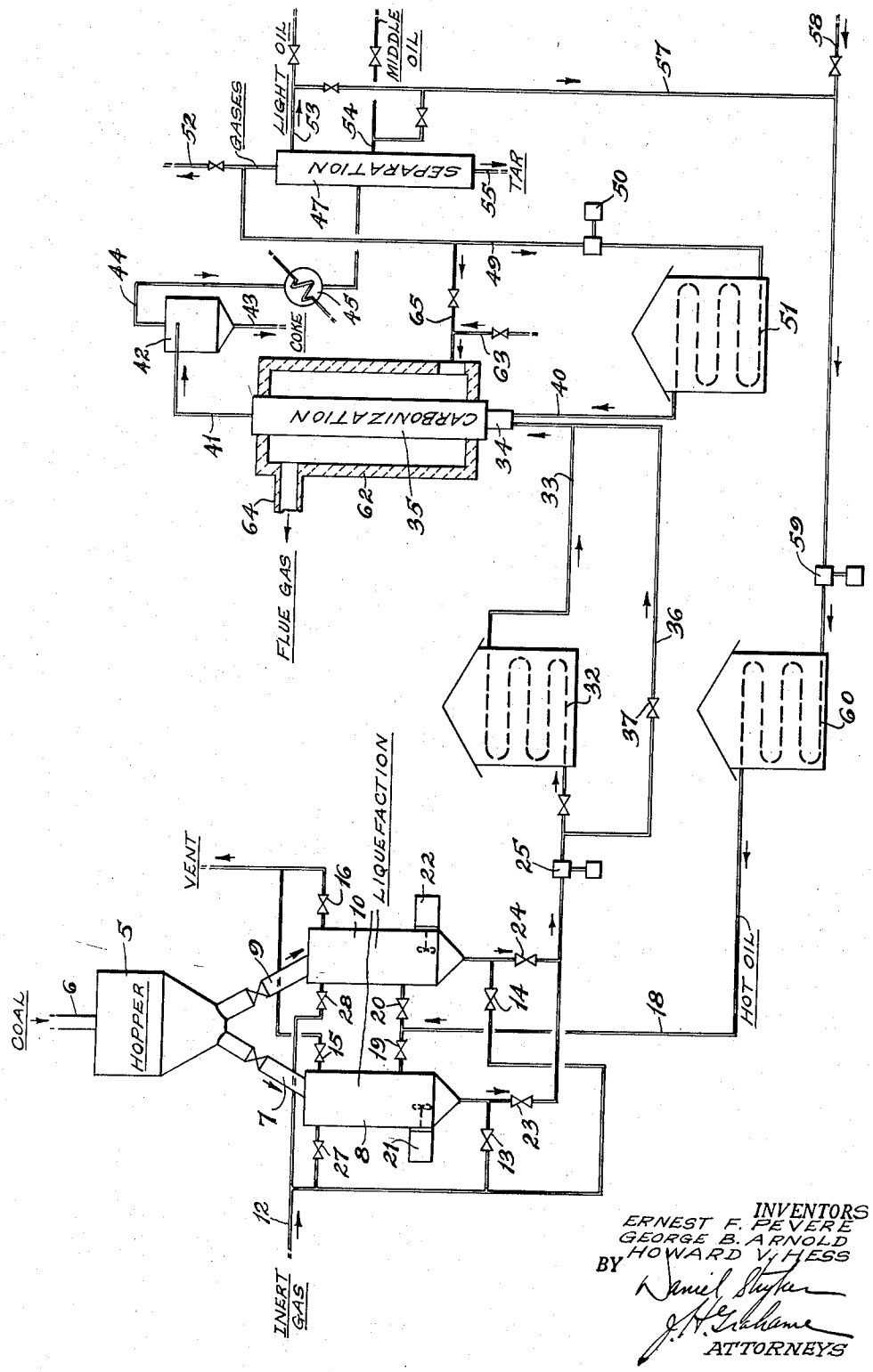
Dec. 29, 1953 E. F. PEVERE ET AL 2,664,390

CARBONIZATION OF COAL

Filed Nov. 19, 1948



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Patented Dec. 29, 1953

2,664,390

UNITED STATES PATENT OFFICE

2,664,390

CARBONIZATION OF COAL

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Application November 19, 1948, Serial No. 61,013

3 Claims. (Cl. 202—11)

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This invention relates to a process for the production of finely divided particles of coke from coal. The process of this invention may be applied to coals of various types, especially bituminous coals and lignite. The process is particularly applicable to the treatment of bituminous coals.

In the conventional processes for the production of coke from coal, the coal is heated, sometimes in the presence of a hot gas, to drive off volatilizable constituents from the coal. Carbonization or coking of the coal is generally carried out in a fixed bed in a coke oven. More recently, processes have been developed for carbonization of coal in particle form by passing hot gasses upwardly through a bed of coal particles under conditions of flow such that the particles are partly suspended in and agitated by the gas stream, effecting fluidization of the entire bed of particles. For successful operation of the fluid bed carbonization process, it is necessary to use coal of a rather narrow range of small particle sizes. This generally involves extensive grinding of the coal and sizing it prior to carbonization.

In the process of the present invention, fine grinding and careful sizing of the coal are not necessary, almost any size lump or particle being suitable for the process. In accordance with this process, coal is liquefied by heating in the presence of a liquid hydrocarbon and atomized. The resulting particles of atomized coal are dispersed in a gas stream and subjected to distillation at an elevated temperature while in a dispersed condition.

An object of this invention is to provide an improved process for the distillation of coal. Another object is to provide an improved process for the production of finely divided particles of coke from coal. Still another object is to provide a process for the conversion of coal particles of random sizes to uniform small particles of coke. A further object is to provide a process for the production of high quality carbon from coal. Other objects and advantages will be apparent to those skilled in the art from the following detailed description of the invention.

Most coals may be rendered plastic by heating in an inert atmosphere. The plastic point and degree of fluidity obtainable on heating varies somewhat with different coals. Coals which contain large amounts of anthraxylon and translucent attritus exhibit greater fluidity than those which contain large amounts of opaque attritus and fusain. Generally, coal becomes plastic at

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a temperature within the range of from about 550 to about 700° F. When coal is heated in the presence of a mineral oil, the coal and oil become miscible and may be mixed to form a substantially homogeneous viscous liquid. Some hydrocarbon oils, especially hydroaromatics, such as tetralin, decalin, and oil fractions obtained from coal, act as solvents and aid in the liquefaction of the coal.

At temperatures above about 700° F., there is a condensation of free radicals or unsaturated compounds (generated by thermal decomposition of coal substance) into materials more stable than the original coal substance. At temperatures above about 825° F., the rate of precipitation of insoluble polymer may exceed the rate of liquefaction. The coal should be kept at a temperature as low as possible during liquefaction and subsequent handling in the liquid state to prevent excessive precipitation of the solid polymer. The optimum temperature for liquefaction is dependent upon the type and source of the coal, and the period of time the coal must be kept in liquid state, and is best determined by trial for any given coal.

When the coal is liquefied with the oil with mechanical mixing, a substantially homogeneous liquid mixture may be obtained. Finely divided solid particles may be present in the mixture, either as a result of incomplete solution, precipitation of polymer, or both. This mixture may be atomized without further treatment other than removal of oversize solid particles. On the other hand, when the particles of coal are extracted with oil without agitation, about 65 per cent by weight of the coal is taken into solution in the oil leaving a porous friable solid residue. The liquefied coal, or extract, obtained by extraction is susbtantially free from ash and may be used in the present process for the production of high purity carbon. The undissolved portion of the coal is particularly suitable for reaction with steam or with a mixture of steam and oxygen for the production of a mixture of carbon monoxide and hydrogen.

In accordance with preferred practice of this invention, coal is liquefied by admixing it with a liquid hydrocarbon at a temperature within the range of about 550 to 850° F. The resulting liquid is atomized into a carbonization zone operated at a temperature within the range of from about 900 to about 1500° F. Near atmospheric pressures are advantageously used. Fixed gases derived from the coal are used as dispersing medium for atomization and suspension of the coal.

The coal may be liquefied at a temperature within the range of 550 to 700° F. and the resulting liquid heated to a temperature within the range of from about 800 to about 1000° F. prior to atomization. The pressure maintained on the liquefield coal feed stream prior to atomization is preferably sufficient to prevent any substantial amount of vaporization. Heat for carbonization may be supplied by heating the liquefied feed stream, the inert gas stream, the carbonization reactor, or by a combination of these methods. The inert gas stream is preferably preheated to a temperature on the order of 1000° F.; it may be heated to a considerably higher temperature.

In a preferred embodiment of this invention, the liquid mixture of coal and oil is atomized and the droplets dispersed in a heated stream of inert gases having a velocity sufficient to suspend the particles. The suspended particles are maintained at a temperature of from about 900 to about 1300° F. during carbonization.

The quantity of oil combined with the coal to form a liquid feed stream which may be atomized with conventional equipment may vary considerably depending upon the type of coal and oil used in the preparation step. Generally, a quantity of oil equivalent to about 50 per cent by weight based on the weight of the coal is sufficient although larger amounts are often advantageous from the standpoint of viscosity reduction.

The particle size of the coal fed to the process is not of particular importance. The smaller particles are more quickly liquefied. The particles should be of such size that substantially complete liquefaction takes place in a reasonable period of time. Generally, it is desirable to use particles ranging from about ½ inch in average diameter to powder.

In the present process, it is not necessary to resort to fine grinding or pulverization of the coal prior to preparation of the feed. It is contemplated that in most applications of the process, the coal will be reduced only to a particle size such that the largest particles are dissolved or disintegrated prior to discharge of the liquid from the liquefaction equipment. Coal may be crushed mechanically to a particle size of ½ to ¼'' in average diameter with a relatively small expenditure of power. The process lends itself to treatment of waste or low market value slack coal produced in mining and handling operations.

The atomized coal may be carbonized under conditions of temperature satisfactory for conventional carbonization of coal. In general, the temperatures used for distillation of coal range from about 900° F. to about 2000° F. or higher. The lower temperatures within this range are preferred, as the yield of valuable products from the coal is favored by low temperature distillation. In the conventional processes it is difficult to carbonize coal at temperatures on the order of 900 to 1000° F.

Carbonization may be carried out efficiently at the lower temperatures by the process of this invention. In atomized form the coal particles are more readily distilled than in conventional particle form. The oil also aids in the distillation of the coal. An oil fraction obtained from the process and having a boiling range of from about 400 to about 500° F., for example, is readily vaporized at the coal distillation temperature. Such volatile oils used for plasticizing or liquefying the coal in the present process aid in the distillation of the coal and recovery of desirable volatilizable constituents therefrom by reducing the effective partial pressure of the volatilizable constituents. Distillation is also favored by the passage of gases through the distillation zone. The coke particles may be maintained in the carbonization zone as a fluid bed. The particle size of the product may be controlled by regulation of the droplet size produced on atomization.

Pulverized coke entrained in the gas stream may be separated therefrom by means of a suitable separator, for example, a cyclone separator. The hydrocarbon liquid and condensible volatilizable constituents from the coal are recovered from the effluent gas stream. A part of the gas resulting from the carbonization may be recycled to the carbonization zone as inert gas for atomization and suspension of the coal. This gas may also be used as fuel, e. g., to supply heat to the distillation process. The carbonized product is in the form of small spherical particles which are eminently suitable for gasification in a fluid bed and are very reactive as fuel.

The invention will be more readily understood from the following detailed description and the accompanying drawing.

The figure is a diagrammatic elevational view illustrating a preferred mode of carrying out the process of the present invention.

With reference to the drawing, coal is supplied to a hopper 5 through a conduit 6. From the hopper, the coal may be fed through conduit 7 into liquefaction vessel 8 or through conduit 9 into liquefaction vessel 10. Any number of such vessels may be employed. While coal and oil are charged to one of the vessels to prepare a liquefied coal feed stream for carbonization, another is discharged to the carbonization step. When a vessel has been filled with coal from the hopper, inert gas may be admitted from line 12 to the vessel to purge it of air and other gases. Valve 13 associated with vessel 8, and valve 14, associated with vessel 10, are provided for this purpose. The purged gases may be vented from the vessels 8 and 10 through valves 15 and 16 respectively.

Preheated oil from line 18 may be admitted to vessel 8 through valve 19 and to vessel 10 through valve 20. When a vessel is charged with coal and hot oil, the valves are closed and the coal at least partially liquefied due to the combined effect of heating the coal and the solvent or plasticizing action of the oil.

Mechanical mixers 21 and 22 are provided to disintegrate and disperse any undissolved residue or precipitated solid material from the coal. A substantially homogeneous liquid mixture of coal substance and oil is thus obtained.

Generally, sufficient heat for the liquefaction may be supplied by preheating the oil stream. Additional heat may be supplied directly to the liquefaction vessels by suitable means not illustrated in the drawing. The oil may be supplied to the vessel at least partially in vapor form under a pressure sufficient to cause condensation in the liquefaction vessel and thereby furnishing additional heat for heating the coal.

The resulting liquid coal feed is discharged from the vessel 8 through valve 23 and from vessel 10 through valve 24 to a charge pump 25.

Inert gas from line 12 may be admitted to the vessel during the period of discharge of the liquefied coal therefrom. The inert gas may be supplied under sufficient pressure to maintain the desired pressure within the vessel to prevent substantial flashing of volatile hydrocarbons and aid in discharging the liquefied coal therefrom.

From the charge pump **25**, the liquefied coal feed stream is passed through a heating coil **32** wherein it is heated to a temperature within the range required for carbonization. The liquefied coal feed is charged through line **33** into an atomizer **34** associated with carbonization zone **35**. Alternatively, part or all of the coal feed stream may be fed without preheating directly from the charge pump **25** to the atomizer **34** through line **36** as controlled by valve **37**.

The atomizer **34** may be of conventional type and is constructed of materials which are resistant to erosion. A suitable atomizer may be one of the spray type wherein the liquid is forced under pressure through an orifice of small diameter. The atomized particles are dispersed in a stream of hot inert gases supplied to the carbonization zone through line **40**.

The carbonized particles of coal, or coke, are entrained in the hot gas stream and discharged through line **41** into a cyclone separator **42**. The coke is separated from the hot gas stream and discharged from the cyclone separator through line **43**. The hot gas stream, which contains vapors of the oil used in the preparation of the feed stream and volatilized constituents from the fresh coal feed, is discharged from the cyclone separator through line **44**, cooled in a cooler **45**, and passed to a separation system **47**.

In the separation system, a gaseous fraction comprising methane and other gases resulting from distillation of the coal is separated from the normally liquid products of distillation comprising, for example, light oil, middle oil and tar fractions. A portion of the gases from the separation zone is recycled through line **49** and pump **50** to a heating coil **51** wherein the gases are heated and introduced into the carbonization zone through line **40**. Part or all of the remaining portion of the gases may be discharged from the system through line **52** for use as fuel or other purposes. A light oil fraction is withdrawn from the separation zone through line **53**, a middle oil fraction, through line **54**, and a tar fraction through line **55**.

The light oil fraction, the middle oil fraction, or a mixture of the two may be recycled to the liquefaction zone through line **57**. The remaining middle oil may be subjected to hydrogenation in vapor phase in accordance with general conventional practice for production of motor fuels. Other oils may be supplied, if desired, to the oil stream in line **57** through line **58**. The oil stream is pased to a pump **59** which forces the oil at the desired pressure through a heating zone **60** wherein it is preheated to the desired temperature prior to admission to the liquefaction zone via line **18**.

Heat may be supplied to the carbonization zone **35** by direct heating of the walls of the vessel or retort in which the carbonization is carried out. For this purpose, a furnace **62** may be provided surrounding the carbonization vessel. Fuel may be supplied to the furnace from line **63** from any suitable source and fuel gases discharged therefrom through duct **64**. A portion of the gaseous fraction resulting from the distillation of the coal may be supplied to the furnace **62** by line **65** as fuel.

It will be understood by those skilled in the art that various alternatives in the way of equipment and methods of handling the various streams and utilizing the heat contents thereof to maximum advantage may be resorted to. In the interest of simplicity, various conventional pieces of apparatus such as control valves, heat exchangers and the like have been omitted from the drawings and description of the process.

In a typical operation of a process of the type illustrated in the drawing, Pittsburgh bed coal containing about 2 per cent water, 31 per cent volatile matter, 58 per cent fixed carbon and 9 per cent ash, as received, is subjected to carbonization. The coal is charged to a liquefaction vessel in the form of lumps ranging from about ½" to about ¼" in average diameter. This coal is mixed with a middle oil fraction obtained from the distillation of coal. Approximately equal parts by weight of coal and oil are charged to the liquefaction vessel. The oil has a boiling range of from about 450 to about 550° F., and is preheated to a temperature of about 825° F. prior to admixture with the coal. A pressure of about 300 pounds per square inch gauge is maintained on the liquefaction vessel to insure liquid phase conditions. The coal and oil are subjected to mechanical agitation to insure dispersion of carbonaceous residue. About 65 per cent by weight of the coal is taken into solution by the oil and the remaining 35 per cent dispersed in the form of minute particles. The resulting liquid coal stream, at a temperature of 600° F., is atomized with an inert gas comprising largely methane. The atomized particles are suspended in the inert gas at a temperature of about 1000° F. The gases and dispersed coal particles are passed through an elongated tubular retort which is externally heated where they are further heated to a temperature of about 1200° F. Coke particles resulting from distillation and carbonization of the coal are separated from the entrained gas stream as a granular product of relatively uniform particle size.

Obviously many modifications and variations of the invention, as hereinbefore 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.

We claim:

1. A process for the carbonization of a fusible coal containing volatilizable constituents with the production of dry solid powdered coke and recovery of said volatilizable constituents which comprises contacting said coal in particle form with a normally liquid hydrocarbon solvent for coal substance, agitating said coal particles with said solvent at a temperature above the temperature at which said coal is rendered plastic and within the range of from about 550 to about 700° F. whereby said coal becomes miscible with said oil solvent and a substantially homogeneous liquid mixture containing all of the original components of said coal is obtained, atomizing said liquid mixture into a stream of a substantially inert gas thereby suspending said atomized liquid therein, passing the suspension upwardly through an externally heated carbonization zone maintained at a temperature of from about 900 to about 1,300° F., maintaining said atomized liquid in suspension in said carbonization zone for a period of time sufficient to effect substantially complete volatilization of said hydrocarbon solvent and volatilizable constituents of said coal and form dry solid particles of coke suspended in vaporous product of volatilization, and subsequently separating said solid particles of coke from volatilized products.

2. A process for the carbonization of a fusible coal containing volatilizable constituents with

the production of dry solid powdered coke and recovery of said volatilizable constituents which comprises contacting said coal in particle form with a normally liquid hydrocarbon solvent for coal substance and agitating said coal particles and said solvent at a temperature above the temperature at which said coal is rendered plastic and within the range of from about 550 to 700° F., whereby said coal is rendered miscible with said oil and substantially homogeneous liquid mixture containing all of the original components of said coal is obtained, heating said liquid mixture to a temperature within the range of from about 900 to about 1,300° F. at a pressure sufficient to prevent substantial vaporization of said liquid, atomizing said liquid mixture into a stream of substantially inert gas, passing said suspension upwardly through an externally heated carbonization zone, maintaining said suspension at a temperature within the range of from about 900 to 1,300° F. and at a substantially atmospheric pressure in said carbonization zone for a period of time sufficient to effect volatilization of substantially all of the volatilizable constituents of said mixture and produce dry solid particles of coke suspended in vaporous products of volatilization, and subsequently separating said solid particles of coke from the volatilized products of carbonization.

3. A process for the carbonization of a fusible coal containing volatilizable constituents with the production of dry solid powdered coke and recovery of said volatilizable constituents which comprises contacting said coal in particle form with a normally liquid hydrocarbon solvent for coal substance, agitating said coal particles with said solvent at a temperature above the temperature at which said coal is rendered plastic and within the range of from about 550 to about 700° F. whereby said coal becomes miscible with said oil solvent and a substantially homogeneous liquid mixture containing all of the original components of said coal is obtained, atomizing said liquid mixture into a stream of a substantially inert gas thereby suspending said atomized liquid therein, passing said suspension upwardly through an externally heated carbonization zone, maintaining said suspension at a temperature within the range of from about 900 to about 1,300° F. within said carbonization zone for a period of time sufficient to effect substantially complete volatilization of said oil and volatilizable constituents of said coal and form dry solid particles of coke suspended in vaporous products of volatilization comprising normally gaseous hydrocarbons and normally liquid hydrocarbons including solvent for said coal substance, subsequently separating said solid particles of coke from the volatilized products, separating normally gaseous hydrocarbons from said volatilized products, supplying said normally gaseous hydrocarbons to said atomization step as said inert gas, separating a normally liquid hydrocarbon fraction containing solvent for said coal substance from said volatilized products, and passing said liquid hydrocarbon fraction into contact with said coal as said hydrocarbon solvent.

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