UNITED STATES PATENT OFFICE

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METHOD OF MAKING COKE BRIQUETTES

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This invention relates to improvements in the carbonization of coal so as to produce coke and valuable by-products, and more particularly to such a method whereby there will be a maximum yield of tar toils together with coke, of the very highest quality.

The invention is related generally to that described in Patent No. 1,689,688, issued February 28, 1928, to Ludwig Kern and myself. In said patent, the coal is described as being ground to 150 mesh or finer, and to such powdered coal there is added enough of an alkali in aqueous solution to react with the freed ash-forming constituents of the coal, to form a plastic binder therewith, which through suitable homogenizing, plastifies the entire mass into a firm moldable body from which the briquettes are formed. These briquettes are thereafter dried, and then subjected to distillation or carbonization to recover the volatiles and to form coke. However, tests have shown that although the process is efficient with some coals, yet with others the results were not entirely satisfactory.

The instant invention, although related to the teachings of said patent, differs therefrom in important aspects, and the results seem uniform ly better. Principally, it differs in that the alkali is replaced by a hydrocarbon compound that is liquid or semi-liquid at normal room temperatures, and which is chemically a close ally to all types of coal and analogous carboniferous materials.

One of the important objects of my invention is to provide a process that is extremely flexible in its application, and wherein the solid carboniferous materials may be treated separately or in combinations, depending upon their physical and chemical make-ups, as for example, by combining and treating bituminous, semi-bituminous and sub-bituminous coals alone, or in combination with anthracites or other substantially resinous-free coals.

Other important objects of my invention are to provide a method of treating materials of the kind described, so that various types of solid carbonaceous products may be produced, as for example, semi-coke briquettes readily ignitable at about the same temperatures as bituminous coal but which will burn smokeless and without requiring drafts, so that it is an ideal fuel for domestic use; an artificial anthracite having all of the desirable properties thereof, but which is ignitable at from 100° to 200° C. lower temperature than said natural anthracite; in a brick form suitable for blast furnaces, foundries, industrial furnaces and boilers; or to coal gasification, hydrogenation, and for other products requiring high fixed-carbon contents.

A further object of my invention is to provide a process of treating solid carbonaceous fuel, so that a favorable exchange is brought about therein in the quantity, speed, and chemical constitution of the by-product hydrocarbons derived therefrom, and at relatively low temperatures.

An added object of my invention is to produce a conversion of highly volatile bituminous coal, rich in resinous content, into relatively large yields of primary tar oils of low specific gravity, averaging between 0.5 and 1.06, and of relatively low boiling points, said oils being composed mainly of saturated and unsaturated paraffines, olefines, napthenes, high-molecular hydrocarbons, and being especially rich in middle oils that contain the highly-prized homologues of the phenolic series.

Another object of my invention is to produce sufficient porosity in the coal being treated, so that it absorbs the plasticizing medium properly while at normal room temperature.

An additional object of my invention is to practice the invention by intermixing the coal with a liquid or semi-liquid hydrocarbon compound flowable at room temperature and having a boiling point of approximately 125° to 300° C. and which in contradistinction to water, has a high affinity for coal, so as to agglomerate and agglutinate therewith and act as a solvent for the resinous constituents thereof, and in itself having lubricating, coking and distillable properties to thereby increase the fixed and reactive carbon content of the coke.

A still further object of my invention is to provide a method of the kind described, that will not only recover the volatiles inherent in the coal, but in addition, will recover those that may have been added thereto in the carrying out of this process, and whereby the process may become cyclic and self-contained.

Other added objects of my invention are to provide a method set forth, wherein the added constituents are introduced at room temperature, carbonization commenced without drying of the briquettes and carried on at low temperatures, and wherein the coal is changed into a moldable and shape-retaining briquettable mass prior to carbonization.

To this end, my invention consists in the novel process herein shown and described, and the ingredients used, as will be more clearly pointed out in the claims hereunto appended.
In the drawings, there is illustrated diagrammatically, a flow sheet of the sequence of steps in the practice of the invention. In carrying out my invention, I may use practically any coal, although I prefer to use a bituminous coal for many reasons, as will be more clearly understood from the disclosures hereinafter, and it may be mentioned that low-grade coals are admirably suited to such use. For example, I have successfully used a low-grade Illinois coal running about 10 percent ash, 3 percent sulfur, 12 percent moisture, 39 percent volatiles, and 37 percent fixed carbon, and by treatment by this process have converted the same into a high-grade semi-coke briquette containing about 78 to 83 percent fixed carbon, easily ignitable and burnable without draft and entirely free of visible smoke; thus enabling the use of such briquettes even in those localities having the most stringent anti-smoke regulations.

Such solid carbonaceous fuel, preferably a coal high in resinic content, as for example the bituminous coals, is crushed or otherwise comminuted to pass through a 30 to 40 standard mesh screen. Fuel, either from the same source, or from another source, may be prepared to a more finely comminuted state, so that it becomes pulverized and will pass through about a 200 mesh screen. If desired, this pulverized coal may be taken from what would otherwise be refuse, as from a coal-dust collector.

These two coals, that is, the coarser and the finer, are to be intermixed in about the proportion of from \( \frac{3}{4} \) to \( \frac{2}{3} \) of the former to from \( \frac{1}{4} \) to \( \frac{1}{3} \) of the latter, say for example about 190 kilo of the pulverized to 250 kilo of the coarser to make up 500 kilo batch. These two coals may be stored separately somewhat as indicated in the flow sheet, and from whence they may be withdrawn as needed.

As coal, no matter what its fineness, is not moldable at room temperature except possibly under unusually great pressure, without some sort of a binder, it will be seen that I have minimized this difficulty, and my process enables the briquetting at relatively low pressure and room temperature, and wherein the briquettes will retain their shape through carbonization and through the subsequent handling, including relatively rough handling.

I have found that coarse and pulverized coals in about the proportions stated will agglutinate under certain conditions without an extraneous binder, at relatively low pressure and at normal room temperature, by introducing said coals into a wet pan grinder and agitator wherein heavy steel millers move, with a slight amount of water, and after said millers have acted for a short time, say about 15 or 20 minutes thereon, to grind, compress, knead and lubricate the mixture, it becomes changed in its chemical and physical form to an elastic, rubber-like lump mass or slab, and the resinic content of the coal has been forced from its original places and states of deposit into the very pores of the coal and especially into the capillary openings formed between the coarse and the pulverized coal particles.

However, I have found that for best results, it is desirable that an additive be incorporated into the coals that are to be intermixed, and have termed this additive an important ingredient of the "hydrocarbon compound," the compound comprising such additive or additives together with the pulverized coal, as will be more clearly hereinafter set forth.

The carrier or diluent for this hydrocarbon compound is liquid or semi-liquid at normal room temperature, with a boiling point ranging broadly between 125° and 300° C., and has the power of being solvent for the volatiles or resinic content of the coal. Such carrier may be derived from various sources and may include those from coal tar or petroleum, including the first fractions or light oils from distillation or carbonization, and may be generally termed "hydrocarbon naphtha.

In the commencement of my operations such liquid may have to be obtained from an extraneous source, but after the process has gotten under way, sufficient amounts may be taken off and returned into the operations to thereby make it a cyclic process. This solvent acts as a vehicle for the resinic constituents of the coal.

If the liquid or semi-liquid hydrocarbon having the foregoing characteristics does not have enough resinic content for the carrying out of this process to best advantage, it may be necessary that a portion of the same be added. If the coal to be used does not have enough resinic content for the carrying out of this process to best advantage, it may be necessary that a portion of the same be added. Such solvent, or asphaltic substance, sometimes called pitch, is added to the mixture in quantities about the same fineness as the pulverized coal.

This resinic substance and the liquid or semi-liquid hydrocarbon described, are preferably intermixed with the pulverized coal, using just enough of the liquid to make into a paste of the desired consistency, and which takes about 20 gallons of the liquid to about 350 kilos of the pulverized coal.

Now, in order to better enable the components of this hydrocarbon compound (the liquid hydrocarbon solvent and asphaltic substance, or pulverized coal) to react fully with one another, it is introduced into an agitator or the like, wherein it is intimately homogenized, with or without heating.

Next, the pulverized coal so treated is introduced in the form of a slurry of hydrocarbon compound or in a wet pan grinder and agitator, where together with added coarser coal, in about the proportions stated, heavy steel millers roll, compress, grind, knead and otherwise intimately intermix the same to form the contents into a slurry, agglomerated lump mass of elastic rubber-like mass. The entire physical and chemical structure of the mass has thereby become changed and sponge-like, that a piece will withstand the impact of a 25 to 30 foot drop onto a brick floor without breaking or chipping.

Such material may be readily imparted to form-retaining briquettes at room temperature, with comparatively low pressure, say with up to 5000 pounds per square inch, depending on the moisture content.

It will be found that the absorbed compound envelopes all of the coal particles with a distinct individual coating, acting as the vehicle for lubricating, agglomerating and otherwise binding into such physical characteristic that the particles permit of compression into the briquettes.
at low pressure. If, in the mixing of the mass, there is not enough moisture for proper molding and briquetting, as will be apparent from visual inspection, additional fluidity may be obtained by added water. Although it is well-known that bituminous coal is by nature water-repellent and resists water, yet it is not to be expected to change its characteristics and become spongy and rubber-like after the aforesaid treatment, and capable of absorbing close to one-half of its weight in liquid hydrocarbon, and that a 55 gram briquette for example holds enough moisture to be reduced to approximately 30 grams after carbonization.

When the mixing has terminated and the charge has assumed the slab-like and rubber-like elastic structure mentioned, and which generally takes only about 15 minutes or so, depending upon the varying conditions that may be encountered, the mass is then preferably disintegrated or fragmented by suitable means, and then, either in such fragments or after shaping into briquettes under relatively light pressure, it is ready for carbonization and distillation without drying.

This elastic mass, shaped or not, as preferred, is carbonized and distilled at relatively low temperatures, as will be more clearly hereinafter set forth, and the crude tar oils obtained will have a low specific gravity and low boiling points and may be separated or fractionated in the well-known manner.

The liquid hydrocarbon naphthas added to the pulverized mass before carbonization act in the best of dry carbonization upon the resinic part of the coal, dissolving and separating the same from the fixed carbon and ash constituents of the coal. This action takes place at low temperatures, and this results in increasing the mechanical strength of the resultant coke, and besides, accelerates extraction. Carbonization takes place in from 2 to 8 hours with my method, as compared with from 20 to 26 hours in the old gas-making and coke-oven methods, with equal amounts of coal.

It will be noted from the chart, that this very important hydrocarbon liquid, as has performed its function, be reclaimed in the recovery of the primary tar oils, and can be returned cyclically, in the process, as can the pitch.

The first fractions, the light oils, are taken off between 0° to 190° C., having a specific gravity of about .860, and include naphthas, benzol, toluol, xyol, some phenols.

The second fractionating stage, between 170° and 270° C., recovers the middle oils, having a specific gravity of about .870, and includes homologues of the phenolic series, phenol derivatives, cresols, cresolate oils, carbolic acids.

The third fractionating stage, between 270° and 360° C., recovers the heavy oils, with a specific gravity of about between .990 and 1.000, and besides some of the middle oil may include some anthracene oil.

Above 360° C., remains the residue of pitch or artificial asphaltic substance, and which may provide the resinic substance for the carrying out of this process.

The foregoing temperatures and gravities may vary within fairly wide limits, depending upon the basic materials employed, and therefore changes in carbonization and fractionating will suggest themselves to the operators. It is to be again stressed that the light oil, the hydrocarbon naphtha, as well as the pitch, may be returned cyclically, to the process.

If desired, said residue tar may be further treated for recovery of valuable constituents, or to refine the same.

It may be more economical or expedient when starting operation, to substitute for said coal-tar naphtha, another naphtha having sufficiently like properties to enable its use herein, as for example some other hydrocarbon naphtha, say petroleum naphtha, having about the same boiling-points and being similarly flowable at normal room temperature, such a distillate may be of the aromatic, aliphatic or naphthenic series or equivalent solvents, and to which may be added a small amount of phenolic compounds, cresols, xyleneols, and the higher phenols, thus enhancing the solvency of the naphthas.

It will be found that the mass remaining after the aggristinizing, is spongy and porous, with a consequent irregularity of its cellular structure, and this is of great importance as it permits of rapid passing of the vapors and gases from the briquettes into the condensers at relatively low temperatures. Such unobstructed volatilization of the tar vapors protects the briquettes from severe secondary decomposition and loss of very valuable primary tar oils. The usual swelling of the briquettes at the plastic stage of carbonization, generally caused by the accumulated and entrapped gases and vapors between 350° and 450° C., is eliminated, and localized overheating of hydrocarbon vapor into pyrogene decomposition is eliminated.

Other advantages obtained by the increasing of porosity of the coal are that there is a rapid accelerated and economical conversion of the coal, besides the unobstructed passage of vapors, without overheating.

If desired, the coke remaining after carbonization as above described may be treated further, and preferably while still hot, by transferring while under exclusion of air, to a suitable coking retort, and gases extracted for further treatment.

It is well known that in crushing coal so as to pass through a standard mesh 20 to 30 per inch, about one-third to one-fourth of the coal will be reduced to dust of a fineness sufficient to pass through about a 200 mesh screen. Since this proportion of crushed coal to dust is about the same proportion used in my invention, the process is very economical. Further, it is not necessary in making a paste by adding a hydrocarbon oil and resinic substances to the dust to first separate the dust from the coarse particles of coal; as the dust will be taken into suspension by the hydrocarbon oils without such separation.

Having thus described my invention, it is obvious that various immaterial modifications may be made in the same without departing from the spirit of my invention; hence I do not wish to be understood as limiting myself to the exact process herein set forth and described, except as limited by the state of the art to which this invention appertains, and by the claims hereunto appended.

What I claim as new and desire to secure by Letters Patent is:

1. A method of converting highly bituminous coal into coke briquettes comprising crushing the coal to pass a 20 to 40 mesh screen, to provide relatively coarse particles, preparing additional coal to pass an approximately 200 mesh screen, to provide pulverized particles, adding liquid
naphtha to the pulverized coal in the proportion of about 20 gallons of naphtha to 350 kilograms of pulverized coal, adding pulverized pitch of about the same fineness to the pulverized coal and naphtha, mixing and agitating the pulverized coal, naphtha and pitch in the absence of water to form a paste, adding to the paste two to three times as much by weight of the coarse coal particles, with respect to the amount of pulverized coal in the paste, intermixing the coarse particles and paste by a combined rolling, crushing, grinding and kneading action at room temperature, adding enough water to maintain fluidity during the intermixing, continuing the rolling, crushing, grinding and kneading for about 15 minutes, whereby the mixture becomes rubbery and slab-like, pressure-molding such rubbery and slab-like material into briquettes, carbonizing said briquettes.

2. A method of converting highly bituminous coal into coke briquettes comprising crushing the coal to pass a 30 to 40 mesh screen, to provide relatively coarse particles, preparing additional coal to pass an approximately 200 mesh screen, to provide pulverized particles, adding liquid naphtha to the pulverized coal in the proportion of about 20 gallons of naphtha to 350 kilograms of pulverized coal, adding pulverized pitch of about the same fineness to the pulverized coal and naphtha, mixing and agitating the pulverized coal, naphtha and pitch in the absence of water to form a paste, adding to the paste two to three times as much by weight of the coarse coal particles, with respect to the amount of pulverized coal in the paste, intermixing the coarse particles and paste by a combined rolling, crushing, grinding and kneading action at room temperature, adding enough water to maintain fluidity during the intermixing, continuing the rolling, crushing, grinding and kneading for about 15 minutes, whereby the mixture becomes rubbery and slab-like, pressure-molding such rubbery and slab-like material into briquettes, carbonizing said briquettes, and recovering the products of distillation.

FERNANDO C. KERN.

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