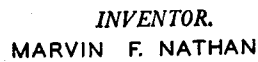


2,844,526

Filed May 16, 1956



G.H. Palmer
V.F. Davies

ATTORNEYS

1

2,844,526

METHOD FOR CARBONIZING CARBONACEOUS MATERIALS

Marvin F. Nathan, New York, N. Y., assignor to The M. W. Kellogg Company, Jersey City, N. J., a corporation of Delaware

Application May 16, 1956, Serial No. 585,334

7 Claims. (Cl. 202—27)

This invention relates to the treating of solid materials and more particularly to a method for the fluidized treatment of carbonaceous materials such as coal, shale, lignite, oil sands, etc., at low temperatures. Still more particularly, it relates to an improved method for recovering solids entrained in fluidizing gases when carbonizing fluidized carbonaceous materials at low temperatures.

This application is a continuation-in-part of my co-pending application Serial No. 517,472, filed June 23, 1955.

The treatment of carbonaceous solids to form valuable liquid, gaseous and solid products is well known in the art. An example of one process frequently employed entails the treatment of solids, such as coal at elevated temperatures whereby volatile materials are released from the solids and a valuable solid residue is formed. It has been the practice in the past to carry out carbonization in both non-fluid and fluid systems; however, the present invention is concerned with a carbonization process of the fluid type wherein the various steps are performed with a finely divided feed material which is maintained in a highly turbulent state of agitation by the passage through of a fluidizing medium.

In carrying out fluidized carbonization of carbonaceous materials, it has been found that several process steps are necessary in order to provide a workable operation and assure a maximum yield of desirable vapor, liquid and solid products. More usually, the first step in the carbonization process concerns the proper preparation of the raw feed material. This involves not only proper selection and sizing of the carbonaceous solids to provide a readily fluidizable feed, but also includes drying the solids to a minimum moisture content prior to further processing.

It has been found that certain finely divided carbonaceous materials, though easily fluidized at low temperatures, when subjected to more elevated temperatures suffer a change in physical characteristics and become soft and gummy and the particles tend to agglomerate. For example, many coals pass through a so-called "plastic" state, usually at a temperature between about 700° F. and about 800° F. wherein the coal particles tend to stick together and form large particles which resist fluidization. Several methods of combating this agglomerating tendency of coals and other carbonaceous materials have been suggested, one of the more successful of which comprises subjecting the finely divided solid particles to a mild oxidation treatment prior to further high temperature processing. It has been found that this method of treatment alters the physical characteristics of the carbonaceous material so as to minimize agglomeration of the solid particles. The changes which take place in the solids in a treatment of this type are not clearly understood; however, according to one theory the mild oxidation "case hardens" the particles thereby substantially nullifying their sticking tendencies when they pass through the plastic state. When processing carbonaceous materials, therefore, the step after the drying operation may

2

involve pretreating the dry solid particles at an elevated temperature in the presence of a limited amount of oxygen. During this process a portion of the vaporizable compounds present in the carbonaceous solids are released. The third and last step in the conventional solids carbonization process concerns the treatment of carbonaceous material at a still higher temperature whereby the remaining vaporizable compounds are released therefrom and a carbonaceous product residue is produced. The vaporizable portion of the coal which is commonly known as "tar" comprises numerous organic compounds having a wide range of boiling points. As used herein, the term "tar" includes any volatile organic compounds released from the coal, either liquid or vapor and either cracked or uncracked. The composition of the carbonaceous residue remaining after vaporization of the tar depends on the type of carbonaceous feed material used. For example, when carbonizing coal the residue material is commonly called "char." Although the description and discussion of the invention will be directed primarily to coal carbonization, the term "char" will be used hereinafter in a broader sense to designate any residue solids remaining after carbonization.

In addition to the three processing steps just described, a fourth operation may be necessary. In carrying out the drying step it is usually preferred to heat the carbonaceous feed only to the minimum temperature necessary to remove the surface water, which is, of course, the boiling point of water at the pressure conditions maintained during this operation. The pretreating operation, however, is carried out at an elevated temperature, substantially above the temperature at which the solids leave the drying zone. Only a portion of the heat required in this operation is supplied by the oxygen consumed therein. The remaining heat required is furnished by what may be called a preheating step. It is possible to provide the required preheat in conjunction with the drying operation or the pretreating operation, or it may be made an entirely separate and independent part of the carbonization process.

Each of the aforementioned processing steps, i. e., drying, preheating, pretreating and carbonization is customarily carried out in a separate vessel, which necessitates the use of numerous transfer lines and standpipes and involves an extensive solids recovery system when substantially all of the solids entering the system are to be recovered.

It is an object of this invention to provide an improved method and means for carrying out a solids carbonization process.

Another object of this invention is to provide improved method and means for increasing product yield in the carbonization of carbonaceous materials.

It is still another object of this invention to provide improved method and apparatus for recovering entrained solids in the successive steps of fluidized drying and preheating of carbonaceous materials.

Still another object of this invention is to recover solids entrained in the drying of fluidized carbonaceous solids.

These and other objects of the invention will become more apparent from the following detailed description and discussion.

In the method of this invention the aforementioned objects are achieved by combining gases containing entrained carbonaceous solids and gases containing entrained char particles and recovering the combined solids as a product by a scrubbing operation. Specifically the carbonaceous solids are entrained in gases released from a fluidized drying and preheating treatment of carbonaceous solids and the char particles are entrained in a gas obtained from a preliminary solids recovery system

for the treatment of char produced in the carbonization of carbonaceous solids.

It is within the scope of this invention to treat various carbonaceous materials in the manner described, including coals, shales, lignites, asphalts, oil sands, etc. The invention is particularly exemplified by its application to the treatment of coal, and further discussion of the invention is directed to the use of this material. It is not intended, however, that this particular application should limit the scope of the invention in any way.

Usually the first step to be considered in a process for the carbonization of coal is concerned with surface water present in the coal feed which may, unless removed, prevent fluidization of the coal. One of the problems encountered when handling carbonaceous materials such as coal in a fluidized system results from the tendency of the finely divided solids to agglomerate because of water condensed thereon. Most coals coming from a treating plant, for example, have a relatively high surface or "free" water content, usually between about 2 and about 15 percent by weight, or higher. Unless removed, this moisture causes the finely divided coal particles to stick together and resist fluidization. Even after fluidization is achieved, moisture may cause packing or bridging in process equipment of restricted cross section, such as, for example in feed hoppers, standpipes, etc. It is not always economically feasible to remove all the moisture from the coal; however, it has been found that agglomeration and packing of coal particles due to the presence of water is minimized if between about 50 percent and about 90 percent of the water initially present is removed.

Several methods may be used for drying coal, such as direct contact with air or other gases at high temperatures, heating the coal by indirect heat exchange with a heated fluid, etc. In the latter method the heat transfer may be effected within a dense phase bed of the solids to be dried, or solids may be withdrawn from such a bed, passed through a heat exchanger and returned to the bed at an elevated temperature. Whichever method is used, a substantial quantity of gases result and these gases upon leaving the dense solids phase must be treated for the removal and recovery of entrained coal solids. Conventionally, gases from a fluid bed are passed through cyclones. However, this provides only a partial recovery of solids and the gases from this treatment may contain from about 0.2 to about 100 grains per cubic foot of solids.

A similar problem of solids recovery may exist in other phases of the coal carbonization process. For example, if a preheating step is provided prior to carbonization, gases from this operation will also contain entrained coal solids. In the carbonization step itself and the pretreating operation, if one is provided, a different situation arises in that the gases contain large quantities of volatile compounds released from the coal. This presents a special problem which is not the concern of the invention herein disclosed.

The recovery of char after quenching, presents a similar problem to that encountered when drying the coal. Thus gases leaving the preliminary char recovery system will contain a quantity of residual solids.

In the method of this invention recovery of residual entrained coal solids and char particles is affected in a common water scrubbing system. The two gas streams may be introduced separately to the scrubbing system; however, more usually, it is preferred to combine the gases beforehand. In the scrubbing step, the gases are contacted with a liquid such as water which is preferably introduced in such a manner as to provide countercurrent contact with the gases. The solids removed during this operation are present as a slurry and after removal from the scrubbing system will combine with the bulk of the char product. The amount of coal recovered in the scrubbing system is very small compared to the total

char product and usually amounts to less than about 0.8 percent thereof by weight. As a result, the properties and physical characteristics of the char are substantially unchanged by the coal present therein.

In a preferred embodiment of this invention coal is treated in a series of steps which involve drying, preheating, pretreating and low temperature carbonization. In carrying out the drying operation, raw coal suitably subdivided for fluidization, that is, of a size between about 10 mesh and about 5 microns is introduced into a first zone wherein it is commingled with dry heated coal in sufficient quantity to elevate the entire mass of coal to a temperature suitable to effect the removal of water. The dry coal is then passed through a heater where it is further elevated in temperature by indirect heat exchange with a hot fluid and then into a second zone. The higher temperature coal in the second zone serves as the source of the coal commingled with the wet coal feed, and in addition, provides preheated coal for the next phase of the carbonization process. The entire drying and preheating step is conveniently conducted in a fluid system with both the low and high temperature zones containing a dense phase bed of fluidized coal superposed by a dilute phase of low solids concentration. Adequate turbulence to maintain each dense phase bed is provided by maintaining a linear gas velocity therein between about 0.5 and about 5 feet per second, or more usually between about 0.75 and about 3 feet per second. Under normal operating conditions the density of the beds thus provided varies between about 10 and about 40 pounds per cubic foot. The temperatures in the two zones may vary, depending on the residence time of the coal in each zone and the moisture content of the raw coal feed. However, usually the first zone is operated at a temperature between about 220° F. and about 325° F., and the second zone is preferably maintained at a temperature of between about 350° F. and about 600° F. Fluidization of the solids in the low temperature zone is partially provided by moisture released from the coal and may be augmented by the introduction into this zone of air or an inert gas such as, for example flue gas, steam, etc. The coal in the high temperature or preheating zone is maintained in a fluid state by the introduction of a similar gasifying medium. It is necessary to circulate a sufficient amount of coal from the low temperature zone through the heater to the high temperature zone and back to the low temperature zone to provide both the sensible heat acquired by the dry solids and the heat of vaporization of the water released therefrom. When operating in accordance with the zonal temperature ranges given, the amount of coal circulated relative to the raw coal feed rate is between about 2 and about 5 pounds per pound.

The heat transfer surface required for drying and preheating the coal is preferably provided by a conventional shell and tube heat exchanger with the solids being passed through the tubes in indirect heat exchange with a hot fluid passed through the exchanger shell. The heat required to dry the coal is provided by a fluid heating medium which may be a petroleum oil or vapor, or mixtures thereof, or other liquid or vapor material which is easily transported and can withstand relatively high temperatures. In general, liquid heating fluids are more satisfactory than gases because of their high specific heats and low volume relative to gases. Examples of suitable heating fluids are residual petroleum oils, synthetic heat transfer liquids, inorganic salt mixtures, lead, mercury, etc. The temperature at which the heating medium is employed varies with the temperature maintained in the drying zone and with the heat transfer characteristics of the heating medium. Usually, it is preferred to introduce the heating medium at a temperature between about 350° F. and about 1000° F. Temperatures greater than this are not desirable because

of the danger of over heating coal particles in contact with the heat transfer surface.

The amount of heat exchange surface required to carry out the drying and preheating operations varies depending on several factors including the quantity of coal to be heated, the amount of moisture in the coal, heat transfer coefficients, etc. More usually a surface area between about 0.02 and about 0.30 square foot per pound of fresh coal feed per hour is sufficient to provide the desired drying and preheat.

The gases leaving the dense solids phase of the drying zone comprise a mixture of steam and fluidizing gas and contain a quantity of entrained dry coal solids. Removal of the major portion of the solids from this gas is effected in a conventional cyclone recovery system. However the gases leaving the dryer still contain some solids, more usually between about 0.2 and about 100 grains per cubic feet of gas.

After leaving the preheating zone, the coal is passed into a pretreating zone wherein it is contacted with air or other oxygen containing gas and partially burned to provide the pretreating and case hardening effect previously discussed. The temperature at which this process step is carried out may vary over a range between about 600° F. and about 825° F.; however, more usually it is preferred to pretreat the coal in a more narrow range of temperature, that is between about 650° F. and about 800° F. As in the previous operations, the coal pretreatment is carried out in a conventional dense phase fluidized bed, wherein the coal is maintained in a turbulent fluid state by passage therethrough of a gasiform medium. Adequate turbulence to maintain the dense phase bed is provided by maintaining a linear gas velocity therein between about 0.5 and about 5 feet per second. Under normal operating conditions, the density of the dense phase bed thus provided varies between about 10 and about 40 pounds per cubic foot. Generally, a portion or all of the fluidizing medium is supplied in conjunction with the oxygen required for pretreating. This may be accomplished by diluting the oxygen with air, by using air alone or by diluting air or oxygen with steam or other inert gas. The amount of oxygen required for pretreating is usually between about 0.02 and about 0.08 pound per pound of dry coal feed. To provide sufficient time for the pretreating combustion reactions to take place, the rate of introduction of coal to the pretreating zone is adjusted to allow an average particle residence time therein of between about 10 and about 60 minutes.

Upon entering the pretreating zone, dry preheated coal at a relatively low temperature becomes intimately mixed with higher temperature pretreated coal and is swiftly elevated to the temperature level prevailing in this zone. As the temperature of the dry coal is increased, a portion of the lower boiling tar components present in the coal are vaporized and passed into the fluidization and combustion gases. Since oxygen is relatively non-selective in its action, this phase of the carbonization process may involve the consumption of a portion of the tar. For this reason, it is desirable to limit the introduction of oxygen to the pretreating zone to the minimum amount necessary to prevent agglomeration of the solids and maintain an operable system.

Following pretreating, the coal is passed into a carbonization zone wherein the major portion of the volatile components in the coal are removed and a valuable residue char is formed. This, the major step of the process, as far as product formation is concerned, is also conveniently carried out in a dense phase fluidized bed similar to the drying, preheating and pretreating beds previously described. In order to effect removal of the volatile coal components, a large amount of heat must be introduced to the carbonization zone. Conventionally, this heat may be supplied from one or more of several sources, for example it may be provided in

an inert gas such as a fuel gas heated to a high temperature, or it may be supplied from a combustible gas such as fuel gas mixed with oxygen or it may be furnished from the combustion of oxygen or an oxygen containing gas with a portion of the carbonaceous feed. When heat is provided by burning either fuel gas or coal, the gasiform fluidizing medium required to maintain the dense phase in the carbonization zone is generally furnished by the combustion gases. If necessary, however, deficiencies in the quantity of fluidizing medium may be made up by the introduction into the carbonization zone of a flue gas, steam or other extraneous inert gas.

The carbonization of coal to remove distillable tars therefrom and produce a char residue product is conducted over a wide range of temperatures usually between about 700° F. and about 2400° F. The preferred thermal range of operation is determined to a great extent by the type of liquid product desired; for example, when it is preferred to distill the coal tars with a minimum of cracking of volatile constituents, namely low temperature carbonization, the temperature is held to a minimum of about 700° F. and not more than about 1000° F. The type of coal is also of importance in establishing the operating temperature since some coals are more difficult to distill than others. The carbonization zone contains a dense phase bed superposed by a disperse or dilute phase which may have a solids concentration as low as 0.001 pound per cubic foot. Gases from the dense phase zone pass into the dilute phase which provides a preliminary rough separation of vapors and solids. Further solids separation is provided by conventional means, such as, for example cyclones, filters, etc.

Substantially all of the desirable constituents of coal are removed at the aforementioned carbonization temperatures within a very short period of time, that is between about 0.25 and about 10 minutes. As a further precaution to prevent agglomeration of the coal particles in the carbonizing zone, it is preferred to maintain a substantial ratio of char to fresh feed therein. This serves to dilute the fresh pretreated coal, which provides the desired beneficial effect; however, it also makes it necessary to substantially increase the coal residence time. At the usual char to fresh feed ratios maintained in the carbonization zone, that is between about 5 pounds per pound and about 50 pounds per pound, the particle residence time therein is between about 2 minutes and about 200 minutes, more usually between about 20 minutes and about 100 minutes.

Carbonization may be carried out over a wide range of pressures; however, the pressure is usually maintained between atmospheric and 500 p. s. i. g., preferably between about atmospheric and about 100 p. s. i. g. Since a driving force is necessary for the passage of coal from the pretreating zone into the carbonization zone the pretreating zone must operate at a pressure above the pressure in the carbonization zone; more usually the differential pressure between the two zones is between about ½ and about 2 p. s. i. By virtue of its physical location above the pretreating zone, the drying zone may operate at a pressure either higher or lower than the pressure in the former zone. More usually, it is convenient to maintain the pressure in the drying zone lower than the pressure in the pretreating zone and as a result the drying zone is ordinarily operated at between about 1 and about 20 p. s. i. less than the pretreating zone.

Hot char product from which the major portion of the volatile constituents of the coal have been removed is withdrawn from the lower portion of the carbonizer and is passed through a cooler wherein the temperature of the char is lowered by indirect heat exchange with a fluid cooling medium. When operating in accordance with the ranges of process variables previously enumerated the amount of this material varies between about 0.6 and about 0.9 pound per pound of wet feed coal. The

remainder of the raw material delivered to the process is now in a vapor state, comprising a mixture of steam, combustion gases and tar vapors. The apparatus used in conjunction with the char cooling preferably comprises one or more conventional tubular heat exchangers similar to those previously described in conjunction with drying and preheating the coal feed. The type and quantity of cooling fluid passed through the exchanger may be varied to meet the particular needs of the process. In general, fluids similar to those previously disclosed for use in drying and preheating the coal are used. This operation is simplified and the cost is substantially reduced, if a common fluid medium is used for both coal drying and preheating, and for cooling the product char. When operating with this type of system, a continuous circulating fluid stream is provided, which extracts heat from the hot char product and transfers it to the fresh coal feed. Inasmuch as the heat removed from the char in the cooling operation may not be sufficient to provide the heat required for drying and preheating the coal feed, or vice versa, it is desirable when using a common heat exchange fluid to provide an additional heat source, such as for example a conventional tubular heater, or an additional source of cooling, such as for example a water cooler, whichever is required.

In this preliminary cooling step, the char temperature is usually reduced to between about 700° F. and about 400° F., although it may be brought to a still lower temperature if desired. The cooling fluid may be introduced to the cooler at any low temperature; however, when a common circulating stream is used the inlet temperature, of necessity, conforms to the temperature of the fluid leaving the heaters which serve the drying and preheating stages of the carbonization process, i. e. between about 650° F. and about 350° F. The size of the cooler required varies with the amount and temperature of the char product, the heat transfer coefficients of the flowing streams and other operating variables; however, more usually a surface area between about 0.01 and about 0.10 square foot per pound of char product per hour is adequate to provide the desired cooling.

Normally, only a portion of the heat contained in the product char can be removed economically by indirect cooling, particularly when using a common circulating heat exchange fluid. To further cool the char and provide a more easily handled product, water is injected into the partially cooled fluidized char which is then passed into a receiver or char hopper. The quantity of water used for this purpose may vary; however, usually it is preferred to limit it to not more than the amount necessary to cool the char to the dew point of water at the pressure existing in the receiver, thus converting the entire quantity of cooling water to steam. By operating in this manner, advantage is taken of the high vaporization heat of water to provide maximum cooling with a minimum of water consumption and at the same time provide additional vapors to maintain the char in the hopper in a fluidized state. The cooled product is then conveniently removed from the hopper, defluidized by contact with additional water which condenses the fluidizing steam and is passed from the system by means of a conveyor or other suitable means.

The water used to cool the hot char may be introduced thereto prior to entry of this material into the char hopper, or after the char enters the hopper, or a portion may be admitted at both localities. It is preferred that the char be cooled to as low a temperature as possible; however, if a suitable use for higher temperature steam exists, the amount of cooling water may be controlled to provide a char temperature in the receiver substantially above the dew point of water. Also, although it is preferred to maintain the char in the hopper in a fluidized state, the defluidization of this material may be accomplished therein by increasing the amount of cooling water introduced into the char to the point where liquid water is present in the hopper. The char is then

removed from this vessel as a slurry rather than as a fluidized mass.

The amount of water required to accomplish the second stage of the char cooling process varies with the initial temperatures of both the char product and the water and the final temperature of the char. More usually the water is introduced at a low temperature, i. e. between about 60° F. and about 100° F. The pressure in the char hopper or receiver is also desirably maintained at a low level, usually less than the pressure in the carbonizer, viz. between about 0 and about 5 p. s. i. g. Setting the pressure establishes the dew point temperature and accordingly the amount of water required as quench, which is usually between about 0.05 and about 0.15 pound per pound of char product.

The cool char which accumulates in the char hopper forms a dense fluidized solids bed above which there exists a conventional dilute phase zone of low solids concentration. The solids density in the dense phase bed is usually between about 15 and about 25 pounds per cubic foot; whereas, the concentration of solids in the dilute phase is very small, often less than 0.1 pound per cubic foot. Vapors and solids leaving the hopper dilute phase pass through conventional separation means, for example cyclones, for the removal of a major portion of the solids. The char solids remaining in the gases, usually between about 0.2 and about 100 grains per cubic foot of gas are recovered in a secondary solids recovery system. In order to minimize the facilities required for separating entrained solids, the overhead gases from the feed coal drier and preheater, which as previously noted contain entrained coal particles, are also introduced to the secondary solids recovery system.

In one embodiment, this system comprises a vertical elongated scrubbing tower, with baffles suitably dispersed therein to provide good liquid-vapor contact. Within this tower, the combined vapors from the drier and preheater and char hopper are scrubbed with water to remove entrained coal and char particles. As in the preceding cooling step, the quantity and temperature of the scrubbing water is controlled to maintain a suitable temperature within the scrubber so that a minimum amount of the steam introduced in the two vapor streams is condensed. Preferably the scrubbing liquid is supplied by recycling a warm solids-water slurry from the bottom of the scrubber and combining with this stream necessary makeup water from an outside source. Since the solids-water slurry is at the dew point temperature of the steam in the scrubber, usually between about 212° F. and about 240° F., this method of operation provides a relatively high temperature scrubbing stream and a minimum of steam is condensed in the process. In addition, by recycling, it is possible to closely control the scrubbing operation for maximum solids removal.

The recovered solids are removed from the scrubbing system as a slurry in the scrubbing water. This slurry is conveniently mixed with char removed from the char hopper in order to lower its temperature and to reduce the dust problem associated with the finely divided solids product. As a result of this, the product char solids contain a mixture of coal and char; however, the amount of coal recovered in this operation is insignificant when compared to the char, being only between about 0.1 and about 0.8 percent thereof by weight, and when combined with the char is insufficient in quantity to alter its properties or characteristics.

It is apparent that the aforescribed method of solids recovery offers several important advantages. The combination treatment of gases from the char hopper and the drying and preheating zones substantially reduces the number of cyclones or other solids recovery equipment required. In addition, controlling the scrubbing operation to prevent condensation of the fluidizing steam provides an important heat economy and reduces the amount of scrubbing water required for the operation. Further-

more, introducing recovered coal into the char product not only provides a convenient method of disposing of this material but also increases the char yield without affecting the properties of the char.

As previously mentioned, this invention is not limited in its scope to the treatment of coal, but encompasses the use of other carbonaceous feed materials, for example shales, asphalt, oil sands, etc. Similar processing considerations are important and similar operations are required when carbonizing these feed materials other than coal. The conditions appropriate for each specific feed material are well known to those skilled in the art and for this reason do not need repeating here.

As previously mentioned, the effluent vapors from the carbonizer comprise gaseous products of combustion and various tar compounds plus a small amount of entrained char. The major portion of the tar materials in the gases condense to liquids at ordinary temperatures and form a valuable product of the carbonization process. To effect the separation of the normally liquid tar, the carbonizer gas stream is passed to a quench tower where the vapors are contacted with a low temperature liquid tar. This material not only provides the cooling effect necessary to condense liquid tars but also effects the removal of entrained solids from the gases. The scrubbing and condensing liquid is preferably obtained by circulating tar condensed in the quench tower through a cooler and recycling it to the upper portion of the tower. Within the tower are provided suitable baffles or plates whereby intimate contact between ascending gases and downflowing liquid is effected. The pressure at which this operation is carried out is controlled by the pressure in the carbonization zone, being somewhat lower, usually between about 10 and about 2 p. s. i. g. It has been found that the major portion of the desirable liquid tar compounds are condensed by cooling the carbonizer gases to between about 150° F. and about 80° F. The remaining vaporous tar compounds and combustion products form a gas, which although low in heat content, may be used as a fuel. If desired, of course, a further separation between the uncondensed tar compounds and combustion and fluidization gases may be effected.

In order to more clearly describe the invention and provide a better understanding thereof, reference is had to the accompanying drawing which is a diagrammatic illustration of process equipment used in carrying out a preferred embodiment of the invention, which equipment comprises a unitary coal carbonization system which includes a dryer, preheater, pretreater, carbonizer, char hopper, solids recovery system, tar quench tower and associated lines and heat exchange equipment.

Referring to the drawing, a finely subdivided coal at a temperature of about 60° F. having a particle size distribution between about 10 mesh and about 5 microns is delivered through conduit 2 into feed standpipe 4 wherein it is maintained in a dense turbulent state by passage therethrough of a fluidizing medium. Entering the system the coal contains about 8% of surface water by weight. From standpipe 4 the fluidized coal passes downwardly through conduit 6 where it is entrained in additional gases, in this instance steam, and passed upwardly through a conduit 8 to a drier and preheater vessel 10 which is at a substantially higher elevation. Within the drier vessel there is maintained a dense highly turbulent bed 12 of dry coal particles at a temperature of about 270° F. The upper portion of this bed occupies the entire cross section of the drier vessel 10; however, in the lower portion thereof, the dry coal is confined within an annular space lying between the walls of the drier and a cylindrical elongated conduit extending upwardly through the bottom of the drier. Within this conduit lies a preheating zone 14 in which there is maintained a higher temperature dense bed of coal particles which overflow continuously into the lower temperature dry

solids bed 12. Above the dense beds of dry and preheated coal is a dilute phase 16 of low solids concentration. Water vapors released from the coal pass upwardly through this space into a cyclone 18 from which separated solids are returned to the dense phase of dry coal, and from which the vapors containing unseparated solids leave the drier through conduit 20.

To provide the sensible heat required to heat the wet coal and the latent heat of vaporization of the water present therein, a stream of dry coal is removed from the annular drying zone 12 through conduit 22, entrained in fluidizing steam and passed upwardly through conduit 26 and coal heater 28 wherein the temperature of the coal is increased to about 480° F. From the heater the hot coal is passed through the conduit 34 into conduit 14 from which it eventually overflows to the drying zone. In order to maintain the desired temperature in the drying zone, it is necessary to overflow about 2 pounds of solids from conduit 14 per pound of wet coal introduced into the unit. Thus the solids circulation rate through the coal heater 28 is about 3 pounds of coal per pound of wet feed. The heat required in the combined drying and preheating operation is supplied at present by passing the circulating solids stream in indirect heat exchange with a suitable heat exchange medium. It is preferred to employ as the heat exchange medium a cat cracker decanted oil having an API gravity of about 15 which is heated by heat removed from the system in the manner described hereinafter. This material is introduced to heater 28 through conduit 30 at a temperature of about 680° F. and exits therefrom through conduit 32 at a temperature of about 400° F. The foregoing method of drying the coal is simple in application and in addition to effecting removal of moisture from the coal, it provides a ready means of adding to the coal the amount of preheat required before pretreating the coal, the next step in the process.

Although the hot coal leaving heater 28 enters zone 14 in a fluidized condition, it may be desirable to introduce additional gases, such as for example steam through conduit 13. Generally, the water vaporized in the drying zone is adequate to provide the desired turbulence in the dry solids bed; however, if necessary, an additional quantity of fluidizing gases may also be introduced to zone 12. The amount of fluidizing gases passed through each zone is controlled to provide a velocity therein of about 2 feet per second, thereby maintaining a solids density in each bed of about 25 pounds per cubic foot. As previously mentioned, the effluent gases from both zones pass through a conventional cyclone 18 for the separation of entrained solids which are returned to zone 12. In spite of this, some solids, in quantity equal to about 0.2 percent by weight of the wet feed are retained in the gases and leave the system through conduit 20.

The combined drying and preheating vessel 10 forms a part of a single unitary vessel structure being superposed above a carbonization vessel 36 which contains within its lower portion a pretreating zone 42. Passage of solids from the preheating zone 14 to the pretreating zone 42 is effected by flowing them downwardly through a standpipe 44 enclosed within the carbonizer vessel. Inasmuch as the standpipe passes through the carbonizer before it reaches the pretreating zone, it is exposed to the high temperatures in the former zone and it may be desirable to provide some form of insulation to protect this conduit. The rate of flow of solids from the preheating zone 14 to the pretreating zone 42 is controlled to maintain a more or less constant level in vessel 10 by a conventional plug valve 58 in contact with the bottom terminus of the standpipe 44. The pretreating zone 42 is separated in part from the carbonization zone 40 by a vertical baffle 66 attached at the bottom and sides to the inner wall of the carbonizer vessel 36. The bottom portion of the treating zone contains a distribution grid 60 for distributing fluidizing gases throughout the pretreated

coal. The pretreating zone opens upwardly into the carbonizing zone 40 and is separated therefrom by a grid 54 through which pretreated solids and vapors pass from the former to the latter zone.

The pretreating operation involves contacting the coal particles with a controlled amount of oxygen, viz., about 0.04 pound per pound of preheated coal, whereby the coal particles are partially oxidized. In this manner, the physical characteristics of the particles are altered so as to nullify their tendency to adhere to each other as they are elevated in temperature and pass through the so-called "plastic" stage. The effectiveness of the pretreating step is dependent not only on the extent to which the coal particles are oxidized, but is also a function of the pretreatment temperature, which is substantially increased over the preheating temperature, that is to about 725° F. The heat required to elevate the coal to this temperature is in normal operation supplied entirely from the heat of combustion of the coal. In carrying out the pretreating step, oxygen is introduced through conduit 64 and is distributed in the lower portion of the pretreating zone through grid 60. The oxygen may be supplied in a relatively pure state; however, more usually, it is preferred to use air, not only from the viewpoint of cost, but also to supply the additional gases necessary to maintain the solids in the pretreating zone in a fluidized state. Although the air admitted to the system normally suffices for this purpose additional gases such as, for example, steam, flue gas, etc., may be introduced through conduit 64 for fluidization purposes.

Coal entering the pretreating zone commingles with the solids contained therein and is partially oxidized and rapidly increased in temperature to that of the dense phase bed. In this process about 4 percent by weight of the preheated coal is reacted with the oxygen and converted to combustion products. The resulting mixture of pretreated coal and combustion gases, along with any portion of unconsumed oxygen, passes upwardly through the pretreating zone and through grid 54 into the carbonization zone 43. Within this zone there is maintained a dense phase turbulent bed of solid char particles at a substantially higher temperature, that is about 950° F. Inasmuch as the pretreating zone is entirely beneath the top level of the solids in the carbonization zone, the grid 54 serves the dual purpose of distributing the solids and gases leaving the pretreating zone and at the same time prevents passage of solids from the carbonization zone to the pretreating zone. By use of this separating means, it is possible to maintain two contiguous, yet distinct and separate dense phase beds of solids at quite different temperatures.

The preheated coal from zone 14 contains a large number of organic tar compounds varying widely in molecular structure and boiling point. The increase in temperature in this zone releases a portion of the lower boiling of these volatile compounds which pass upwardly into the carbonization zone 43 along with the pretreated solids and other gases. Upon entering the latter zone, the pretreated solids are quickly elevated to the temperature prevailing therein and large additional amounts of volatile components are released from the coal. The total time required in the two zones to carry out the process of tar removal is of short duration; however, in order to prevent solids from agglomerating and thereby assure an operable fluid process, it is desirable to maintain a large excess of pretreated solids in the pretreating zone and a similar excess of carbonized solids or char in the carbonization zone. This is effectively provided by sizing the pretreating and carbonization zones to allow an average particle residence therein of about 25 minutes and about 60 minutes, respectively. The pretreated solids bed is maintained in a highly turbulent state by controlling the flow of vapors therethrough to provide a gas velocity of about 1.2 feet per second and a solids density of about 25 pounds per cubic foot. Usually, this is effected by

varying the oxygen rate through conduit 64; however, if necessary, an extraneous gas (not shown) is admitted to zone 42. The degree of turbulence and density of the solids in zone 43 is regulated in a similar manner.

The heat required for carbonizing the coal feed is also supplied by burning a portion of this material. For this purpose, about 0.03 pound of oxygen per pound of pretreated coal feed is introduced into the carbonization zone 43. In this operation also the oxygen is introduced in the form of air rather than in pure state, for the reasons previously given. Since one of the important features in optimizing liquid product yield is minimum contact between oxygen and volatile tar constituents, the oxygen required for carbonization is introduced into the bottom of the carbonization zone through conduit 70 which is at a point remote from the area of introduction of pretreated coal into the same zone. Oxidation and combustion of the carbonized coal particles proceeds rapidly and is substantially completed before the carbonizer fluidizing and combustion gases reach the elevation at which the pretreated coal is present in quantity. The heat released by the combustion reactions is quickly transmitted throughout the dense char bed providing a hot turbulent mass into which lower temperature pretreated solids are introduced. The transfer of heat from the char particles to the pretreated solids in turn is equally swift and these solids reach the general char bed temperature level within a very short period of time. The process of devolatilization also proceeds at a fast rate and, by the time the pretreated solids reach the zone of combustion, they are substantially free of volatile tars.

By reason of the location of withdrawal conduit 62, char product from the main upper portion of the carbonization solids bed is forced to flow downwardly through the space provided between baffle 66 and the wall of the carbonizer vessel 36. Hot combustion and fluidizing gases flow upwardly through the same space countercurrent to the descending char and provide a stripping action which assists in the removal of tar compounds from the char. The removal of volatile components from the coal in the carbonization zone, therefore, is effected in two ways, i. e., by elevating the pretreated coal particles to the carbonization temperature and by passing these particles downwardly countercurrent to ascending combustion and fluidizing gases before withdrawing them from the carbonization zone. While increased temperature is the major factor in effecting tar removal, the stripping action of the combustion gases contributes to the total tar yield by removing some residual volatile materials.

The final products of the carbonization process comprise a mixture of tar vapors, steam and combustion gases, and carbonaceous char solids. Distribution of these products, based on the wet coal feed, is approximately 8 percent steam, 14 percent tar compounds and 76 percent char. The remainder of the coal is converted to combustion products to supply the process heat requirements. The gaseous products pass from the dense phase bed of char 43 upwardly into a dilute phase 47 and from thence through a cyclone separator 46 and conduit 52. Solids recovered in the cyclone are returned to the dense char bed below the surface thereof. Char solids product are removed from the bottom of the carbonizer 36 through conduit 62, are picked up by a stream of fluidizing steam and are passed through conduit 72 upwardly through a char cooler 74 for preliminarily cooling. The fluidized char solids enter the cooler at a temperature substantially the same as that maintained in the carbonization zone, i. e., about 925° F., and exit from the cooler at a temperature of about 500° F. To extract the heat from the char, a cat cracker decanted oil of about 15 API gravity is introduced into the cooler 74 through conduit 73 at a temperature of about 400° F. This material flows through the cooler 74 countercurrent to the char and exists therefrom through conduit 75, being heated in its passage

through the cooler to about 600° F. To provide a process of maximum thermal efficiency, a continuous circulating fluid system (not shown) is used in which a common hydrocarbon fluid accomplishes both char cooling and the drying and preheating of the coal feed. Substantially more heat is required in the drying and preheating operation than is obtained by cooling the char. Therefore, in order to thermally balance the system, it is necessary to supply an additional amount of heat to the oil prior to its introduction into the coal heater 28. This may be done in any conventional manner, such as, for example, by passing the decanted oil through a conventional fired heater (not shown) or other conventional heating means.

The lower temperature char leaving cooler 74 is passed into a char pot 78 from which it flows downwardly through conduit 80 into a char hopper 84 where it accumulates in a conventional dense phase fluidized bed 88, superposed by a dilute phase 86. Although a substantial amount of heat is removed from the char in the cooler 74, it is still much too hot to be yielded as product. It is preferable, for convenience in handling the char, that it be cooled to a much lower temperature and, if possible, by a more efficient method than indirect heat exchange. The large amount of additional cooling required is conveniently and economically furnished by introducing water into the char through conduit 82 prior to passage of the char into the char hopper 84. The water is immediately converted to steam, thus providing, in addition to the cooling effect, additional fluidizing medium suitable for maintaining the solids in conduit 80 in a turbulent state. The amount of water combined with the char is controlled to provide a temperature in the char hopper at or slightly above the dew point of water at the pressure existing therein. In this specific illustration, the hopper pressure is about 3.5 p. s. i. g. and the temperature of the dense char bed 88 is about 230° F. These conditions are maintained by cooling the char with about 0.07 pound of 80° F. water per pound of char. Operating in this manner prevents liquid water from passing into the hopper, and the solids contained therein are readily maintained in a fluid state.

Steam which results from the char cooling disengages from the solids in bed 88, passes upwardly through dilute phase 86 and a conventional cyclone separator 90 for the removal of entrained solids, and thence through conduit 108 into a secondary solids recovery tower 98. Before entering this tower, the char hopper gases are joined through conduit 20 by the gaseous effluent from the drier and preheater 10. Within the solids recovery tower 98, which contains a number of baffles 106, the combined gases containing both char and coal solids are scrubbed with water introduced through conduit 100, and spraying 104. The resulting solids-water slurry is withdrawn from the bottom of the recovery tower through conduit 110, is diluted with additional water from conduit 94 and then combined with char removed from the bottom of the char hopper through conduit 92. The slurry water serves to condense any steam remaining in the char released from the hopper, thereby defluidizing this material. The total solids product comprising char admixed with a small amount of coal is then removed from the unit by a conveyor or by other suitable means (not shown).

The temperature in the solids recovery tower is about 216° F., which, at the pressure existing therein, that is about 2 p. s. i. g., is equal to the dew point of water. It is preferred in carrying out the solids recovery process that a minimum amount of the steam introduced to tower 98 be condensed. In order to assure this result, the temperature of the scrubbing water is maintained at substantially the same level as the temperature within the tower. This is conveniently accomplished by heating the water prior to its introduction to the recovery tower, or more preferably by recycling hot slurry from conduit 110 to the top of the recovery tower (not shown). Even when using recycle slurry for scrubbing, however, it is neces-

sary to introduce extraneous warm make-up water through conduit 100 to compensate for water in the slurry combined with the char product. The scrubbed gases, consisting of essentially solids-free steam, accumulate in the upper portion of tower 98 and are removed therefrom through conduit 102. This gas, although low in pressure and temperature, contains a large amount of latent heat and may be used in any conventional service where low pressure steam is of value.

Tar vapors formed in the pretreating and carbonization zones, together with the gaseous products of combustion, pass from the carbonizer 36 through conduit 52 and are introduced into a tar quench tower 112. A substantial portion of the tar in these gases consists of compounds which are liquid under normal atmospheric conditions. These compounds are readily condensed in the quench tower by contacting the hot gases with a quantity of cool liquid tar. The liquid tar also serves as a scrubbing medium and operates to remove char solids entrained in the hot gases. In carrying out this step, the vapors are introduced into the bottom of the tar quench tower and pass upwardly around baffles 114 countercurrent to liquid tar introduced into the tower through conduit 142. The cooler vapors subsequently pass through a number of perforated trays 116, through a mist extractor 118 to remove entrained liquid droplets and exit from the quench tower through conduit 120. The liquids and solids removed from the vapors by the scrubbing tar are transferred from the bottom of the quench tower through pump 136 and are passed through conduit 138 and cooler 140. A portion of the cooled material is returned to the quench tower through conduit 142 and the remainder is yielded as product through conduit 138. The temperature of the gases leaving the top of the quench tower is about 160° F. This is still substantially above atmospheric temperature and in order to lower the temperature of the gases still further they are passed through a water cooler 122, where additional tars are condensed, and then into an accumulator 124 where a further separation of gas and liquid takes place. This final cooling step reduces the temperature of the gases to about 100° F. The gases are released from the accumulator through conduit 126 and pass into a Cottrell precipitator 128. Liquid is removed from the precipitator through conduit 132, is combined with accumulator liquid from pump 130 and conduit 134, and this combined stream is in turn added to the tar product passing through conduit 138. The final vapor product comprising primarily combustion gases and steam leaves the precipitator and the system through conduit 130.

The preceding discussion has been directed to a preferred embodiment of the invention, however it is not intended that the material presented be construed in any limiting sense, but that other equipment, process conditions, flows, etc. are also used within the scope of the invention. For example, to limit the possibility of equipment plugging prior to the drying operation it may be desirable to introduce wet coal directly from a mechanical conveyor, such as a bucket elevator, into the dryer vessel 10. Again, to provide a more flexible process, less dependent, or even separate, drying and preheating facilities may be provided with solids recovery from the gases from both operations being provided in a common scrubbing system as described.

The following data are presented to illustrate a typical commercial carbonization operation based on the processing arrangement of the drawing.

Flows:		Lb./hr.
Wet coal—10 to 400 mesh	-----	450,000
Water content	-----	35,000
Dry coal	-----	415,000
Bases from drying zone—		
Water	-----	61,500
Air	-----	16,000
Coal	-----	900

Char product.....	345,000
Volatile product.....	60,000
Solids content.....	1,000
Pretreater air.....	80,000
Carbonizer air.....	50,000
Char recycle (for pretreater temperature control).....	25,000
Feed coal heater—	
Coal circulation rate.....	1,250,000
Heating fluid—15° API hydrocarbon oil.....	1,550,000
Product char cooler—cooling fluid—15° API hydrocarbon oil.....	1,550,000
Cooling water injected into char product.....	28,000
Water to solids recovery tower.....	96,000
Tar quench tower reflux.....	580,000
Temperatures:.....	° F.
Wet coal.....	60
Drying zone.....	270
Preheating zone.....	480
Pretreating zone.....	725
Carbonization zone.....	950
Char hopper.....	230
Solids recovery tower.....	216
Tar quench overhead.....	165
Feed coal heater—	
Coal in.....	270
Coal out.....	480
Heating fluid in.....	500
Heating fluid out.....	400
Product char cooler—	
Char in.....	950
Char out.....	500
Cooling fluid in.....	400
Cooling fluid out.....	470
Cottrell precipitator.....	150
Product char.....	190
Tar product.....	350
Pressures:.....	P. s. i. g.
Drying zone.....	3.8
Preheating zone.....	6.0
Pretreating zone (top).....	11.0
Carbonization zone (disperse phase).....	8.0
Char hopper.....	3.5
Solids recovery tower.....	2.0
Tar quench tower.....	6.0
Average residence time of coal in:.....	Minutes
Pretreating zone.....	60
Carbonization zone.....	30
Gas velocity in:.....	Ft./sec.
Drying zone.....	2.0
Preheating zone.....	2.5
Pretreating zone.....	1.0
Carbonization zone.....	1.5
Density of solids in:.....	Lb./cu. ft.
Drying zone.....	25
Preheating zone.....	25
Pretreating zone.....	22
Carbonization zone.....	18

Having thus described the invention by reference to a specific example thereof, it is understood that no undue limitations or restrictions are to be imposed by reason thereof, but that the scope of the invention is defined by the appended claims.

I claim:

1. In the process for treatment of carbonaceous solids

to remove volatile constituents therefrom in which the solids are heated in a drying zone in a dense phase fluidized bed to remove water therefrom, gases leaving said dense phase bed are subjected to solids recovery treatment for removal of the major portion of the solids entrained therein, the dry solids from said dense phase bed are passed into a carbonization zone wherein a char product is formed, the char product is removed from the carbonization zone, cooled and accumulated in a product zone, the improvement which comprises passing the gases from the drying zone after solids recovery treatment to remove the major portion of the solids entrained therein a scrubbing zone, contacting said substantially solids free gases while in the scrubbing zone with a liquid to form a solids-liquid slurry, removing the solids-liquid slurry from said scrubbing zone and combining therewith char products from said product zone.

2. The process defined in claim 1 in which the carbonaceous solids is coal.

3. The process defined in claim 1 in which the liquid which contacts the gases in the scrubbing zone is water.

4. In the process for treatment of carbonaceous solids to remove volatile constituents therefrom in which the solids are heated in a drying zone in a dense phase fluidized bed to remove water therefrom, gases leaving said dense phase bed are subjected to a solids recovery treatment in which the major portion of the solids entrained therein are removed, the dry solids from said dense phase bed are passed into a carbonization zone wherein a char product is formed, the char product is removed from the carbonization zone, cooled and accumulated in a product zone as a dense phase bed fluidized with steam, the improvement which comprises passing the gases from the drying zone after the major portion of the solids entrained therein have been removed to a scrubbing zone, passing gases from said product zone to said scrubbing zone, contacting said gases with a liquid while in the scrubbing zone to form a solids-liquid slurry, removing the solids-liquid slurry from said scrubbing zone and combining therewith char products from said product zone.

5. The process defined in claim 4 in which the carbonaceous solids is coal.

6. The process defined in claim 4 in which the liquid which contacts the gases in the scrubbing zone is water.

7. An apparatus for treatment of carbonaceous solids for the removal of volatile constituents, a vessel adapted to define a drying zone, a vessel adapted to define a carbonization zone, a vessel adapted to define a char product zone, a vessel adapted to define a scrubbing zone, means for conducting gases from said drying zone vessel to said scrubbing zone vessel, means for conducting solid product from said carbonization zone vessel to said product zone vessel, means for conducting gases from said product zone vessel to said scrubbing zone vessel, means for supplying water to said scrubbing zone vessel, conduit means for removing char product from said char product zone vessel, means for introducing the solids-liquid slurry from said scrubbing zone vessel to said conduit means.

References Cited in the file of this patent

FOREIGN PATENTS

3,780/56	Great Britain	Feb. 7, 1956
K28,090	Germany	Feb. 13, 1956
708,372	France	Feb. 13, 1956