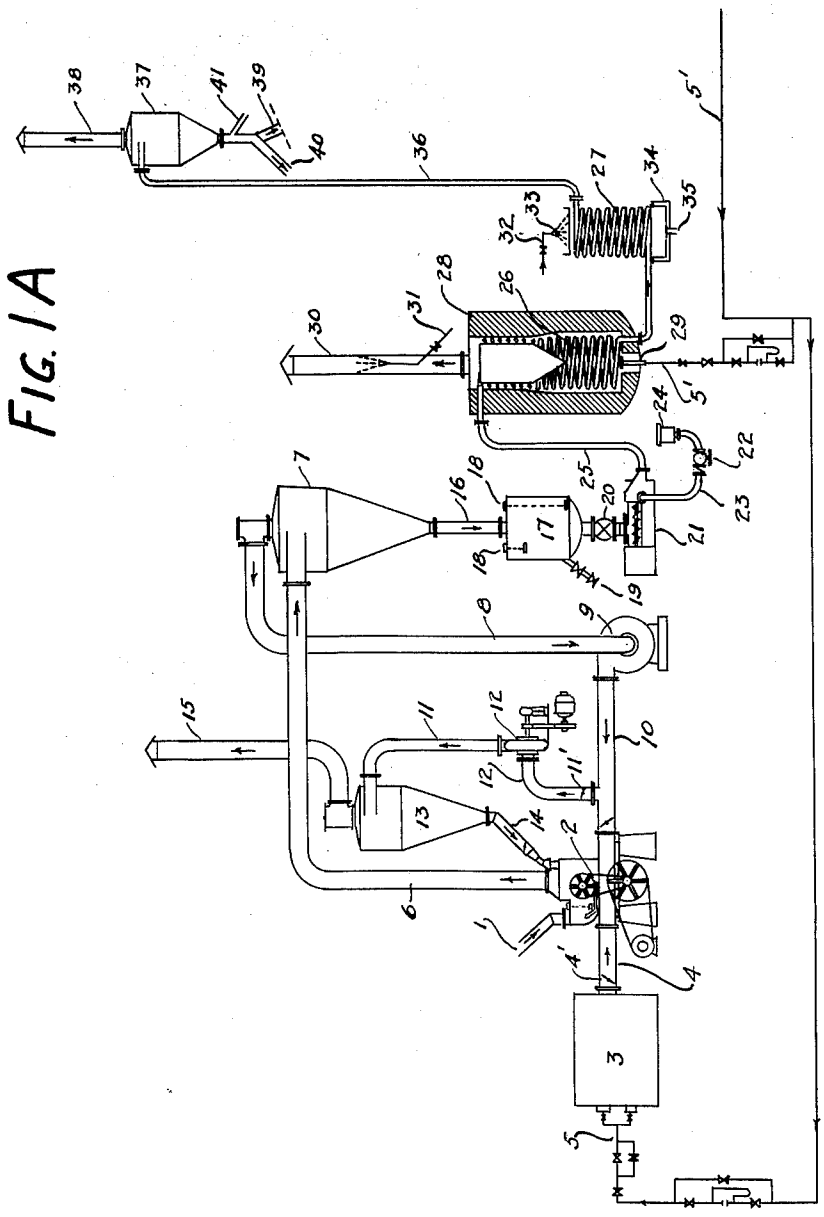
June 18, 1963 W. J. KRUPPA 3,094,467

CARBONIZATION OF COAL

Filed July 30, 1954 2 Sheets-Sheet 1

FIG. 1A



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CARBONIZATION OF COAL

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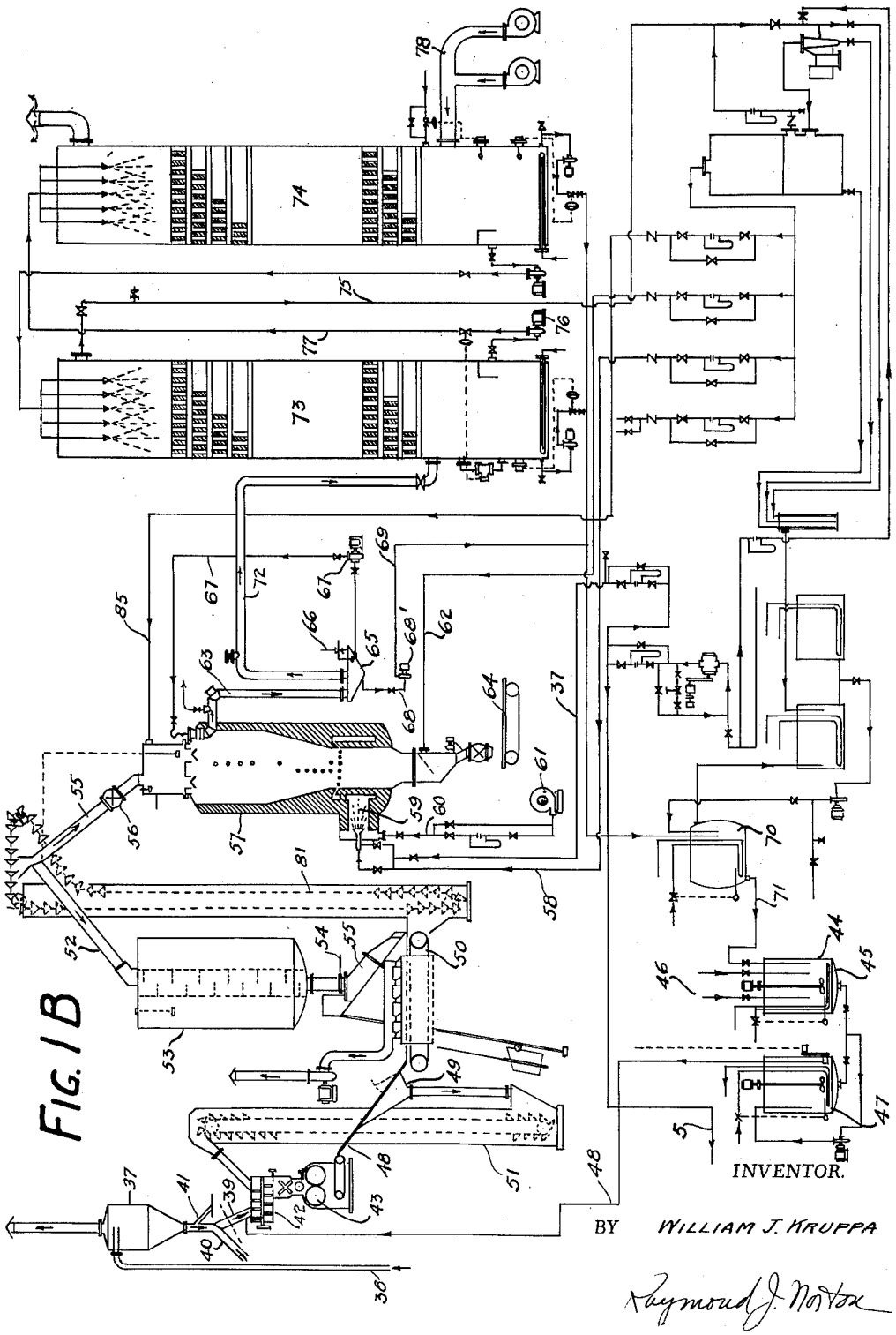


FIG. 1B

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3,094,467

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CARBONIZATION OF COAL

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11 Claims. (Cl. 202—22)

This invention relates to the carbonization of coal, more particularly to a method of producing high grade metallurgical coke while concomitantly producing high yields of valuable liquid products.

The metallurgical coke employed in industry is largely produced in by-product coke ovens; but a minor amount is produced in beehive ovens. As is known, in the by-product oven, the coke is produced by the high temperature distillation of selected grades of bituminous coal in a closed retort without access to air. The primary product of such an operation is metallurgical coke and the charge material is selected and the operating conditions adjusted so as to produce a hard, porous coke suitable for the reduction of iron ore in a blast furnace. The co-products of the operation are coal tar, coal gas and ammonia. The desideratum in such operations is a high quality coke and this results in a relatively low yield of liquid products and a high yield of gas. Because of the high temperatures employed, necessitated by the indirect heating utilized and to the physical structure of the ovens, the originally evolved, volatile materials are subjected to excessive pyrolysis and hence the ultimate liquid products produced are comprised largely of heavy, residual products of a tarry nature.

The physical structure of the by-product coke oven and the nature of the operation impose certain restrictions on the charge material, that is to say, to selected grades of coal that do not unduly swell or cake. There are many grades of bituminous coals that swell and cake badly but which are otherwise suitable for the production of metallurgical coke and other products such as partially coked briquettes.

The present invention relates to an improved method of producing an excellent grade of metallurgical coke from bituminous coal and is of particular economic value because it may utilize a wide range of bituminous and particularly badly swelling and caking coals which cannot be utilized in a by-product coke oven.

In order to more clearly explain the invention, a preferred embodiment of a process unit will be described and is illustrated in the accompanying drawings in which:

FIG. 1A is a view, partially in elevation and partially in vertical section, of the coal preconditioning unit.

FIG. 1B is a similar view of the connected units in which the preconditioned coal is briquetted and carbonized.

The process of the invention, as will be seen more fully hereinafter, combines the advantages of the high temperature by-product coking process and the low-temperature carbonation method in that, like the by-product oven operation, it produces an excellent grade of metallurgical coke and as in the earlier low-temperature carbonization operation, it insures a large yield of primary tars. It also produces appreciable amounts of gas of substantial fuel value. The novel process of the invention is designed to economically process a wide range of bituminous coal, operating efficiently on grades from those having slight coking properties to those characterized as badly swelling and caking coals.

Considered generally, the process comprises a special preconditioning of the coal in finely divided form to modify its caking and swelling properties and to improve the coal in other particulars; briquetting or extruding the

coal with an improved binder derived from a coproduct of the process and distilling and carbonizing the briquettes by direct contact with a stream of hot recycle gas from which the originally evolved condensables have been removed.

It has long been known that weathering reduces the swelling power of bituminous coals and such reduction could be achieved by accelerated oxidation employing heated oxygen containing gas or other oxidizing agents. The oxidation methods proposed heretofore, generally considered, contemplated a treatment at relatively low temperature for prolonged periods of time as, for example, at a temperature of around 150° C. for a period of from 24 hours to 120 hours or more, depending on the type of coal processed. Such earlier treatments presented many disadvantages included in which was the excessive storage capacity required for treatment and the batch nature of the operation. The prior art discloses many suggestions as to methods of shortening the oxidation treatment such as, carrying out the oxidation under superatmospheric pressures of the order of 50 to 150 p.s.i. and temperatures of the order of from about 150° C. to 200° C. or by subjecting heated coal alternately to pressure and vacuum. Such methods, however, still entail a relatively prolonged treating period of several hours and are essentially batch operations requiring retention periods in expensive pressure tight containers of substantial capacity.

The present process invokes and utilizes a novel coal preconditioning treatment. It is found that when this is properly correlated with proper briquetting or extrusion techniques, an improved carbonization of a wide permissive range of charge material may be effected to produce a superior grade of metallurgical coke and a high yield of valuable co-products.

The method of preconditioning the coal is based on the concept of oxidizing the coal at high temperatures, but for a very brief interval of time, followed by immediate, rapid cooling of the heated coal to stop the oxidation. This preconditioning essentially is a continuous flash oxidation conducted on particulated coal fluidized in the oxidizing gaseous medium which conveniently is ordinary air, followed by a flash cooling of the oxidized coal.

As will be more fully appreciated from a consideration of the subsequent disclosure, the novelty of the present invention is the more arresting because the basic procedure is contra-indicated in the prior art. As will be explained more in detail, the oxidation is carried out in the presence of an oxygen-containing atmosphere such as air and the coal is heated in the presence of such oxygen-containing gas to temperatures to and above the threshhold ignition temperature. In these circumstances complete combustion of the coal, rather than controlled oxidation, would be expectable. However, as will be more fully elucidated, this high temperature oxidation is carried out under such conditions of high thruput velocity and rapid quenching that the retention or dwell period at the high temperatures is exceedingly brief, and of the order of 10 seconds, more or less, so that actual ignition does not ensue. Another characteristically novel factor or condition in the process is that the particulated coal is suspended in high concentration in an air stream and is heated in the so-called plastic range so that the actual oxidation of the coal is largely effected while the coal is in a special physical condition or state, namely as a viscous plastic or quasi-liquid particles dispersed in a gaseous magma. Such a procedure, per se, i.e., heating powdered coal to the viscous or plastic range, would be expected to, and normally would, result in the agglomeration of the plasticized coal particles into larger cake-like

bodies. However, in the novel procedure, this high temperature treatment is carried out on the solid or plasticized particles of coal which move at a very high veolcity in a constricted path of confined cross-section and while maintained in a constricted path are shock cooled or rapidly quenched from the platsic to the solid phase. In these circumstances but very little agglomeration of the coal particles occur as the mesh size of the oxidized quenched coal particles is only slightly above that of the charge stock. This retention of the desired particle size is due, among other things, to the fact that the coal particles, whether in solid or semi-fused form, constitute a dispersed phase in a high velocity air stream; to the low retention period which the coal is in the plastic state and probably to some considerable degree to the mutual attrition of the resolidified coal particles which occurs in the rapidly moving fluid stream.

This method of preconditioning has many advantages and, as will be seen, lends itself ideally to the continuous production of metallurgical coke. Inasmuch as the thermal preconditioning treatment is but for a very brief period of time and the retention period of the coal in the heating apparatus is extremely short, a very large thruput is achieved in a relatively small oxidizing unit. This method of flash oxidation in the plastic range of the coal and quick cooling thus eliminates the necessity for the large and costly pressure equipment required by earlier proposed methods.

This coal preconditioning treatment insures many other advantages which are of peculiar and special import with respect to the character of the coke ultimately produced. The flash oxidation not only reduces the swelling and caking properties, but also, with most types of coal, unexpectedly lowers the sulfur content of the coke produced from the oxidized coal. This is of very considerable significance in the production of metallurgical coke because, as is known, the presence of sulfur in the coke is detrimental, especially when the coke is used in blast furnace reductions. In such blast furnace operations, it costs approximately about twenty-five cents to remove each tenth of a percent of sulfur, this cost being represented by the extra coke and limestone required.

Another advantage of the flash oxidation method is its effect on the ash content of the ultimately produced coke of the treated coal. When such preconditioned coal is employed in the present invention, the finished coke shows a lower ash content than a coke produced from coal which is not subjected to such preconditioning treatment. Another factor of not inconsiderable significance, is that the softening temperature of the ash in the flash oxidized coal is substantially higher than the ash of the same coal which is unoxidized, for example, it was determined that the softening temperature of the ash of a sample of raw Heisley coal was 2320° F. whereas this same type of coal which was subjected to flash oxidation was found to have the softening temperature of the ash at 2570° F. The reason for this reduction in the ash content as effected by the flash oxidation is obscure, and difficult to precisely explain. It may be due to a physical separation of some of the ash forming constituents as a result of a winnowing effect taking place in the turbulant stream of the triturated coal or possibly to a chemical conversion of metaliferous components to a more evolvable form. Whatever may be the mechanism of reaction, the described beneficial reduction in ash is accomplished by the flash oxidation.

There is another inexplicable action which takes place during or because of the flash oxidation, and that is the reduction of the critical character of the rate of heat input during the so-called critical range in the heating of the briquettes or extrusions in the carbonizing retort. This critical range may be defined as the temperature range extending from approximately 350° C. to about 500° C. in which softening or fusion of the coal takes place and in which upon evolution of volatile material the coal mass resolidifies. In the past it was thought that the rate of heat input to the coal compacts in this range had to be carefully controlled and the limit of safety was of the order of about 2.5° C. per minute; if the rate of temperature rise, in this range, materially exceeded this rate, briquettes, for example, tended to distort and adhere to each other and in addition suffer a loss in head load strength. This critical effect was found, in the past, to be somewhat reduced by forming briquettes of pulverized coal of minus 20 mesh. It was discovered that when the coal from which the briquette or extrusion was formed was first subjected to the novel flash oxidation, this critical effect was mitigated to a surprising degree, so much so, in fact, that the critical character of the range does not obtain to the same extent when carbonizing the flash oxidized coal. Operations have been carried out, in the manner to be more fully described hereinafter, in which briquetted or extruded flash oxidized coal has been carbonized and in which the rate of heat input within the range 350° C. to 500° C. was of the order of up to 5° C. per minute. This, as will be appreciated, is most important, because in earlier operations the slow rate of heat input during the plastic or critical range was a limiting factor on the thruput of coal preforms in the retort. There is thus an intimate and unobvious correlation between the type of flash high temperature preconditioning of particulated coal and the retort operation which was unappreciated and unutilized heretofore.

In carrying out the process, the bituminous coal or blends or mixtures of selected grades of coal are dried and pulverized prior to the oxidizing treatment. It has been ascertained that the presence of water vapor in the oxidizing gas tends to somewhat retard the oxidation of the coal. It is thus desirable to dry the coal particles to less than one percent free moisture.

The particle size of the coal to be treated, while not critical is quite important. It has been found that improved results are assured by pulverizing the coal so that 100 percent is minus 20 mesh and at least 90 percent is minus 28 mesh. Controlling the mesh size is quite important since it has a direct bearing on the flash oxidation treatment and on the physical character and thermal behavior of the subsequently formed briquettes. If the coal particles are too large, it is more difficult to achieve uniform oxidation and the time required for satisfactory oxidation is prolonged, furthermore, the large coal particles result in a sandy texture of the raw briquette or other preform and a reduction in abrasive strength of the briquette.

In operating the process, the coal or blends of coal to be treated are continuously taken from a suitable storage bin (not shown) and fed through the conduit **1** to any suitable unit in which the coal is particulated and dried to the desired degree. This obviously may be done in different specific manners utilizing any suitable apparatus. Thus the coal may be ground to a particle size suitable for drying, using any suitable type of mill such as a hammer mill, dried in an economical dryer such as a rotary kiln dryer and then passed to an air swept pulverizing mill. In the illustrative embodiment the coal is pulverized and dried contemporaneously. The coal passes from the conduit **1** to a suitable mill such as the roller mill **2**. In lieu of the roller mill, any other effective triturating apparatus may be employed.

In the embodiment depicted in the drawing, the coal which is pulverized in the mill **2** is picked up in a stream of a hot drying medium such as hot gas admitted from the furnace **3** through the conduit **4**, the flow of which is controlled by damper **4'**. The furnace **3** may be of any desired type but in the preferred embodiment, i.e., one in which coal is continuously preconditioned, briquetted or extruded and carbonized, the furnace conveniently is a gas fired type the fuel for which is fuel gas derived in the carbonization treatment which, as shown, may be fed to the furnace in controlled amounts through the gas

feed line 5. This line preferably is provided with a flow meter and thermostatically controlled feed valve so as to establish and maintain the temperature of the exit gases at the desired value.

The pulverizer, as shown, is connected in a classifier circuit which includes the discharge conduit 6, a solids separator such as the cyclone 7, the fluid recycle conduit 8, circulating fan 9 and the return conduit 10 which discharges back into the mill 2. The operation of this air swept mill is apparent. When the unit is on stream, the fine coal particles formed in the mill, i.e., those less than about 20 mesh, are entrained in the stream of hot gas entering from the furnace and are forced by circulating fan 9 through the discharge conduit 6 to the cyclone 7. The larger coal particles drop out of the circulating stream and are further reduced in the mill. In the cyclone 7 most of the suspended coal particles are separated and the hot gas largely freed of particulated coal is returned to the circuit through line 8.

As shown, the pulverizing-drying unit also includes a vent circuit. This includes line 11 controlled by valve 11', having the interposed motor drive vent fan 12 which line connects at one end with recirculating conduit 10 and at the other with a fluids-solids separator such as the cyclone 13. The solids separated out in cyclone 13 are returned to the mill through a suitable return line 14 and the solids-denuded gas is vented from the system through vent line 15. During operation the positively driven vent fan 12 removes heated gas from the main recirculating circuit, in amounts controlled by valve 11' and discharges this gas into the cyclone 13. Herein the entrained coal particles are separated and returned to the mill and the waste gas is discharged through vent 15 to atmosphere. As will be appreciated during the operation, the vent circuit is controlled so as to discharge or vent from the system a quantity of gas equal to that admitted from the furnace 3 in addition to the water vapor evolved from surface moisture and some water of constitution in the coal.

The above described pulverizing and drying unit produces a suitably pulverized dried coal adapted for the improved preconditioning treatment. As explained previously, such pulverized dried coal may be prepared in any other suitable manner. The hot dried coal which collects in the low sections of the cyclone 7 is fed through the discharge line 16 to a unit in which the coal is fluidized prior to the flash oxidation. The coal removed from the cyclone 7 is discharged through line 16 into an accumulator or feed tank 17 which in turn feeds the coal to any suitable fluidizing unit. This feed tank 17 is desirably provided with suitable level indicators 18 and with a pressure indicator (not shown). The tank also preferably is provided with manually controlled valved draw off line 19 from which coal may be withdrawn for current tests and checks.

The coal may be fluidized in any suitable type of fluidizing unit. In the descriptive embodiment the unit is illustrated as a conventional Fuller-Kinyon pump. In the continuous operation coal is fed from the feed tank through any suitable pressure retentive valve such as the rotary vane valve 20 to the fluidizing pump 21. This coal is transported by a continuous screw to the discharge side of the pump and in transit is aerated and fluidized by air forced into the pump from the compressor 22 through the line 23. The air feed to the compressor is preferably filtered in filter 24. The fluidized coal, as shown, is discharged from the pump into transfer line 25 which is connected to the flash oxidizing zone. As will be understood, the pump inlet line 23 and discharge line 25 are preferably provided with suitable pressure indicators.

It has been found during test operations of the process that the quantity of air required for adequate aeration and fluidizing of the coal may vary somewhat. Effective oxidizing has been obtained using a ratio of substantially 8 lbs. of coal per pound of air, however, higher ratios of the order of about 12 to 15 lbs. of coal per pound of air may desirably be employed.

In the preferred oxidation treatment, the fluidized coal is passed in a continuous stream through an elongated confined passageway and is rapidly heated to a high temperature in one portion of the passageway and is then immediately quickly cooled or quenched in an adjoining portion of the passageway.

As shown in the drawing, the flash oxidation and flash cooling can most conveniently be effected by passing the fluidized coal through a continuous coil. The fluidized coal, passing from the pump 21, is forced through transfer line 25 into the heating coil 26 and thence through the connected cooling coil 27. The heating section of the coil, as shown, may be located in a suitable furnace 28 and desirably in the combustion zone so as to insure rapid transfer of heat to the coal stream; preferably the stream of fluidized coal passes through the furnace in a direction countercurrent to the flow of combustion gases. As will be understood, the furnace 28 may be of any desired type and conveniently may be a gas fired furnace the fuel for the burner 29 of which is fuel gas derived from the subsequent carbonization operation and fed through line 5'. As this line 5' previously described is provided with a suitable flow meter and temperature recorder so that the operation may be effectively, currently checked.

The flue gas from the furnace passes out through stack 30 under a draft created by steam or water injected into the stack through valve-controlled line 31.

The heated fluidized stream passes immediately and continuously from the coil 26 to the quench coil 27. Preferably the fluidized stream, as shown, passes upwardly in coil 27 and during such passage is quickly cooled in any suitable manner as by means of a spray of water or other coolant fed through the line 32 and spray head 33, immersion in a tank of a coolant or by any other effective means.

When the spray cooling technique is employed, the used cooling water may be collected in the tank 34 from which it may be withdrawn through line 35.

The cooled fluidized stream of oxidized coal passes continuously from the coil 27 through the transfer line 36 to a fluid-solids separator such as the cyclone 37 in which the oxidized coal particles are separated from the entraining gas and water vapor which latter are vented to atmosphere through the stack. The cooled oxidized coal particles accumulating in the cyclone 37 may be drawn off through the line 39 (FIG. 1B) and passed directly to the briquetting or extrusion unit of the plant or, when required, all or a portion of the coal may be directed through line 40 to storage from which it may later be withdrawn for the fabrication of briquettes or extrusions. The cyclone preferably is provided with the sample draw off line from which samples may be secured for periodic check tests.

The improved preconditioning of coal, as effectuated in the described apparatus can be more readily appreciated and evaluated from a consideration of a typical operation. A large number of runs have been carried out in a unit such as that described, having a thruput capacity, through the oxidizing zone, of 13 tons per hour. In this unit the oxidizer coal 26 consisted of 600 linear feet of 6 inch diameter alloy steel pipe and the connected cooling coal 27 was of the same size and length. In the usual operation when the unit is on stream, the temperature of the drying air passing through the furnace 3 to the pulverizer is controlled at about 500° F. The furnace 28 is operated so as to raise the temperature of the fluidized coal stream to between about 675° F. and about 800° F. and desirably between 700° F. and 750° F. The pressure on the fluidized stream at the exit of pump 21 may be between about 20 and 25 lbs. per sq. in. In such circumstances the actual velocity of the coal stream at the exit of the fluidizer pump ranges from about

1200 to 2000 or more ft. per min. With such velocity in an oxidizer coil of the dimensions given, the retention period of the coal is only about 10 to 15 seconds. As will be appreciated during passage through the oxidizer and cooling coils the pressure progressively diminishes and is reduced to substantially zero pressure at the cooler.

The cooler is operated so as to very rapidly reduce the temperature down to the lowest convenient value to check the oxidation reaction and to cool the coal to a degree sufficient to insure convenient subsequent handling. The fluid stream, passing through the cooler coil, as will be understood, contains water vapor evolved from water of constitution and partial decomposition of the coal.

The rapid chilling or cooling of the oxidized particulated coal, as previously pointed out, is a very important feature of the invention. If the particulated coal which is highly heated in coil **26** is not cooled quickly, over oxidation of the coal would result thus negating the advantages of the treatment. In the treatment described, the amount of oxidation is controlled within selected narrow limits. Thus in the specific example an analysis of the exit air (from cyclone **37**) shows an oxygen content of 16% so that but approximately 5% of oxygen in the air carrier stream is utilized in the controlled oxidation.

It will be understood that the requirements that the particulated coal be flash heated and flash cooled in a brief period of time imposes some limitations on the diameter of the coil, i.e., the cross-sectional area of the air-coal mass to be heated and cooled. It has been found that for effective commercial operations the heating and cooling coils should not be materially below two inches and not substantially more than six inches internal diameter.

It will have been appreciated that the improved method of preconditioning bituminous coal to alter its swelling and caking properties essentially involves extremely rapid or flash oxidation in a gas stream of substantial oxygen content at elevated temperatures at or above the threshhold ignition temperature of the coal and at or above the plastic range of the particulated coal. This flash heating and oxidation is instantaneously followed by a flash or shock chilling of the confined stream of dispersed particulated coal to abruptly check or inhibit further oxidation. The apparatus shown in the drawings is designed and has been found to operate effectively to achieve the desired results. It is apparent, however, that other and specifically different designs and types of apparatus of a functionally equivalent nature may be utilized in effectuating the process. Thus while the illustrative embodiment involves a particular pulverizing and drying circuit in which the coal is pulverized in an atmosphere of drying gas, it is apparent that the same results may be achieved by specifically different methods employing apparatus other than that shown herein. Thus the raw coal may be preliminarily crushed in any suitable crushing apparatus, dried in any suitable drying unit, such as a rotary kiln dryer, and then fed to the pulverizing mill which may or may not be air swept. Other apparatus and methods for producing a dried, suitably particulated coal are within the compass of the invention.

In the course of study and investigation of the effects of the described flash oxidation treatment on different types of bituminous coals and various blends of such coals, it was ascertained that, with some rare exceptions, coke which is made from compacted units of particulated coal oxidized by the described procedure and carbonized as hereinafter described displayed a significantly lower sulfur content than coke produced from the same coal which was not preoxidized. It was similarly ascertained that coke made from the oxidized coal, in all cases, showed less ash content than coke made by the same procedure from the same coal which was not preliminarily oxidized.

The above mentioned reduction in the sulfur and ash content of the coke produced from oxidized coal is shown in the following table. The results recorded are from runs in a semi-commercial unit in which coal blends were flash oxidized and flash cooled in accordance with the described procedure and the oxidized coal was briquetted using a sulfuric acid-treated tar binder, in accordance with U.S. Patent 2,314,641 using approximately 11% of binder on the coal. The briquettes were carbonized in a vertical retort under the conditions prescribed in the U.S. Patent to Berry 2,131,702. The results recorded in Table I represent the analysis of composite samples of a one day run and a five day run.

TABLE I

Typical Analyses of Solid Materials

COMPOSITE SAMPLES OF ONE DAY'S RUN—FIN. TEMP. COKE 830° C.

	Raw Coal	Pulv. Coal Grab Sample	Oxid. Coal	Raw Briq't	Coke
Vol. Matter (Dry)	18.39	18.13	17.67	24.39	0.75
Fixed Carbon	74.85	75.57	77.05	69.92	91.97
Ash	6.76	6.10	5.28	5.69	7.28
Sulfur	1.40	1.23	0.95	1.31	1.04

COMPOSITES—FIVE DAY'S RUN

Vol. Matter (Dry)	18.41	18.02	17.50	24.12	0.89
Fixed Carbon	74.75	74.68	77.02	70.46	92.85
Ash	6.84	7.30	5.48	5.42	6.86
Sulfur	1.58	1.65	1.26	1.53	1.03

The raw briquettes designated in the table contained 11% tar pitch binder which contained approximately 10% of sulfuric acid.

A study of the data recorded in the above table reveals the striking effects of the flash oxidation treatment on the ash and sulfur content of the oxidized coal and finished coke. The ash and sulfur content of the oxidized coal is significantly lower than that of the raw coal and grab samples. Notable, too, is the fact that the softening temperature of the ash in the oxidized coal is measurably higher than that of the raw coal.

The particulated coal processed as above described by flash high temperature oxidation and flash or shock cooling may be formed into units of the desired shape and size, by any suitable procedure such as briquetting or extrusion, and may be then carbonized in a continuous retort to a metallurgical grade coke. Since the optimum conditions for briquetting the particulated coal and extruding preforms of such coal vary somewhat, these procedures will be separately described.

In one procedure under the invention, the oxidized particulated coal is withdrawn from line **39** (or from storage) and passed to a mixing unit where the coal is thoroughly mixed with a hot binder and is passed to the briquetting machine **43** of any standard or conventional type. One type of binder which has been found to be effective comprises a heavy tar or pitch fraction derived wholly or in part from a subsequent distillation of products evolved during the carbonization of the coal and charged to the binder make-up tanks **44** and **47**. This binder may be chemically modified by the addition of from about 8% to about 15% of sulfuric acid admitted through lines **46**. The acid is thoroughly mixed with the binder by suitable agitating paddles, as shown, and retained at the desired elevated temperature by control of steam through the steam coil **45**. The pre-mixed binder is passed from mixer **44** to the heated binder supply tank **47** from whence it is withdrawn, in the desired amount, through line **48** and is discharged to the mixer **42**.

The hot fluent binder is thoroughly mixed with the particulated coal in mixer **42** in amounts sufficient to produce briquettes of the desired characteristics of high raw and coked strength and good abrasive resistance. In

usual circumstances, from 5% to 10% of this binder is employed in the briquetted coal mixture. If desired, a corrosion inhibitor, such as potassium dichromate, may be added to the binder to inhibit acid corrosion of the briquetting rolls. While such an acid treated binder does serve effectively, it does have the disadvantage of requiring the mixing of acid and does present a corrosion problem. The preferred binder, to be described subsequently, comprises an acid-free pitch product which may be derived wholly or in part from distillation products of carbonization.

The binder-coal mix, as noted, is passed to the briquetting rolls and briquettes are discharged from this press usually at a temperature of from about 40° C. to 95° C. At this stage, the binder is relatively soft. The hot briquettes are then passed over a chute 48 provided with a screen 49 to the conveyor 50. Fines passing through screen 49 are returned by a conveyer 51 back to the mixer 42. In passage along the conveyer, the warm briquettes are cooled with minimum tumbling or movement down to a temperature of between about 30° C. to 40° C. at which temperature they are hard enough to withstand normal handling, i.e., transportation by typical conveyors such as elevator conveyor 31 for discharge into the carbonizing retort. Any excess of briquettes above that required for a retort charge may be directed to temporary storage, as for example, by diversion through chute or passageway 52 to storage bin 53 from which they may be charged as required through gate valve 54 and conduit 55 back to conveyor 50.

The briquettes which are to constitute a retort charge are fed by gravity through the line 55 controlled by valve 56 to the vertical retort 57.

In the carbonization operation, the briquettes pass by gravity continuously down the retort and during such passage are subjected to direct intimate contact with a countercurrently flowing stream of non-reactive gas wherein they are carbonized to a finishing temperature of from about 850° C. to 1000° C. or more to produce tough, dense carbonized units of the desired low volatile content of the order of 1% to 2%. Such finishing temperature, as will be understood, is chosen in relation to the desired reactivity and volatile content of the coke.

The heating gas stream in the retort is made up, in effect, of two merged streams which are derived from the carbonization operation and which have been stripped of condensable components. One such gas stream is fed through line 58 to a combustion chamber 59 where it is heated by partial combustion in the combustion chamber with air introduced through line 60 by the action of blower 61 and after such partial combustion enters the tuyeres.

The second stream of gas comprises a quench gas which enters the lower or quench section of the retort through line 62 in controlled quantities. This gas directly contacts and abstracts heat from the hot carbonized briquettes, being thereby preheated, and in passing upwardly in the retort merges or mixes with gas entering through the tuyeres and the merged streams pass upwardly in the retort. The exit gases pass out of the upper section of retort through the main 63 and are treated in any desired manner to remove the tar and to provide the desired tar-denuded gas for recycling to the retort.

When bringing the retort up to temperature, preparatory to putting it on stream, the gas feed line 37 may be employed since for such preheating operation the volume and temperature of the tuyere gas is not important.

In this operation the heating of the briquettes or extrusions is effected in a progressive manner, as they pass down the retort, by direct contact with relatively large volumes of stripped recycle gas. In such passage the units are subjected to an increasing temperature gradient during which passage condensable volatile material and fixed gases are evolved. In the operation of the process, the quantities of gases fed through the lines 58 and 62 and the temperature of the tuyere gas are so correlated and controlled, that the volume of gas passing upwardly in the retort has a heat capacity which substantially balances that of the charge of briquettes or extrusions being heated. In the passage down the retort the compacted units are subjected to increasing temperatures in an atmosphere of hot gas which is characterized by a very low partial pressure of the volatile or evolvable components. The heating is controlled at such a rate as to avoid distortion and cracking of the compacted coal units. In the lower section of the retort, as shown, the compacted units are subjected to the desired high finishing temperature of from 800° C. to 1000° C. and are thereafter quenched by the stream of quench gas and are discharged to conveyer 64 and conveyed to storage. The upwardly flowing gas sweep out evolved tars and lighter vapors into the upper section of the retort thus minimizing excessive pyrolysis or cracking of such evolved products.

An additional or supplemental stream of hot product gas may be advantageously utilized in the retort. This stream is fed to the upper section of the retort through the line 85 after being preheated to a temperature of the order of from 200° C. to 400° C. The sensible heat of this gas stream is imparted to a substantial degree to the incoming coal units, preheating such units and at the same time minimizing or inhibiting the condensation of tar from the tar-containing vapors; such condensation normally would occur if such vapors contacted cold briquettes or extrusions. Such entering stream of flush gas enters the retort countercurrently to the main upwardly flowing merged gas stream and buffers or checks the flow of the latter into the top of the retort where condensation of tar might otherwise occur.

The vapor-laden gases pass through the main or downcommer 63 to any suitable tar settler or knockout box 65. Preferably the effluent stream of gas discharged from the retort is quickly cooled by any suitable coolant, such as water, which is admitted to the effluent stream through the line 67. This coolant, which can comprise a supernatant liquid layer from knockout box 65, may be continuously recirculated by withdrawing it from the box 65 and forcing it by pump 67′ to the main 63. The make-up coolant, in required amounts, is admitted through line 66.

The liquid tar fraction, settling out in unit 65, is discharged through line 68 and is forced by pump 68′ through line 69 and after suitable heating, for example in heated still 70, a portion may be utilized for the binder requirements for the binder-make-up by passing the desired fraction from still 70 through line 71 to the binder make-up system.

The gas, from which the entrained tar has been scrubbed, is passed from knockout box 65 through the main 72 and is treated by any suitable or conventional method as, for example, in the scrubbing tower 73 to separate additional quantities of tar and to separate and condense the condensable components. The gas separated in such operations and withdrawn from tower 73 through line 75 is split into several streams to serve as a source of recycle gas for the carbonization operation and if desired for heating gas, passed through line 5 to the coal heating furnace. This recycle gas, when desired, may be supplemented by gas introduced into the cycle from an extraneous source. The scrubbing liquid collecting in the base of tower 73 is withdrawn and passed through pump 76 and line 77 to the top of tower 74, where it is regenerated for reuse by contact with air introduced through line 78.

The carbonization retort employed may be of any suitable design and construction and of the desired capacity. The retort shown comprises essentially a steel shell provided with a refractory lining, such a firebrick. The steel shell may be lagged on the exterior with suitable insulating material. If desired, however, the retort may be of any other desired shape. In a typical case, a 200 ton a day production may be carried out in a

round retort of substantially twelve feet in diameter and of the order of about thirty feet or more in height.

The product produced by the above described method of flash oxidation of particulated bituminous coal followed immediately by shock cooling, subsequent compaction of the oxidized coal into units of predetermined size and shape, and the controlled carbonization of these units in the manner described, results in coke-like products of eminent utility. The carbonized units are uniform in size and composition and are of controlled density thus assuring uniform operation in use, as for example, in a blast furnace or cupola operation. The size and shape of the fuel units readily can be controlled to insure the desired or requisite permeability of the bed to the passage of gases therethrough.

In the preceding description, the novel operation of the invention was described, illustratively, with reference to the briquetting of specially preconditioned coal with a specific binder such as sulfuric acid-treated coal tar pitch. This described operation has been found to be quite effective. However, the permissive scope of the invention extends considerably beyond this particular operation. The special preconditioning of the coal has been found to so modify the coal that it is rendered amenable to bonding with other and specifically different binders and utilizing different techniques for compaction of the coal.

Continuing research on the problem of compacting oxidized coal particles and preforming these into self-sustaining units of sufficient strength to undergo or withstand the conditions incident to carbonization has eventuated in certain novel findings of profound technological value. As will be explained more fully hereinafter, it has been found, among other things, that the head load strength of briquettes and extrusions may be considerably increased by the incorporation of certain water-soluble binders in the main binder pitch mix. It has also been ascertained that while acid-treated binders operate effectively, the addition of acid is not essential. It has been found that effective bonding of the particulated coal may be achieved with acid-free coal tar or pitch binders, provided certain conditions are established and maintained. It has further been determined that by proper control of binder-to-coal ratios, water content and aqueous wettability of the coal particles, coal-binder mixes may be produced which are readily extrudable to produce extruded units of excellent green strength and which can be carbonized, under the general conditions previously described, to produce strong, tough coked units of predetermined shape and controlled density which perform excellently as metallurgical coke in reduction operations involving the use of such coke.

One of the important factors in the successful operation of the process is the development of adequate headload strength in the briquettes or other preformed units. It has been found that the headload strength of preforms of compacted oxidized coal is somewhat limited. Such headload strength may be increased by invoking a number of expedients. Evidence, adduced in operation of the process, indicates that the failure of briquettes or other preforms, under headload, apparently is due, among other things, to the fact that because of the general homogeneity of the structure of the briquette lines of cleavage tend to develop in the units which cleavage lines or planes induce or facilitate fracture of the units. It has been determined that this tendency can be substantially minimized by incorporating in the coal-binder premix a heterogeneous and preferably a substantially inert solid phase. This apparently serves as an aggregate which imparts considerable strength to the compacted unit. Such an additive or aggregate may comprise, for example, a selected amount of up to the order of 25% of anthracite fines, coke breeze, over oxidized coal and the like.

The headload strength of green briquettes and other preforms can also be measurably improved by utilizing a small amount of a water-soluble binder, and of the order of two percent, more or less, of the weight of the coal. Such water-soluble binder may comprise starchy or farinaceous materials, waste sulfite liquor, molasses and the like. While the rationale of their function or action is not fully comprehended, it has been found in actual operation that they significantly improve the headload strength of briquettes or other preforms in which they are incorporated.

It has been found that certain variables quite profoundly affect the character of the binder-coal mix and the physical properties of the briquettes or other preforms molded from such a mix. It has been ascertained that the moisture content of the coal and the proportion of binder to coal are of salient importance in the production of briquettes which behave satisfactorily in the subsequent carbonization operation. Similarly, as previously pointed out, the heating rates during the carbonization operation are of major importance and are intimately correlated with the character and structure of the preformed units.

As a result of extensive investigation of the several variables or factors which might or could influence the quality of the briquettes, it was determined that the optimum condition for the fabrication of briquettes of the desired characteristics involved in careful control of a number of conditions.

It was found that the moisture content of the coal to be extruded was of major significance. It was learned that such moisture content should be held within the range of approximately 5% to 8% and that improved results could be achieved by introducing a small amount of an effective wetting agent that lowers the interfacial tension between the coal particles and the binder magma.

It was also determined that the relative quantity of the binder in the permix was a very important factor. It was found that excellent preforms could be made by employing a relatively low melting point coal tar pitch (100° F.M.P.) and using this in the preparation of between about 12% and 15% on the weight of the coal.

While the addition temperature of the pitch and the temperature of mixing are not critical, it has been found from experience that adding the pitch at a temperature of between about 325° F. and 350° F. and holding the temperature of the mix at between about 180° F. and 190° F. insure excellent mixing and the production of formed units of consistently high quality.

While the above specific mixing procedure has been found to be very effective and is thus recommended, it will be understood that the comprehensive scope of the invention is not confined to such mixing technique.

Although certain recommended procedures have been described, it is to be understood that these are given didactically to illustrate the underlying principles of the invention and not as limiting its useful scope to the descriptive embodiment.

This application is a continuation-in-part of United States application Serial No. 158,125, filed April 26, 1950, now abandoned, and United States application Serial No. 267,158, filed January 18, 1952, now Patent No. 2,815,316.

I claim:

1. A process of producing a metallurgical grade coke from swelling coals which comprises subjecting such coal to flash oxidation with a gas containing free oxygen, in particulated and fluidized form, at a temperature within the plastic range of the coal for a controlled brief period of time; shock cooling the oxidized particulated coal to a temperature at which oxidation of the coal is inhibited; admixing the oxidized coal with a fluent binder to produce a plastic, formable mass of uniform consistency; forming the plastic mass into units of selected size and shape; carbonizing the formed units by contacting such units with a flowing stream of non-reactive gas characterized by a low partial pressure of the components

of the coal which are volatile at such carbonization temperature; controlling the rate of heat input from such non-reactive gas to said formed units substantially at the rate of between 2.5° C. to 5° C. per minute up to substantially 500° C. and thereafter controlling such heat input at the rate of from substantially 0.75° C. to 1.25° C. per minute up to the final finishing temperature.

2. A process in accordance with claim 1 in which the binder is comprised essentially of coal tar pitch.

3. A process in accordance with claim 1 in which the binder is comprised essentially of coal tar pitch in which is incorporated a wetting agent substantive to the coal.

4. A process in accordance with claim 1 in which the binder is compised essentially of coal tar pitch in which is incorporated an adjuvant which imparts headload strength to the said formed units.

5. A process in accordance with claim 1 in which a predetermined percentage effective of a carbohydrate binder is incorporated in the coal tar pitch.

6. A process in accordance with claim 1 in which the headload strength-imparting adjuvant is chosen from the group consisting of waste sulfite liquor, amylaceous and saccharogenic coal-binding compounds.

7. A process in accordance with claim 1 in which the oxidizing period in the plastic range is less than about twenty seconds.

8. A process in accordance with claim 1 in which the binder comprises essentially a coal tar pitch having a melting point of approximately 130° F.

9. A process in accordance with claim 1 in which the binder is coal tar pitch and comprises from approximately 12.5% to 15% of the binder-coal mix.

10. A process in accordance with claim 1 in which the oxidation of the coal is effected at a temperature above about 500° F.

11. A process in accordance with claim 1 in which the oxidation of the coal is effected at a temperature of between about 675° F. and 800° F. and within a period of less than approximately 15 seconds.

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