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FURNACE FOR CARBON BLACK PRODUCTION

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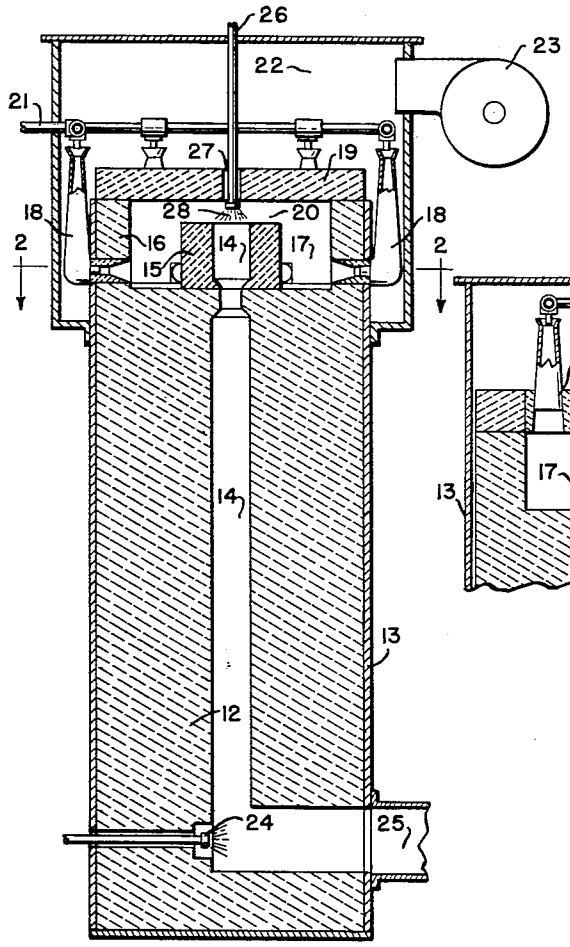


FIG. 1

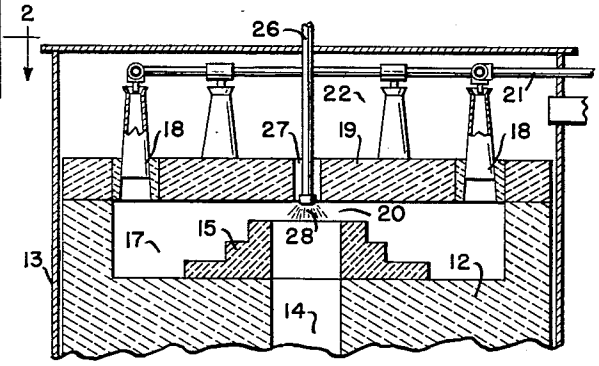


FIG. 3

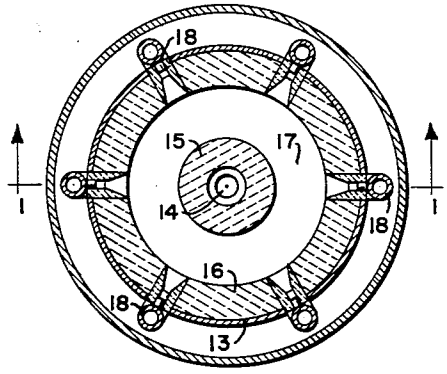


FIG. 2

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FURNACE FOR CARBON BLACK PRODUCTION

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This invention relates to a process and an apparatus for making carbon black of fine particle size and high reinforcing value in rubber, by the thermal decomposition of hydrocarbons. It is more particularly concerned with a process for producing carbon black by the thermal decomposition of liquid hydrocarbons under such conditions that the carbon is produced wholly or substantially wholly from one hydrocarbon stock and the heat necessary for the decomposition is supplied by the combustion of a second hydrocarbon stock.

It is known in the art to produce furnace carbon by the decomposition of liquid hydrocarbons in admixture with separately produced hot combustion gases. For example, United States Patent No. 2,625,466 of Ira Williams discloses a process generally of that nature.

Many of the properties of the carbon produced in a furnace process are determined by properties of the particular hydrocarbons from which the carbon is formed. For this reason, it is desirable that the carbon be produced only from that hydrocarbon stock which will yield carbon particles having the desired specific properties. We have found, however, that in the operation of the methods and furnaces generally used heretofore, much of the carbon obtained is produced from the hydrocarbon fuel stock rather than from the hydrocarbon cracking stock. The production of carbon from the fuel is particularly noticeable when a material of low free energy such as methane is used as fuel, due to the fact that the slow burning of such fuel leads to mixing with the cracking stock before it is completely burned. This slow-burning property results not only in forming carbon from part of the fuel stock but in burning part of the hydrocarbon intended to be cracked for the production of carbon black. Such prior processes are not only difficult to control, but they produce a carbon black now known to be inferior to that which can be produced.

One of the important objects of this invention is to provide a process and furnaces for practicing the process, by which carbon is produced substantially entirely from the desired cracking stock although this stock is decomposed in admixing with hot gases formed by the combustion of a hydrocarbon fuel of different composition.

A further object is to provide a way and means for evenly distributing the cracking stock with hot combustion gases so that decomposition of the cracking stock will take place in a well regulated manner to yield carbon of controlled quality.

Another important object of the invention is to provide a process and furnaces for producing an extraordinary fine grade of carbon black which is particularly valuable for rubber reinforcement.

According to this invention, carbon black is produced by burning suitable hydrocarbon fuel stock completely to form hot combustion gases substantially free of unburned fuel, flowing these hot gases in a constricted stream at high and accelerated velocity into a limited mixing zone at the mouth or inlet end of an elongated reaction chamber and thence into that chamber, distributing hydrocarbon cracking stock evenly into the hot gases as they enter the mixing zone, and decomposing the cracking stock in the resulting mixture to form the carbon. The decomposition is completed as the mixture passes through the elongated reaction chamber, and the decomposition products then are cooled before being discharged from the furnace for further treatment to separate and collect the carbon.

The hot combustion gases are formed by completely burning suitable fuel stock in a combustion chamber which preferably surrounds one end of the reaction chamber and leads into that end laterally through a constricted annular passage surrounding and defining the mixing zone. The hot gases thus converge from all sides into a central mixing zone, flowing into it at greatly increased velocity. As they enter it, the cracking stock is distributed symmetrically into the hot gases, as by injecting the cracking stock in the form of an expanding coaxial cone of atomized or vaporized hydrocarbon.

Thorough combustion of the fuel stock for the avoidance of unburned fuel in the hot combustion gases is promoted by supplying an abundant amount of air for the combustion and by causing the burning fuel mixture to impinge against a hot refractory wall surface and to change the direction of its flow before it can leave the combustion chamber.

The high velocity of the flow of hot gases into the mixing zone has been found to have a major influence upon the quality of the carbon produced. The product quality can be regulated by varying the gas velocity through adjustment of the cross-sectional area of the constricted flow passage, as by adjusting its height in the case of a constricted annular flow passage. For example, when using a reaction chamber 12 inches in diameter for the production of carbon from a heavy residue oil having and A.P.I. gravity of 5, a constricted annular passage 6 inches in height operating under a combustion gas pressure equal to 20 inches of water resulted in carbon having a surface area of 80 square meters per gram. When the height of the passage was reduced to 3 inches and the combustion gas pressure was equal to 90 inches of water, the surface area of the carbon product increased to 200 square meters per gram.

Other objects, features and advantages of this invention and preferred ways of practicing the invention will be apparent from the following detailed description, and from the accompanying illustrative drawings in which like numerals are employed to designate like parts throughout the same:

FIGURE 1 is a vertical section through a preferred furnace construction embodying the invention;

FIGURE 2 is a horizontal cross section taken at line 2—2 of FIGURE 1; and

FIGURE 3 is a fragmentary vertical cross section through the top portion of a modified form of furnace embodying the invention.

Referring first to FIGURES 1 and 2 of the drawings, the main body of the furnace there illustrated is formed by a long massive refractory cylinder **12** which is vertically disposed and enclosed in a steel shell **13**. An axial cylindrical passage **14** in this upright cylinder constitutes the reaction chamber in which the reaction is completed. The chamber **14** is shown as having a slight constriction near its upper end.

A thinner refractory ring **15** is mounted centraly upon the upper end of cylinder **12** and defines the top portion and the mouth or inlet of the reaction chamber **14**.

A second comparatively thin refractory ring **16** of greater diameter and greater height than ring **15** is also mounted on the upper end of body **12** so as to leave a space **17** between the rings **15** and **16** for the combustion of fuel stock. A series of inspirator burners **18**, shown as being six in number, opens into the space **17** near the outer extremity and near the bottom thereof. These burners are mounted so that they direct combustion gases and flame radially toward the outer cylindrical surface of ring

15. A refractory cover 19 is mounted over the top of ring 16 and serves both to enclose the combustion space 17 and to form a constricted annular passage or aperture 20 leading from that space radially over the upper end of ring 15 into a mixing zone between cover 19 and the mouth of the reaction chamber.

The burners 18 are supplied with fuel gas through a manifold 21. A plenum chamber 22 mounted at the top of the furnace encloses the inspirator burners 18, and this chamber is supplied by blower 23 with air under a pressure sufficient to maintain the desired air flow through the burners.

The bottom end of the reaction chamber 14 is provided with a water spray 24 serving to cool the gas-carbon mixture issuing from the chamber 14. The cooled mixture passes out of the chamber 14 through duct 25 to collecting equipment (not shown).

A pipe 26 for the introduction of cracking stock has a spray injector on its end, which is located above the mouth of the reaction chamber 14 in approximately coaxial relation thereto. This pipe passes from the plenum chamber through an opening 27 in the cover 19. The injector or nozzle at its end may be of any type suitable for uniformly injecting the cracking stock in an expanding conical spray pattern, as indicated diagrammatically at 28. It is arranged to discharge substantially at the bottom surface of the cover 19. The opening 27 is slightly larger than the pipe 26, which permits air from the plenum chamber 22 to flow along and cool the pipe and nozzle.

In operation, a mixture of air and hydrocarbon fuel containing enough air for complete combustion of the fuel is forcefully introduced into the combustion chamber 17 and burned therein from the several burners positioned about the perimeter of this chamber. While six burners ordinarily will accomplish an adequately uniform production and distribution of combustion gases, a greater or lesser number of burners may be used if the burners are of proper capacity and construction. At least four burners should be used in any arrangement in which the combustion chamber surrounds the reaction chamber. The burners may open in any desired direction toward a refractory wall surface of the combustion chamber.

The combustible fuel mixture is completely burned in chamber 17, and the resulting hot combustion gases then flow through the constricted annular passage 20 from all sides thereof into the mixing space at the mouth of the reaction chamber. As the hot gases converge at high velocity into that space they meet the cracking stock which is distributed evenly thereinto in the form of a conical spray. The spray cone preferably has such a wide cone angle that the cracking stock enters the hot gases before the gases enter the reaction chamber. The combustion gases generally have temperatures between about 1150° and 1500° C. as they flow into admixture with the cracking stock.

FIGURE 3 of the drawings illustrates another embodiment of a furnace structure capable of achieving the objects of this invention. In each embodiment a sheet-like flow of hot combustion gases is supplied at high velocity into admixture with a conical spray of cracking stock at the mouth of an elongated reaction chamber.

FIGURE 3 shows a modification of the furnace of FIGURE 1, in which the combustible fuel mixture enters the combustion chamber in a direction parallel to the axis of the reaction chamber and blasts against an end wall of the combustion chamber before it changes direction and flows through the restricted annular passage 20.

The practice of the invention is further illustrated by the following examples:

Example 1

A furnace of the design of FIGURE 1 was constructed with a refractory body 12 (outside diameter 66 inches) forming an inner cylindrical reaction chamber 14 of 12 inches in diameter and 12 feet in length. A refractory ring 16 of the same outside diameter as the body 12 and 16 inches high was mounted on top of body 12. A smaller refractory ring 15 having an inside diameter of 12 inches and a height of 12 inches was mounted on body 12 at the center, leaving an annular space 17 of 8 inches in width to serve as the combustion chamber between the two rings and providing a restricted annular passage 20 having a height of 4 inches between the top of ring 15 and the lower surface of cover 19. Six evenly spaced radially directed inspirator burners 18 were provided near the closed end of the combustion space 17. Cracking hydrocarbon was supplied under pressure through the pipe 26 to and through a nozzle which formed a hollow cone spray having a conical angle of about 80 degrees, so that the spray entered at an angle of about 40 degrees to the axis of the reaction chamber. The other parts used to complete the furnace were of known construction.

Natural gas of 1060 B.t.u. per cubic foot was supplied through the burners at the rate of 238 cubic feet per minute and was burned with 3120 cubic feet of air per minute. The pressure in the combustion chamber was equivalent to 61 inches of water. A residual oil having an A.P.I. gravity of about 3 was pre-heated to 650° F. and fed under pressure through the nozzle of pipe 26 at the rate of 2.62 gallons per minute. The air flow through the opening 27 along pipe 26 amounted to 100 cubic feet per minute.

The decomposition products formed in the reaction chamber 14 were cooled to 1200° F. by quenching with a water spray at the exit end of the furnace. The carbon was finally collected by means of conventional electrical precipitating apparatus.

This operation produced 4.5 pounds of carbon per gallon of cracking stock introduced. The carbon gave a substantially colorless solution when extracted with benzene.

To illustrate the advantage of the foregoing operation, the procedure was duplicated except that the furnace was altered by removal of the inner ring 15. This change provided an enlarged combustion chamber united with the mixing zone, which permitted mixing of much of the unburned fuel with the cracking stock before combustion of the fuel was complete. In this way only 3.7 pounds of carbon were produced per gallon of cracking stock, and the benzene extract of the product was dark brown.

The carbon products from these two procedures were incorporated in identical tire tread compounds which were vulcanized under similar conditions and tested for abrasion resistance. The test pieces prepared with carbon produced according to the present invention lost 0.396 inch in thickness while the test pieces prepared with carbon produced by the changed procedure lost 0.513 inch under the same wear conditions. The rubber made with the carbon produced according to the invention had an abrasion resistance of 129% of the rubber containing the carbon produced in the unseparated combustion chamber.

Example 2

In this example the height of the ring 15 of the furnace of FIGURE 1 was increased to 13 inches, leaving a constricted passage 3 inches high between this ring and the cover 19. Burners were altered to use No. 2 fuel oil. The pressure in the combustion chamber was equal to 87 inches of water. The cracking stock was an aromatic recycle oil having an A.P.I. gravity of 12. A yield of 4.1 pounds of carbon per gallon of cracking stock was obtained. The carbon was free of benzene extractable color. In a parallel operation in which the ring 15 was omitted, only 3.3 pounds of carbon were produced per gallon of oil used, and benzene extraction of the carbon produced a large amount of dark brown material. The rubber containing the carbon produced according to the present process had an abrasion resistance of 141% of the rubber containing the carbon produced in the unseparated combustion chamber.

The number of burners to be used in the furnace for best results will depend on the diameter of the combustion chamber. The burners should be spaced in such a manner that the chamber will be well filled with burning gases in order to assure the desired uniform flow of gases through the constricted annular passage leading to the mixing zone at the mouth of the reaction chamber.

It will be understood that the details and particulars of illustrative embodiments and examples set forth hereinabove are given by way of example and that the invention extends to all equivalents which will occur to those skilled in the art upon consideration of this disclosure and the claims appended hereto.

We claim:

1. A furnace for producing carbon black comprising a massive, vertically disposed refractory cylinder defining an elongated cylindrical reaction chamber, an outer refractory ring of greater internal diameter than said chamber extending upwardly from the upper end of said cylinder, an inner refractory ring on said upper end around the inlet of said reaction chamber, a refractory cover on said outer ring forming with it and with said upper end and said inner ring a combustion chamber surrounding said reaction chamber, said inner ring forming a constricted annular passage between its upper end and said cover to accelerate the flow of combustion gases from the combustion chamber to the reaction chamber, a plenum chamber over said cover, a series of inspirator burners leading from said plenum chamber into said combustion chamber and arranged to direct flames against a wall of the combustion chamber from a plurality of points spaced about the periphery thereof, said combustion chamber comprising at least one wall positioned to change abruptly the direction of flow of all the flame gases therein to assure complete combustion of said gases, means in said plenum chamber for supplying hydrocarbon fuel into said burners, means for supplying compressed air into said plenum chamber for inspiration by said burners, and means extending through said cover in coaxial relation to said inlet for spraying hydrocarbon cracking stock, in a conical pattern coaxial with said reaction chamber, substantially evenly into the hot combustion gases issuing from said passage so that a homogeneous reactive mixture of the cracking stock with said combustion gases flows continuously into said reaction chamber.

2. A furnace for producing carbon black comprising a massive, vertically disposed refractory cylinder defining an elongated cylindrical reaction chamber, a refractory ring of greater internal diameter than said chamber extending upwardly from the upper end of said cylinder, a smaller refractory ring mounted on said upper end inside the first-mentioned ring and forming the inlet of the reaction chamber, a refractory cover on the larger of said rings, said cover forming with said rings and with said upper end a combustion chamber surrounding the reaction chamber, said cover and said smaller ring being narrowly spaced apart to form a constricted passage for accelerating the flow of combustion gases from the combustion chamber to the reaction chamber, a plenum chamber over said cover, a seies of inspirator burners leading from said plenum chamber into said combustion chamber and arranged to direct flames against a wall of the combustion chamber from a plurality of points spaced about the periphery thereof, means in said plenum chamber for supplying fuel into said burners, means for supplying compressed air into said plenum chamber for inspiration by said burners, and means extending through said cover in coaxial relation to said inlet for spraying cracking stock substantially evenly into the hot combustion gases issuing from said passage.

3. A furnace for producing carbon black comprising, a massive refractory cylinder defining an elongated cylindrical reaction chamber, an outer refractory ring of greater internal diameter than said chamber extending axially from one end of said cylinder, an inner refractory ring extending axially from said one end of said cylinder and surrounding the inlet of said reaction chamber, a refractory cover on said outer ring forming with it and said one end of said cylinder and around said inner ring a combustion chamber, said inner ring having a free end spaced from said cover and forming between said free end and said cover a constricted annular passage to accelerate the flow of combustion gases from the combustion chamber to the reaction chamber, the distance between said one end of said cylinder and said cover being substantially less than the internal diameter of said outer ring and the distance between said free end and said cover being substantially less than the axial length of said inner ring, a series of hydrocarbon burners arranged to direct flames against a wall of said combustion chamber from a multiplicity of points spaced about the periphery of said combustion chamber, means connected to each of said burners for supplying hydrocarbon fuel to said burners, means for supplying compressed air to each of said burners, said combustion chamber having at least one wall positioned to change abruptly the direction of flow of all flame gases directed into it from said burners to assure complete combustion of said gases, and means extending centrally through said cover for introducing hydrocarbon cracking stock substantially evenly into the hot combustion gases passing through said annular passage.

4. A furnace for producing carbon black comprising, a massive refractory cylinder defining an elongated cylindrical reaction chamber, an outer refractory ring of greater internal diameter than said chamber extending axially from one end of said cylinder, an inner refractory ring extending axially from said one end of said cylinder and surrounding the inlet of said reaction chamber, a refractory cover on said outer ring forming with it and said one end of said cylinder and around said inner ring a combustion chamber, said inner ring having a free end spaced from said cover and forming between said free end and said cover a constricted annular passage to accelerate the flow of combustion gases from the combustion chamber to the reaction chamber, the distance between said one end of said cylinder and said cover being substantially less than the internal diameter of said outer ring and the distance between said free end and said cover being substantially less than the axial length of said inner ring, a series of hydrocarbon burners extending through said outer ring adjacent said one end of said cylinder and arranged to direct flames against a wall of said combustion chamber from a multiplicity of points spaced about the periphery of said combustion chamber, means connected to each of said burners for supplying hydrocarbon fuel to said burners, means for supplying compressed air to each of said burners, said combustion chamber having at least one wall positioned to change abruptly the direction of flow of all flame gases directed into it from said burners to assure complete combustion of said gases, and means extending centrally through said cover for introducing hydrocarbon cracking stock substantially evenly into the hot combustion gases passing through said annular passage.

5. A furnace for producing carbon black comprising, a massive refractory cylinder defining an elongated cylindrical reaction chamber, an outer refractory ring of greater internal diameter than said chamber extending axially from one end of said cylinder, an inner refractory ring extending axially from said one end of said cylinder and surrounding the inlet of said reaction chamber, a refractory cover on said outer ring forming with it and said one end of said cylinder and around said inner ring a combustion chamber, said inner ring having a free end spaced from said cover and forming between said free end and said cover a constricted annular passage to accelerate the flow of combustion gases from the combustion chamber to the reaction chamber, the distance between said one end of said cylinder and said cover being substantially less than the internal diameter of said outer

ring and the distance between said free end and said cover being substantially less than the axial length of said inner ring, a series of hydrocarbon burners arranged to direct flames against a wall of said combustion chamber from a multiplicity of points spaced about the periphery of said combustion chamber, means connected to each of said burners for supplying hydrocarbon fuel to said burners, means for supplying compressed air to each of said burners, said combustion chamber having at least one wall positioned to change abruptly the direction of flow of all flame gases directed into it from said burners to assure complete combustion of said gases, means extending centrally through said cover for introducing hydrocarbon cracking stock substantially evenly into the hot combustion gases passing through said annular passage, and an annular member of reduced diameter within said reaction chamber adjacent to the inlet thereof for constricting the flow of reaction materials thereinto.

References Cited in the file of this patent

UNITED STATES PATENTS

2,625,466	Williams	Jan. 13, 1953
2,656,254	Heller	Oct. 20, 1953
2,769,692	Heller	Nov. 6, 1956
2,785,054	Bethea et al.	Mar. 12, 1957
2,865,717	Krejci	Dec. 23, 1958
2,918,353	Heller	Dec. 22, 1959
2,924,512	Webster et al.	Feb. 9, 1960

FOREIGN PATENTS

525,031	Canada	May 15, 1956