the present invention involves carbonization of the feed material and gasification by the concurrent reaction with oxygen and steam. In carbonization, the volatile constituents are driven off by heat to produce a char or coke residue. Gasification of the carbonized material may be carried to substantial completion leaving only ash or low carbon content char as the residual solid.

In accordance with this invention, the solid carbonaceous material is comminuted to a particle size suitable for fluidization, generally less than one-quarter inch in diameter, and preferably less than about 0.1 inch in diameter, and charged into an agitated mass of particles of hot

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char. Carbonization of the feed material in the resulting mixture takes place in a fluidized bed. The resulting char is passed to a separate zone wherein it is gasified by reaction with oxygen and steam. Gases produced in the gasification zone are used for fluidization in the carbonization zone. Char of reduced carbon content withdrawn from the gasification zone may be used as fuel or for generation of additional product gas. The process of my invention permits operation of the two zones simultaneously with simple positive and flexible temperature control in each zone.

An object of this invention is to provide an improved process for the gasification of solid carbonaceous material.

A further object is to provide a process wherein each step can be carried out under optimum conditions for that step.

Another object is to provide a process for the gasification of solid carbonaceous material which is particularly applicable to carbonization and gasification of coal, lignite, oil shale and similar materials.

Still another object is to provide a process for the gasification of solid carbonaceous materials wherein those materials which tend to soften and agglomerate on heating may be effectively handled.

Other objects and advantages will be apparent from the following detailed description and the accompanying illustrative drawings.

The present invention will be described in detail with reference to coal as the carbonaceous material as typifying the operation and applications of the process of this invention. It will be understood that coal is used as a specific example and that the apparatus and method described are not limited to the use of coal as the carbonaceous feed material. Since the gasification of various materials is known in the art, the application of the present invention to other solid carbonaceous materials will be evident to one skilled in the art from the detailed description of this invention and illustrative examples of its application to treatment of coal.

Fig. 1 is a diagrammatic elevational view, partly in cross section, of one form of apparatus suitable for carrying out the process of my invention.

Fig. 2 is a diagrammatic elevational view, partly in cross section, of an arrangement of apparatus illustrating a modification of the process of my invention.

With reference to the drawing, ground coal, for example, is fed through line 5 into a hopper 1.

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An inert gas may be supplied to the hopper through line 8 to build up pressure in the hopper. The gas also forms an inert blanket avoiding explosion hazards. The particulate coal is fed from the hopper into the gasification apparatus by means of a screw conveyor 9.

The gasification apparatus comprises a carbonization zone 11, a gasification zone 12 and a mixing zone 13 intermediate the carbonization zone and the gasification zone. In the carbonization zone, the volatilizable constituents of the coal are volatilized by heating. In the gasification zone 12, the char or devolatilized coal from the carbonization zone is gasified by reaction with oxygen and steam. Both the carbonization zone and the gasification zone are operated as fluidized beds. In the mixing zone 13, fresh crushed coal is admixed with char and with hot product gases from the gasification zone. The char may be entrained char or ash carried by the hot product gases or char recycled from the carbonization zone. Generally, both will be present to a greater or lesser extent in the mixing zone. The hot gases from the gasifier suspend the particles of coal and char and carry them into the carbonization zone. The gases serve as a fluidizing medium in the carbonization zone to maintain the carbonaceous particles in a fluidized bed and aid in the distillation of the volatile constituents from the fresh coal feed. The quantity of entrained char from the gasification zone may vary considerably depending upon design and operating conditions.

When the particles of coal are brought into the mixing zone into the stream of hot gases and suspended char, the turbulent motion of the gases causes rapid mixing of the coal particles and the char. This prevents agglomeration of particles of coal on heating and facilitates fluidization of the entire mass of solids in the carbonization zone. Preferably the mixing zone is a cylindrical conduit operated with a gas velocity in excess of the terminal velocity of the particles of coal and char. The ratio of char to fresh feed in the mixing zone may vary within the range of from about 3 parts char by weight per part of fresh feed to about 10 parts char per part of feed. A weight ratio of char to feed of about 5:1 is generally satisfactory.

Char from the carbonization zone is supplied to the mixing zone through line 14 as controlled by a suitable valve 16, for example, a slide valve. The char from the carbonization zone is introduced into the product gas stream from the gasification zone at a point upstream from the point of introduction of the coal so that the char is admixed with the gases prior to contact with the fresh feed.

Product char from the carbonization zone is fed through line 17 into admixture with steam in line 18 and passed into the gasification zone 12. A portion of the char from the carbonization zone may be withdrawn, if desired, through line 19 as a high quality product char for other use. Oxygen from line 21 is supplied to the gasification zone through a series of tuyères 22.

Char product from the gasification zone is withdrawn through line 23. Generally, the char product from the gasifier will be of relatively low carbon content and contain substantially all of the ash from the feed material. The carbon content may be controlled by control of the extent of gasification in the gasification zone to produce a char of any desired carbon content (for example, 40 per cent) suitable as fuel for genera-

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tion of steam or the like. The char withdrawn from the gasifier may be subjected to further reaction with oxygen to produce carbon monoxide.

Gases from the carbonizer 11 are passed through a separator 24 to recover entrained solid particles from the gas stream, and the resulting gas, substantially free from solids, is discharged through line 25. The separator 24 may be, for example, a centrifugal type separator capable of removing over 99 per cent of the 70 micron particles. The recovered solid particles are returned to the fluidized bed in the carbonizer through line 26.

The mixture of gases discharged through line 25 may be treated to separate sulfur compounds, carbon dioxide, tar, and oil therefrom. Alternatively, the gases from line 25 may be reacted with steam and oxygen to convert the tar, oils and gaseous hydrocarbons to carbon monoxide and hydrogen. The resulting gases from either mode of procedure may be used as fuel gas or as a source of feed gas, after suitable purification and adjustment of the proportions of carbon monoxide and hydrogen, for the synthesis of hydrocarbons.

The apparatus illustrated in Fig. 2 of the drawing is particularly useful in generation of fuel gases, but may also be utilized to generate other product gases, e. g., synthesis feed gas. The apparatus is generally similar to that of Fig. 1 with some modifications.

With reference to Fig. 2, the gasification apparatus comprises a carbonization zone 31, a gasification zone 32 and a mixing zone 33. Crushed coal enters the feed hopper 27 through line 28; pressurizing gas is admitted through line 29. Coal from the feed hopper 27 is fed into line 34 from which it is introduced to the mixing zone. The coal may be conveyed through line 34 by any suitable means, for example, with a stream of gas, suitably a recycle gas produced in the gasification operation. Alternatively, a mechanical conveyor may be used. The coal introduced into mixing zone 33 is admixed with char and hot gases from the gasifier 32. The coal is suspended in the gases and carried into the fluidized bed in carbonizer 31.

Provision may be made in the carbonizer 36 for cooling the char and gases to remove excess heat and thereby control the operating temperature. The cooling may be effectively accomplished by indirect heat exchange with water, generating steam. The gaseous effluent and entrained carbonaceous particles from the carbonization zone are discharged through line 37 into a separator 38 wherein the particles are separated from the gas stream. A cyclone type separator is suitable for use as separator 38. The resulting gases substantially free from entrained solids are discharged into product gas line 39. The amount of entrainment of char in line 37 may vary considerably depending upon operating conditions.

The char separated in separator 38 may be passed to the mixing zone 33 or to the gasification zone 32 through lines 42 and 43 respectively. Generally, char is fed into both the mixing zone and the gasification zone. The char introduced through line 42 into the mixing zone may be supplemented, if desired, by additional char withdrawn directly from the carbonizer through line 44. This recycled char is admixed with hot gas and entrained char from the gasification zone 32 and with coal from line 34 and returned to the carbonizer 31.

Char from line 43 is admixed with oxygen and steam in line 46 and passed into the fluidized bed in the gasifier 32. The oxygen and steam serve as reactants and as fluidizing gases in the gasifier 32. Oxygen may be admitted separately as in Fig. 1, if desired.

A portion of the product gas from the gasifier 32 is diverted through line 47 to a separator 48. Entrained char is separated from the product gas by the separator which may suitably be of the cyclone type and the gas is discharged through line 49 into the product gas line 39 for further treatment. The separated char may be returned to the gasifier via lines 51 and 46. The gas stream in line 49 may be quenched by introduction of a suitable cooling medium, water, for example, through line 50. Quenching to a temperature below that at which the water gas shift reaction takes place at an appreciable rate, e. g., below about 1500° F., is particularly useful in the generation of fuel gases of high heating value.

Residual char from the gasifier may be taken through line 52 for fuel or other purposes or through line 53 into a supplemental gas generator 54. Gas generator 54 may be, for example, a generator of the burner type wherein the residual char is reacted with a suitable oxygen-containing gas introduced through line 56. The gas from the generator 54 is passed through line 57 from which it may join the product gas stream or be separately treated. The residual solid or ash is discharged from the generator through line 58.

It will be obvious that various economies may be effected by utilizing heat contents of the various streams of gases and solids. This may be accomplished, for example, by passing hot streams in heat exchange with cooler streams or by utilizing heat from the hot gases or solids to generate steam. These various expedients are known in the art and may be resorted to without departing from the scope of the invention.

The process of the invention is particularly advantageous for the generation of fuel gas or synthesis gas from coal in that it provides flexible operating characteristics such that the temperature of the gasifier and the carbonizer may be at any desired value and substantially independent of one another. Normally, where hot gases from the gasification zone are utilized as a source of heat for the carbonization zone, the gasification apparatus is relatively inflexible, the temperature of the two zones being interdependent, as determined by the composition of the coal, the rate of feed of the coal, rate of feed and concentration of the oxygen, steam, etc.

Heat may be supplied to the carbonizer from the gasifier by way of hot gases or hot carbonaceous solid particles, or both, passing from the gasifier to the carbonizer. The quantity of heated solid particles passing from the gasifier to the carbonizer may be varied widely depending upon conditions of operation. If the gasifier is operated with a bed level below the top of the vessel, only those particles actually carried in the gas stream in the gasifier are passed with the effluent gas into the carbonizer. On the other hand, when the gasifier is operated full, fluidized particles from the gasifier are entrained in the stream of relatively high velocity gases leaving the gasifier through the restricted transfer line, which also serves as a mixing zone for the fresh feed. All of the carbonized particles admixed with fresh feed may be supplied from the gasifier.

Heat for the carbonization step is derived from

the materials, i. e., gases and any entrained solids, from the gasification zone. Heat is supplied to the carbonizer by heat transfer between these materials and the carbonaceous feed. Also, in many instances, heat may be supplied the carbonizer as a result of exothermic reactions of the gases. The formation of methane sometimes takes place by the interaction of carbon monoxide and hydrogen or by direct hydrogenation of carbon in the carbonization zone. Under these conditions exothermic heat of reaction is released in the gasifier. Carbon which may be incidentally formed by reversal of the producer gas reaction or by the cracking of hydrocarbons is carried by the carbonized solids to the gasification zone where it is regasified. Methane formation in the carbonization zone is influenced considerably by the composition of the carbonaceous feed material. Components of the ash, particularly iron, tend to catalyze the methanation reaction while sulfur compounds in the gas stream tend to deactivate the catalysts. The type of char made in the carbonizer also influences methane formation, lignite char being particularly active in this respect and the cokes of coking coals being relatively inactive.

It will be evident from a consideration of the foregoing that the present invention provides utmost flexibility of operating conditions. The temperatures of operation of the gasification and carbonization zones are almost completely independent of one another. Furthermore, no complex arrangement of vessels and solid transfer equipment is required to obtain this important advantage. By reason of this flexibility any desired type of gas: city gas, synthesis gas or pipe line gas may be produced with high efficiency.

The present process provides for passing any required quantity of the product gas from the gasifier to the carbonizer, for passing any desired quantity of carbonaceous material from the gasifier to the carbonizer, and for removal of excess heat, if necessary, from the carbonizer. Thus, a variety of feed materials may be handled. For those feed materials which tend to catalyze the disproportionation of carbon monoxide and formation of methane, a minimum amount of heat may be supplied the carbonizer by gases, or solids, or both, from the gasifier and the temperature controlled by the heat exchanger. For those feed materials which have large heat requirements to effect carbonization, such as those having high moisture contents or large percentages of volatile constituents or those which exhibit little or no tendency to catalyze exothermic reactions like the methanation reaction, heat requirements may be readily satisfied by passing the required quantity of hot solids, as well as gases, from the gasifier to the carbonizer. Thus the temperature of each zone may be controlled independently of the other.

Gasification with oxygen and steam in the gasification zone preferably is carried out at a temperature within the range of about 1600 to 2000° F. and at a pressure within the range of from about atmospheric to 600 pounds per square inch gauge, or even higher. Generally a pressure within the range of from about 200 to about 500 pounds per square inch is preferred. The carbonization may be carried out at temperatures within the range of from about 900° F. to about 1300° F. and at pressures comparable to those of the gasification zone. Pressures and tempera-

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tures may vary somewhat from these ranges and are not limited thereto.

The following are examples of the application of the process of my invention to the gasification of coal.

Example I

A coal, crushed to a particle size such that 85 per cent passes a 100 mesh screen, is maintained in a feed hopper under a pressure of 350 pounds per square inch gauge and fed at the rate of 10.5 tons per hour into gasification equipment similar to that illustrated in Fig. 1. Char from the carbonization zone at 1250° F. is recycled at a rate of 52.5 tons per hour to a mixing zone where it is admixed with hot gases at 2000° F. from the gasification zone; the resulting mixture is admixed with the incoming coal and passed to the carbonization zone. The gasification zone is operated at a pressure of 350 pounds per square inch gauge and the carbonization zone at a pressure of about 345 pounds per square inch gauge.

Char from the carbonization zone at a rate of 7.3 tons per hour is fed to the gasification zone in admixture with 1315 pounds of steam per hour at 375 pounds per square inch gauge and 1000° F. In addition, 4460 pounds of steam per hour in admixture with 225,000 standard cubic feet of oxygen per hour at 300° F. is fed into the gasification zone. The gasification zone is operated with a superficial gas velocity of about 0.6 foot per second and a bed density of about 14 pounds per cubic foot. The velocity in the mixing zone is 25 feet per second. The carbonization zone is operated with a superficial gas velocity of 0.6 foot per second and a bed density of about 14 pounds per cubic foot.

A 40 per cent carbon char is taken from the gasification zone at the rate of 2.67 tons per hour, and utilized as fuel for production of steam requirements for the plant. 885,000 standard cubic feet per hour of product gas is produced. The product gas may be converted to synthesis feed gas by suitable treatment, for example, removal of tar and oil, conversion of hydrocarbon gases to carbon monoxide and hydrogen, conversion of carbon monoxide and steam to hydrogen and carbon dioxide, and separation of carbon dioxide and sulfur.

Example II

Gasification of coal at 1650° F. and 300 pounds per square inch gauge is carried out in gasification equipment similar to that illustrated in Fig. 2.

Coking coal of high volatile content which yields a relatively inactive char is charged to the gasifier at the rate of 10 tons per hour by suspension in a stream of recycle gas preheated to 900° F. The recycle gas stream is essentially a mixture of carbon monoxide, hydrogen, and light hydrocarbons obtained as a residual gas after separation of tar, oil, water, and carbon dioxide from the product gases. Only sufficient recycle gas is used as is required to transport the feed and recycled char into the carbonizer.

The carbonizer is operated at 1250° F. and 280 pounds per square inch gauge. Product gases and hot char are passed from the gasifier to the carbonizer. All of the product gas passes through the carbonizer. Just sufficient carbonaceous material (or char) is passed from the gasifier to the carbonizer to satisfy the heat requirements in the carbonizer. Both the gasifier and carbonizer are operated under fluidized conditions and fluid

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full. The carry over from the gasifier to the carbonizer and from the carbonizer to the separator is thus dependent upon the rate of feed to these zones.

About 100 tons per hour of char is supplied to the carbonizer for admixture with the fresh feed. Of this amount about 58 tons is recycled to the carbonizer from the separator while about 42 tons is passed from the gasifier to the carbonizer.

About 108 tons per hour of carbonized material passes overhead from the carbonizer to the separator. From the separator, the carbonaceous material is returned to the gasification apparatus. One stream of carbonaceous material is sent to the carbonizer as the recycle char for admixture with fresh feed and a second is sent to the gasifier.

About 1,100 pounds per hour is passed to an auxiliary generator operated at 2300° F. for carbon cleanup and ash disposal. The carbon content of the carbonized material from the separator is about 50 per cent by weight.

The product gas is processed for the removal of water vapors, condensible oils and tar, and carbon dioxide. The resulting gas has a total heating value of about 410 B. t. u. per cubic foot and the following approximate analysis:

	Volume per cent
Hydrogen.....	35.2
Carbon monoxide.....	52.7
Methane.....	11.0
Nitrogen.....	0.4
Illuminants.....	0.6

Gas is produced at the rate of about 570,000 standard cubic feet per hour. About 48,000 pounds per day of light oil and tar are separated from the product gas stream. Steam at 900° F. is supplied to the gasification steps of the process at the rate of about 250,000 standard cubic feet per hour. Oxygen at 600° F. is supplied at the rate of about 134,000 standard cubic feet per hour.

Obviously many modifications and variations of the invention as hereinabove set forth may be made without departing from the spirit and scope thereof and therefore only such limitations should be imposed as are indicated in the appended claims.

I claim:

1. A process for the carbonization and gasification of solid carbonaceous material containing volatilizable constituents which comprises maintaining a dense phase fluidized bed of particles of solid carbonaceous material in a carbonization zone wherein volatilizable constituents are distilled from said particles producing carbonized material, passing carbonized material from said carbonization zone to a separate gasification zone, subjecting said carbonized material to reaction with oxygen and steam in said gasification zone to produce a primary gas consisting essentially of carbon monoxide and hydrogen, passing a stream of said primary gas through an intermediate zone of restricted cross-sectional area directly into the lower portion of said carbonization zone thereby effecting fluidization of the carbonaceous material therein and distillation of said volatilizable constituents, introducing particles of fresh carbonaceous material containing volatilizable constituents into said stream of primary gas in said intermediate zone, introducing particles of carbonized material substantially free from volatilizable constituents from said carbonization zone directly into said stream of primary gas in said intermediate zone upstream

from the point of introduction of said fresh carbonaceous material and in controlled amounts whereby at least 3 parts by weight of carbonized material is added per part of said fresh carbonaceous material, maintaining the velocity of said stream of primary gas through said intermediate zone at a velocity sufficient to entrain said particles of said fresh and carbonized material therein, and discharging from said carbonization zone the resulting gas containing volatilized constituents and products of reaction.

2. A process as defined in claim 1 wherein said dense phase fluidized bed of solid particles maintained in said carbonization zone substantially completely fills said carbonization zone whereby particles of carbonized material are entrained in gases discharged from said carbonization zone, carbonized material separated from gases discharged from said carbonization zone, a portion of the separated carbonized material passed directly to said intermediate zone, and a further

portion of the separated carbonized material passed to said gasification zone.

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