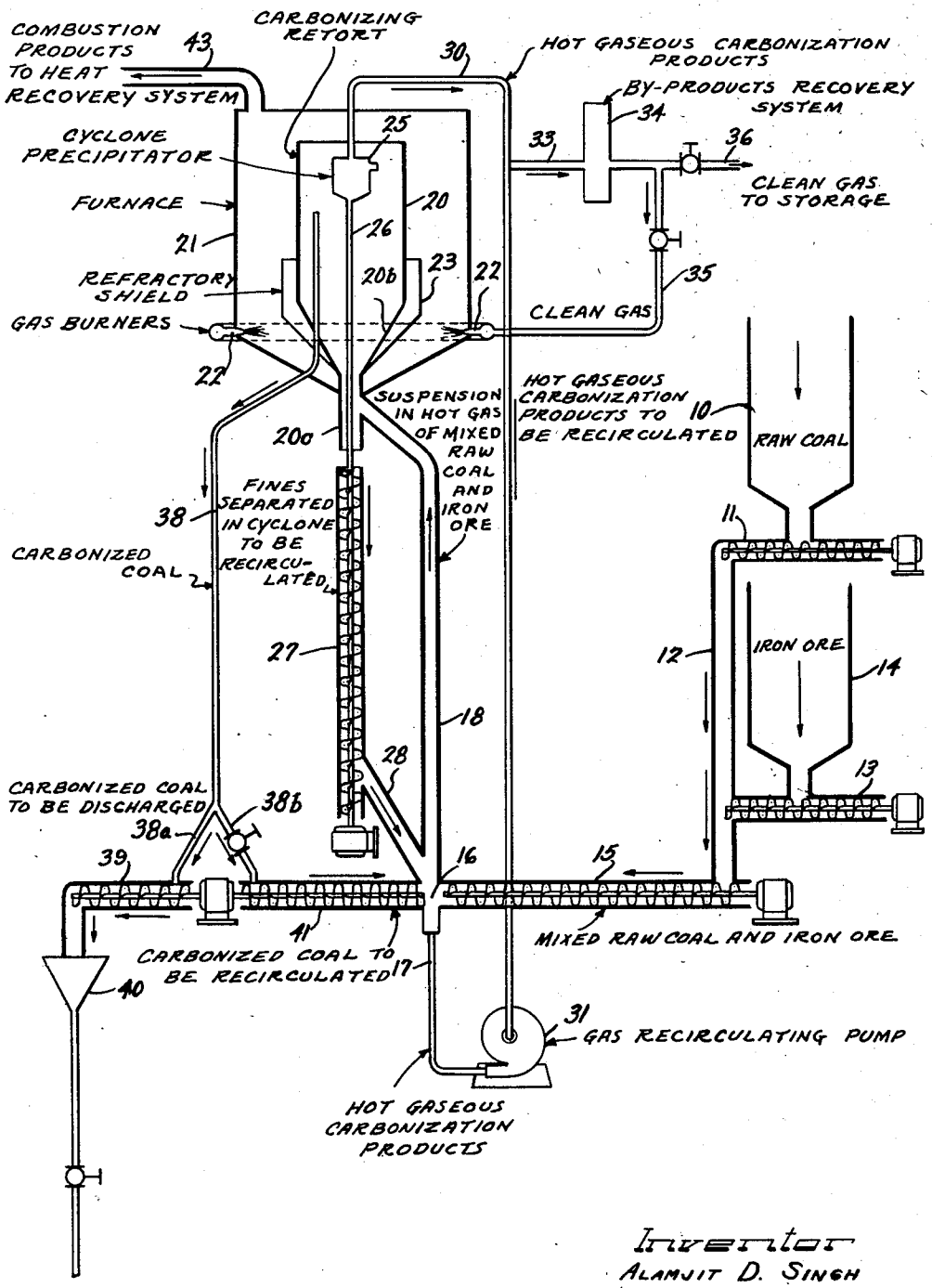
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METHOD OF CARBONIZING COAL WITH IRON OXIDE

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METHOD OF CARBONIZING COAL WITH IRON OXIDE

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This invention relates to a method of carbonizing coal and, more particularly, medium or high volatile coking coal.

Coals are now generally divided into two broad classes known as coking and non-coking coals. In this sense coking coals include all classes of coals that coke or cake, or swell or agglomerate in any way, and not merely those coals known as coking coals that are suitable to make blast furnace coke. In other words, coking coals are those that whenever sufficient heat is applied fuse to form coke masses. Non-coking coals, on the other hand, will not ordinarily coke when heated to a fusion or plastic temperature, but may instead disintegrate.

Depending on their content or volatile matter, coking coals are referred to as low volatile, medium volatile and high volatile coking coals. These three classes of coking coals contain, respectively, less than 25%, from 25 to 30%, and from 30 to 40% of volatile matter that is removed on carbonization. Metallurgical coke is ordinarily prepared from low volatile coking coals in a high temperature carbonization treatment requiring less than 24 hours. A satisfactory metallurgical coke can also be prepared from medium or high volatile coking coals by a carbonization treatment requiring from 24 to 36 hours.

I have now invented a method for producing coke from medium or high volatile coking coals by means of a carbonization treatment that may be completed in from 16 to 18 hours or less time. For this purpose, I first flow upwardly through a body of comminuted medium or high volatile coal a hot more or less inert gas at a rate such as to maintain said body of coal in a state of agitation. The gas and the retort containing the comminuted coal are maintained at such a temperature as to effect a low temperature carbonization of the coal leaving a char or semicoke while avoiding such agglomeration of the comminuted coal particles as would prevent free agitation of the body of comminuted coal or free passage of the gas therethrough. The semicoke or char thus obtained is then admixed with additional amounts of comminuted medium or high volatile coking coal, and the resulting mixture is subjected to a high temperature carbonization yielding a high grade metallurgical coke in from 16 to 18 hours or less time.

For the purpose of preventing such agglomeration or sticking together of the coal particles in said low temperature carbonization as would prevent agitation of the coal by a gaseous medium, I may employ steam as the agitating gas;

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or I may subject the coal to a preliminary heat treatment (if desired, by agitation with a hot gas) at a temperature not above 430° C. until from 4 to 5% (by weight of the coal) of the volatile content has been driven off, as by continuously moving the coal through a carbonizing retort in such a manner as to gradually raise the temperature of the entering coal from about 400° to about 430° C. over a space of not less than five feet (when a retort of six inch diameter is used); or I may incorporate with raw coal a small amount of sodium carbonate (1 to 3% or more); or I may recycle some of the semicoke or char so that the charge entering the retort will contain, say, about 40% of recycled char or semicoke.

I have found that the semicoke or char produced by the low temperature carbonization described hereinabove has a low bulk density, ranging from 25 to 28 pounds per cubic foot. This low bulk density causes difficulties in the blending of the semicoke or char with raw coal for the high temperature carbonization, so that the resultant high temperature carbonization product is lacking in uniformity. Further, the low bulk density of the semicoke or char lowers to an undesirable extent the bulk density of the final high temperature carbonization product which, if used in a blast furnace, would impair the capacity of the blast furnace.

I have further found that metallurgical coke having the desired bulk density can be produced by the method hereinabove described by the use of a suitable amount of an iron oxide or ore incorporated with the carbonaceous material being treated, not indiscriminately at any stage of the process, but by admixture with raw coal prior to the low temperature carbonizing step. For, as I have found, when the iron ore or oxide is incorporated with raw coal prior to the low temperature carbonizing step, then the particles of iron ore or oxide are entrapped within the particles of semicoke or char produced in the low temperature carbonization step, so that this step will yield a char or semicoke of uniformly increased bulk density (practically equal to that of raw coal) that will not segregate in the subsequent high temperature carbonizing step. No segregation of any particles of iron ore or oxide will occur at any stage of my coking process, if the iron ore or oxide is incorporated with the raw coal prior to the low temperature carbonization step. The resultant semicoke or char is magnetic and lends itself particularly well to agitation by a gaseous medium.

It is therefore an important object of the pres-

ent invention to provide a method for materially reducing the time heretofore required for the production of high grade metallurgical coke from medium or high volatile coking coals.

Another object of the present invention is to provide a two-stage process for the production of high grade metallurgical coke from medium or high volatile coking coals and including an initial low temperature carbonization of a fraction of the coal to be carbonized followed by a high temperature carbonizing step carried out with a mixture of the initially carbonized fraction with the remaining fractions, said first step being carried out by subjecting the coal to be carbonized in comminuted form and at an elevated temperature to the agitating action of a hot gaseous medium flowing upwardly through the coal.

A further object of the present invention is to provide a method for carrying out the low temperature carbonization of comminuted coal normally tending to agglomerate at carbonizing temperatures under the agitating action of a hot gas but without such agglomeration as would interfere with said agitating action, so that said agitation can be maintained throughout said carbonizing step.

A further object of the present invention is to provide a method for increasing the bulk density of the low temperature carbonization product as well as the high temperature carbonization product of the methods described hereinabove and to effect such increased bulk density by the addition to the coal of an iron oxide or iron ore without segregation of such iron ore or iron oxide in any carbonizing step.

Still another object of the present invention is to provide a method for carbonizing medium and high volatile coking coals for various purposes, in particular, for metallurgical purposes.

Other and further objects and features of the present invention will become apparent to those skilled in the art from the following description, accompanying drawings and appended claims.

By way of an illustrative example, applicant will hereinbelow describe the carbonization of a specific coal according to the methods of the present invention. More particularly, applicant refers to Wheelwright slack coal having the analysis tabulated hereinbelow:

Volatile Matter	Fixed Carbon	Ash	Sulfur	Moisture	B. t. u./#
Dry 34.69	58.60	6.71	0.69	----------	13,338
Natural 34.17	57.72	6.61	0.68	1.50	13,186

The screen analysis of this coal is tabulated as follows:

Screen size	Wheelwright Coal
	Per cent
On 30 Mesh	0.6
On 40 Mesh	5.1
On 60 Mesh	19.8
On 80 Mesh	15.1
On 100 Mesh	6.3
On 200 Mesh	20.2
Minus 200 Mesh	32.5
Total	99.6

Reference is made to the accompanying flow sheet which illustrates diagrammatically the apparatus and methods of the present invention. As shown on the flow sheet, the raw coal is drawn

from a hopper 10 and delivered by a screw conveyor 11 to a duct 12 into which a conveyor 13 brings iron ore, blast furnace flue dust or the like, drawn from the hopper 14. Suitably from about ½ to 20% (by weight of the total charge) of flue dust or the like is added to the coal. The mixed coal and ore or blast furnace flue dust are discharged from the duct 12 into a feeder 15 which conveys the mixture to a point 16 where the mixture is picked up by a hot gas issuing from a conduit 17 and carried through a conduit 18 into the bottom of a carbonizing retort 20. As shown, the carbonizing retort 20 is generally cylindrical in form but has a conical bottom 20*b* terminating in a constricted cylindrical extension 20*c*. The retort 20 is disposed within a furnace 21 heated by interior gas burners 22 that impinge upon the refractory shield 23 that serves to prevent direct flame impingement upon the lower portion of the retort.

Within the retort 20 there is disposed near the upper cover of the retort a cyclone precipitator 25. The finely divided solid material entrained by the gas leaving the retort 20 and entering the cyclone precipitator 25 is discharged into a conduit 26 which extends downwardly from the cyclone 25 within the conical bottom 20*b* and the constricted cylindrical extension 20*c* of the retort bottom for delivering the fine material to a conveyor 27 which in turn discharges into a duct 28. This duct 28 delivers the fine material to the point 16. The fine material is therefore recirculated through the retort.

Preferably the retort 20 is filled with coal approximately to the level of the bottom of the cyclone precipitator 25.

The gaseous and tarry products of the carbonization are drawn off from the cyclone 25 through a conduct 30 extending to a gas pump 31 which delivers the gas under pressure to the gas conduit 17. Excess gas not required for recirculation as well as tar formed in the retort are carried through a conduit 33 branching off from the conduit 30 to a by-product recovery system indicated by the reference numeral 34 from which the clean gas is partly sent through a valved conduit 35 to the burners 22, the excess of clean gas being piped through a valved conduit 36 to a storage gas holder.

The carbonized coal may be displaced upwardly by additional raw coal and withdrawn from the retort 20 through a conduit 38 having an upper end extending upwardly within the retort almost to the level of the cyclone separator 25. Ordinarily the upper end of the conduit will not extend above the body of coal or coke in the retort 20. At its lower end, the conduit 38 is bifurcated to form branches 38*a* and 38*b*. The branch 38*a* discharges into a conveyor 39 to deliver the semi-coke to a hopper 40 for further blending with raw coal to be charged to high temperature coke ovens. The other branch 38*b* is valved and permits flow of carbonized coal to a conveyor 41 which delivers semicoke to the point 16 for recirculation through the retort 20.

The hot combustion gases from the furnace 21 are vented through a conduit 43 to a heater recovery system for preheating gas and air (not shown).

The above described apparatus may be operated continuously or intermittently, and with or without recirculation of semicoke. For instance, intermittent operation can be carried out by introducing, as described hereinabove, a predetermined amount of raw coal into the retort 20 and

thereafter merely recirculating gas through the retort 20. In the event that no recirculation of semicoke has been effected, and the charge in the retort 20 consists entirely of raw coal, the temperature in the retort is maintained at from about 390° to 430° C. until the weight of the coal has been reduced by from 4 to 5% by removal of volatile matter. The temperature may then be raised to 450° to 500° or higher (up to 850° C.) until the desired devolatilization has been effected. If the charge in the retort contains at least 40% of semicoke, there will be no need for maintaining a temperature of not higher than 430° C. until 5% of the coal has been volatilized, but the temperature can be maintained at a higher level, if so desired.

When the above described apparatus is operated continuously, the valve in the branch conduit 38b may be set so that at least 40% of the semicoke is recirculated. In that event there is no need for maintaining the coal in the retort at a temperature not higher than 430° C. until the weight of the coal has been reduced by 4 or 5%. In the event the valve in the branch conduit 38b is closed, so that no recirculation is effected, it is necessary to maintain the coal introduced into the bottom of the retort 30 at a temperature not higher than 430° C. until at least 4 or 5% of the weight of the coal has been volatilized. This can be done, when the retort 20 has a diameter of six inches, by maintaining a temperature gradient in the lower six feet of the height of the retort of from about 400° to 430° C. The semicoke should leave the retort at a temperature of at least 450° to 500° C., although the maximum temperature may be as high as 850° C.

Recirculation or devolatilization to the extent of 4 or 5% at a temperature not higher than 430° C. serves to reduce agglomeration of the coal particles during carbonization. If normal agglomeration took place, the agitation of the coal particles by the gas traversing the coal in the retort would be prevented. Such reduction of agglomeration may also be effected by using, as agitating gas, hot steam (in place of coal gas). The same end may also be reached by incorporating with the coal, prior to introduction in the retort, a few per cent of sodium carbonate.

The carbonization is carried out to produce a semicoke having a volatile content of from 16 to 23%, and preferably from 17 to 18%, when the semicoke is to be used for the manufacture of metallurgical coke. One part of such semicoke may be admixed with two parts of raw Wheelwright coal and subjected to a high temperature coking process in a conventional coking oven.

When a domestic fuel is to be manufactured, the semi-coke may have its volatile content reduced to from 4 to 20% by the use of appropriate retort temperatures and time of exposure of the coal to such temperatures. The semicoke may be admixed with additional amounts of raw Wheelwright coal and then subjected to a medium temperature carbonization in a conventional coking oven, or the semicoke may be admixed with suitable proportions of a binder volatile at elevated temperatures and formed into briquettes having a volatile content determined by the extent of devolatilization of the semicoke and the amount of binder used.

The semicoke produced by the present method may also be employed for the production of fuel gas by complete gasification of the coal (water gas process). In that event, the volatile content

of the semicoke may be reduced to 20% or less in the low temperature carbonization.

The methods of this invention are not limited to the use of Wheelwright slack coal, but are applicable to all coking coals. For best results, the coal should be comminuted to a maximum particle size of 4 or, preferably, of 8 mesh, and, preferably, the particles should not be less than 140 mesh size, although, as illustrated by the specific examples described hereinabove, the process is operative with coal containing considerable proportions of particles smaller than 140 mesh. However, when particles smallers than 140 mesh are present, more recycling of the smaller particles may take place.

Low volatile coking coal breeze may be carbonized according to the present invention, and by the use of a binder volatile at an elevated temperature may be formed into briquettes of appropriate size and volatile content.

The maximum particle size of 4 mesh disclosed herein above assures that, when a gas traverses the body of coal in the retort, this body will be thoroughly agitated by the gas, with resultant efficient heat transfer from the gas to the solids and prevention of local over or under heating. Any inert or mildly oxidizing gas may be used. The use of a mildly oxidizing gas, such as steam cuts down the natural tendency of the coal particles to agglomerate. Further, agglomeration is reduced by the rapid flow of gas through the body of coal in the retort with resultant agitation of the coal.

Air may be used as an agitating medium at 350° C. or lower temperature. The coal may also be subjected to a mild preoxidizing treatment in order to reduce or cut down agglomeration.

It should be noted that in the low temperature carbonization method disclosed hereinabove, the formation of semicoke or char having any desired volatile content can be effected throughout temperature ranges wherein normally a volatile coking coal would agglomerate to an extent preventing agitation by a gaseous medium. The limited reduction of agglomeration necessary for the purposes of the present invention has been obtained by providing the coal in the form of particles not greater than 4 mesh size and by flowing through the body of coal a hot gas at such a rate as to keep the body of coal in constant agitation as well as by reducing the tendency of the coal to agglomerate by including with the body of coal being carbonized previously prepared semicoke or char or sodium carbonate, by effecting the removal of the first 5% of the volatile in the coal at temperatures below 430° C., by the use of steam as the agitating gas, or by mild preoxidation of the coal. At the same time, the coal is sufficiently plastic at some stage in the carbonization process to permit the entrapment of particles of iron ore or iron oxide, so that the bulk density of the semicoke can be made sufficiently great to prevent any segregation on subsequent high temperature carbonization in admixture with additional amounts of raw coking coal. The metallurgical coke thus produced has satisfactory properties and can be used, pound for pound, as a substitute for ordinary metallurgical coke without reducing the capacity of the blast furnace. In other words, I have succeeded in devising a process for the low temperature carbonization of coking coals in which the coal passes through a definitely plastic stage in comminuted form permitting entrapment of iron oxide particles but without

such agglomeration of the individual coal particles as would prevent agitation by a gaseous medium.

As pointed out hereinabove, many details of procedure and of construction may be varied within a wide range without departing from the principles of this invention and it is therefore not my purpose to limit the patent granted on this application otherwise than necessitated by the scope of the appended claims.

I claim as my invention:

1. A method for the production of metallurgical coke from a coking coal comprising providing said coal in comminuted form with a maximum particles size of 4 mesh, incorporating an iron oxide with said comminuted coking coal, agitating a body of the resulting mixture by flowing therethrough a stream of hot gas at a low carbonizing temperature, reducing the plasticity of said coal at said carbonizing temperatures to the point where said iron oxide particles will be entrapped by said coal particles without such agglomeration of said coal particles as would prevent said agitation, after said low temperature carbonization incorporating additional amounts of coking coal with the resultant semicoke, and subjecting the resulting mixture to a high temperature carbonization.

2. A method for the production of metallurgical coke from a coking coal selected from the group consisting of medium and high volatile coking coals and comprising providing said coal in comminuted form with a maximum particle size of 8 mesh, incorporating an iron oxide with said comminuted coking coal, agitating a body of the resultant mixture by flowing therethrough a stream of hot steam at a low carbonizing temperature, after said low temperature carbonization incorporating additional amounts of coking coal with the resultant semicoke, and subjecting the resulting mixture to a high temperature carbonization.

3. A method for the production of metallurgical coke from a coking coal selected from the group consisting of medium and high volatile coking coals and comprising providing said coal in comminuted form with a maximum particle size of 8 mesh, incorporating with said comminuted coking coal a semicoke derived from said coking coal by a low temperature carbonization in an amount sufficient to prevent agglomeration of said comminuted coking coal in the subsequent low temperature carbonization step, incorporating with said comminuted coal an iron oxide, agitating a body of the resulting mixture by flowing therethrough a stream of hot gas at a low carbonizing temperature, after said low temperature carbonization incorporating additional amounts of coking coal with the resultant semicoke, and subjecting the resulting mixture to a high temperature carbonization.

4. A method for the production of metallurgical coke from a coking coal selected from the group consisting of medium and high volatile coking coals and comprising providing said coal in comminuted form with a maximum particle size of 8 mesh, incorporating an iron oxide with said comminuted coking coal, agitating a body of the resulting mixture by flowing therethrough a stream of hot gas at a temperature of from 390° to 430° C. until 5% of said comminuted coking coal has been volatilized and thereafter at a temperature rising at least to 450° C. until a semicoke has been formed, after said carbonization incorporating additional amounts of coking coal with the resultant semicoke, and subjecting the resulting mixture to a high temperature carbonization.

5. A method for the incorporation of an iron oxide with a coking coal selected from the group consisting of medium and high volatile coking coals for the production of a ferruginous coke which comprises providing said coal in comminuted form with a maximum particle size of 4 mesh, incorporating an iron oxide with said comminuted coking coal, agitating a body of the resulting mixture by flowing therethrough a stream of hot gas at low carbonizing temperatures, reducing the plasticity of said coal at said carbonizing temperature to the point where said iron oxide particles will be entrapped by said coal particles without such agglomeration of said coal particles as would prevent said agitation, after said low temperature carbonization incorporating additional amounts of said coking coal with the resultant semicoke, and subjecting the resulting mixture to a high temperature carbonization.

6. The method for the production of semicoke from raw coking coal selected from the group consisting of medium and high volatile coking coals and comprising providing said coal in comminuted form with a maximum particle size of four mesh, incorporating a comminuted iron oxide with said coking coal to form a mixture containing from one-half to twenty percent of iron oxide and agitating a body of the resulting mixture by flowing steam through said body at a low carbonizing temperature whereby said iron oxide particles will be entrapped by said coal particles without such agglomeration of said coal particles as would prevent said agitation to yield a semicoke of increased bulk density and characterized by magnetic properties.

7. A method for the production of semicoke from raw coking coal selected from the group consisting of medium and high volatile coking coals and comprising providing said coal in comminuted form with a maximum particle size of four mesh, incorporating a comminuted iron oxide with said comminuted coking coal to form a mixture containing from one-half to twenty percent of iron oxide, and agitating a body of the resultant mixture by flowing therethrough a stream of hot gas at from 390° to 430° C. until about five percent of said coal has been volatilized and thereafter at a temperature rising at least to 450° C. until a semicoke has been produced whereby the comminuted iron oxide will be entrapped by the coal particles without such agglomeration of said coal particles as would prevent said agitation to yield a semicoke of increased bulk density and characterized by magnetic properties.

8. A method for the production of semicoke from raw coking coal selected from the group consisting of medium and high volatile coking coals and comprising providing said coal in comminuted form with a maximum particle size of four mesh, incorporating with said coking coal semicoke in an amount sufficient to prevent such agglomeration of said comminuted coking coal in a subsequent low temperature carbonization step as would prevent agitation of said coal by gas, incorporating with the resulting coal-semicoke mixture a comminuted iron oxide to form a mixture containing from one-half to twenty percent of iron oxide, and agitating a body of the resultant mixture by flowing therethrough a stream of hot gas at low carbonizing temperature whereby the comminuted iron oxide will be entrapped by said coal particles to yield a semicoke of increased bulk density characterized by magnetic properties.

9. A ferruginous semicoke characterized by magnetic properties and comprising ferruginous particles enclosed within semicoke particles, said semicoke containing ferruginous particles in an amount such that the bulk density of said semicoke is substantially in excess of 28 pounds per cubic foot.

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