Oct. 15, 1929. J. J. NAUGLE 1,731,473

METHOD OF TREATING CARBONACEOUS MATERIAL IN AN ELECTRIC FURNACE OR THE LIKE

Filed April 21, 1923 2 Sheets-Sheet 1

Fig. 1.

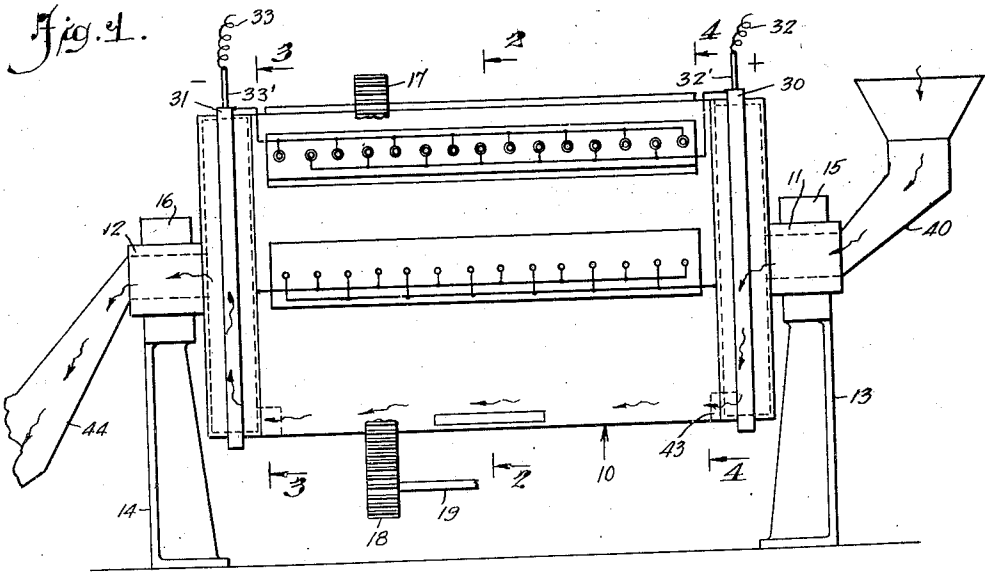
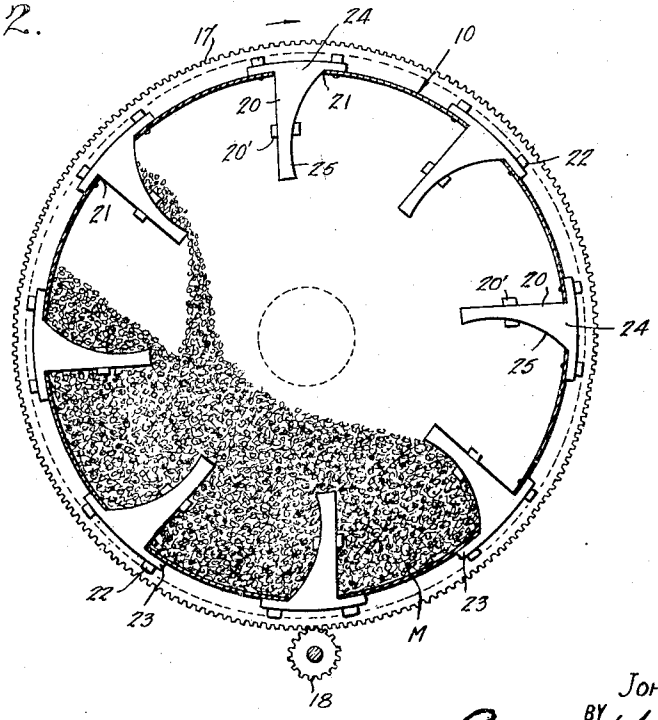


Fig. 2.



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Fig. 3.

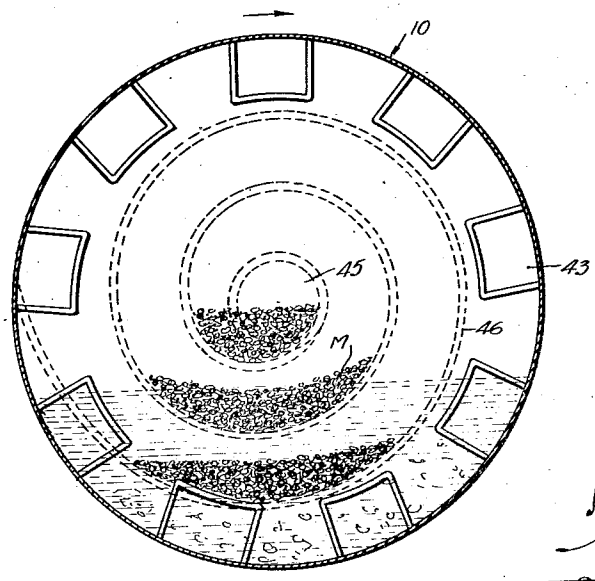


Fig. 4.

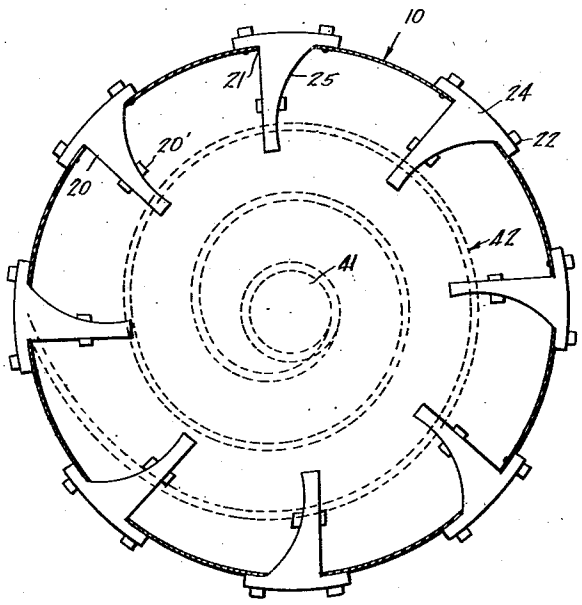
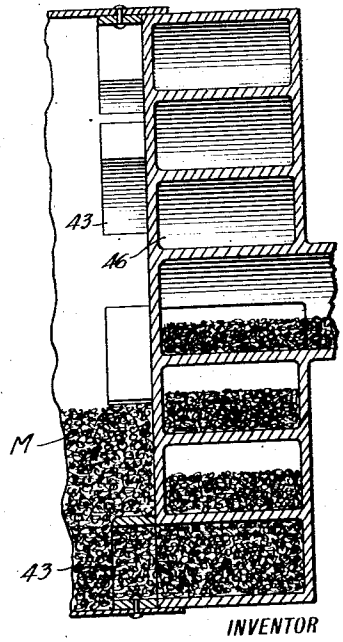


Fig. 5.



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1,731,473

UNITED STATES PATENT OFFICE

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METHOD OF TREATING CARBONACEOUS MATERIAL IN AN ELECTRIC FURNACE OR THE LIKE

Application filed April 21, 1923. Serial No. 633,640.

My present invention relates to methods of treating carbonaceous material, particularly comminuted carbonaceous material, such as comminuted carbonized lignin residues obtained by carbonizing the waste liquors derived from the treatment of wood by the alkali (soda) process in the production of wood pulp, for the production from the materials designated of a very pure grade of carbon characterized by a very low "ash" or mineral content, high conductivity, and a remarkably high degree of activation corresponding to a high degree of decolorizing power. It is an object of the present invention to devise methods of the character specified above which shall be exceedingly simple in operation, requiring comparatively simple apparatus and a minimum of time, labor and material for this purpose; which shall produce the desired product in the desired condition of high purity, high conductivity and a high degree of activation and decolorizing power; and which shall render possible a very high yield of the desired product in proportion to the raw material used and the amount of power consumed.

In the accompanying specification I shall describe an illustrative embodiment of the method of the present invention. In the accompanying specification I shall also describe, and in the annexed drawing show, one form of apparatus in which the method of the present invention may be conveniently practiced. It is, however, to be clearly understood that my invention is not limited to the specific embodiment of the process described herein, nor to the use of the particular form of apparatus shown and described herein, said apparatus forming the subject matter of a separate application for Letters Patent filed by me of even date herewith.

In order that my invention may be more clearly understood, reference may be made to a co-pending application of mine, filed of even date herewith, Serial No. 633,643 and entitled "Process for the treatment of comminuted carbonaceous materials and the like, in an electric furnace and the like."

In practicing the aforesaid illustrative embodiment of the method of the present invention, I take first any suitable raw material, preferably a carbonaceous material in comminuted or finely divided form. While various types of comminuted carbonaceous material may be treated with success in accordance with the principles of the present invention, either in the production of a highly activated carbon from the raw material, or in the revivification of spent decolorizing carbon, I shall describe the present invention as applied more particularly to the preparation of a highly activated carbon from carbonized lignin residues in comminuted form, such residues being obtained by carbonizing the cooking liquors derived from the treatment of wood by the alkali process in the production of wood pulp.

Taking, for example, the raw material specified above, which comprises comminuted carbonized lignin residues, such material may have an average analysis as follows:

Analysis of comminuted carbonized lignin residues

	Per cent
Carbon	90
Ash	10

Analysis of ash

Silicious matter	0.13
Silica	0.75
Ferric oxide	0.34
Alumina	0.47
Lime, CaO	0.23
Magnesia, MgO	0.11
Chlorine as chlorides	0.47
Sulphuric anhydride	2.65
Carbon dioxide	1.00
Alkalies (by diff.)	3.85
	10.00

This material contains certain volatile matter and also certain fusible mineral substances which have a high resistance and which serve substantially to reduce the conductivity of the raw material.

I now heat the mass of raw material specified above so as to expel the volatile substances and to fuse the mineral substances, characterized by a high resistivity, for the

subsequent removal of such fused mineral substances. This I preferably do by passing an electric current of regulated density through the mass of the raw material until the mineral substances present therein have been fused. The degree of current employed should be such as to cause the mineral substances to aggregate or sinter together in the formation of clinkers.

The fused mineral substance is now removed in any suitable manner so as to yield a product which is characterized by a very high conductivity, amounting often to as much as five times the conductivity of the raw material treated, and characterized, further, by a very substantial reduction in the mineral content of the final substance, which mineral content is often reduced to as little as one-fourth or less of the mineral content of the original raw material treated. I find that the conductivity of the final product is a function of the degree of activation or decolorizing power of such material, or vice versa, increased conductivity indicating increased activation and decolorizing power, and vice versa. Accordingly, by the removal of the fusible mineral substances, which are of comparatively high resistivity and which, therefore, reduce the conductivity of the final product, I am enabled to effect a very remarkable increase in the degree of activation and decolorizing power of the material treated in accordance with the principles of the method of the present invention.

The present method may be combined with considerable success with the means and method described and claimed in my copending application, filed May 22, 1922, Serial No. 562,828, and entitled "Prefiltering means and method", especially so far as the preliminary treatment of the raw materials is concerned, particularly to effect by decantation from a watery suspension of such material a partial preliminary separation by gravity of the portions of the material richer in carbon from the portions of the material poorer in carbon and containing an excess of heavy mineral substances. I may also use the feature of effecting a purification of the final product by forced filtration, under pressure, through a layer of the final product, of an aqueous acidified fluid and of wash waters. I may also add to the charge of material being treated in accordance with the principles of the method of the present invention, certain reagents, such as sodium fluoride, which will have the property of eliminating one or more of the mineral substances present in the charge, particularly silica, in the form of volatile by-products, such as silicon fluoride.

The resulting product possesses many advantages for the intended purpose, since it can be very cheaply made in accordance with the method of the present invention, and possesses the desired quality of high conductivity, corresponding to a remarkably high degree of activation and decolorizing power. An average analysis of such material is as follows:

Analysis of the treated product

	Per cent
Carbon	97.60
Ash	2.40

Analysis of ash

Calcium	0.16
Sodium, Na_2O	0.35
Magnesium, MgO	0.16
Chlorides, (Cl_2)	0.06
Sulphates (SO_3)	0.78
Silica, SiO_2	0.50
Iron and alumina, (Fe_2O_3, Al_2O_3)	0.39
	2.40

This product is also characterized by a very high degree of purity, corresponding to a high conductivity and a high degree of activation and decolorizing power. The conductivity of the final product is often increased as much as five times that of the raw material treated, and its mineral content is often reduced to as low as one-fourth or less that of the raw material treated in accordance with the method of the present invention. The method itself is exceedingly simple to practice, requiring comparatively simple apparatus and a minimum of time, labor, power and material for this purpose. The method is also admirably calculated to produce the desired end product in the desired condition of purity, high conductivity, and high activation and decolorizing power. The method is also very economical in view of the very high yield which may be obtained by its practice as compared with the methods hitherto employed for revivifying spent decolorizing carbon or for producing highly activated carbons for decolorizing purposes.

Referring to the drawing wherein I have illustrated one form of apparatus in which the method outlined above may be conveniently practiced:

Fig. 1 is a view in longitudinal section, partly broken away, and partly in elevation, showing the furnace comprising the aforesaid form of apparatus;

Fig. 2 is a cross-sectional view of the same taken along line 2—2 of Fig. 1 of the drawing;

Fig. 3 is a cross-sectional view of the same taken along line 3—3 of Fig. 1 of the drawing;

Fig. 4 is a view similar to Fig. 3 taken along line 4—4 of Fig. 1 of the drawing; and

Fig. 5 is an enlarged detail longitudinal sectional view at the outlet end of the machine.

Referring to the aforesaid form of ap-

paratus in which the aforesaid illustrative embodiment of the method of the present invention may be conveniently practiced, the furnace comprising such apparatus includes the casing 10, preferably of substantially cylindrical form, having the inlet tube 11 and the outlet tube 12, said tubes being likewise of substantially cylindrical form and comprising bearings for the casing 10. The casing 10 and the tubes 11 and 12 are supported by a plurality of standards 13 and 14, providing bearing sleeves 15 and 16 for the tubular bearings 11 and 12.

In the form illustrated by way of example herein, I provide means for effecting a rotation, which is preferably continuous, of the casing 10 and the parts carried by and within the same. For this purpose, I provide any suitable means, such as the annular toothed gear 17 carried by and surrounding the casing 10, and adapted to cooperate with a spur gear or pinion 18 carried by a shaft 19 driven from any suitable source of power not here deemed necessary to be shown. By the meshing of the gears 17 and 18, the casing 10 and its associated parts may be rotated at any desired rate of speed.

Within the casing 10 are a plurality of stirring and mixing members, which may serve also as supports for the electrodes, said members being generally indicated by reference character 20. Each of the members 20 is preferably replaceable or readily removable from the casing 10, as by having each such member pass through an opening or orifice 21 in the wall of the casing 10. By means of suitable bolts 22, passing through flanges 23 forming a part of the base 24 of each member 20, each member 20 may be readily installed and as readily removed for cleaning, repair or replacement, as desired.

In order to bring about an effective mixing of the material and the proper distribution of current through same, I prefer to provide some or all of the members 20 with a hollowed-out surface for the reception and subsequent discharge of a portion of the material in the furnace. For this purpose, that face of each or some of the members 20 which, during the rotation of the casing 10, first comes into contact with the material M in the furnace, is dished-out or made slightly concave, as indicated at 25.

It will be noted that each set of members 20 is disposed in spaced relation along the circumferential periphery of the inner face 10′ of the casing 10. It will also be noted that the members 20 of each such set are substantially radially disposed. I have found, by trial and experiment, that this disposition of the members 20 and the electrodes carried by the same is of considerable importance for the proper treatment of the material within the furnace, by insuring proper mixing of the material and the proper distribution of current through the mass being treated.

The electrodes 20′ are in the form of longitudinally arranged knobs or studs, preferably located on both sides of each of the members 20, which members are preferably of insulating material or are properly insulated, so as to avoid connection with the electrodes, and also to avoid short-circuiting the furnace through the metallic portions of the casing and other parts of the machine. While various types of electrical connections may be employed with considerable success for the electrodes 20′, I prefer to use the means illustrated in the drawing and comprising conductor rings 30 and 31 mounted at the inlet and outlet ends of the casing 10, respectively, and suitably insulated from the metallic structure of the same. At 32 and 33 I have indicated the leads or wires for supplying current to the conductor rings 30 and 31, respectively. I prefer that the electrodes 20′, of which several peripheral sets or rows, here shown as fifteen peripheral rows each including eight peripherally disposed electrodes, may be employed, shall be so connected to the conductor rings 30 and 31, and the leads 32 and 33 supplying current thereto, that successive electrodes in each peripheral set will be oppositely charged and that successive electrodes in each longitudinal set will be likewise oppositely charged. I find that this arrangement of electrodes is highly desirable in that it assists in effecting the proper distribution of current through the charge of material within the furnace.

In order to continuously feed material into the furnace for treatment and out of the furnace after the same has been subjected to the desired treatment for the desired length of time, I provide any suitable means, but preferably an inlet hopper 40 for feeding untreated material into the machine through the tubular bearing 11. The tubular bearing 11 communicates with the central chamber 41 of a spiral feeding device, generally indicated by reference character 42. The spiral feeding device 42 is located at the inlet end of the machine, and is provided with the discharge openings 43 leading into the heating chamber 10″ within the casing 10. Similarly, the discharge outlet is in the form of a conduit 44 communicating at its upper end with the tubular bearing 12 which leads into the central opening 45 of a spiral outlet feeding device, generally designated by reference character 46, and similar in most respects to the spiral inlet feeding device 42. The feeding device 46 has the inlet openings 4 communicating with the spiral passage in the feeding device 46 for the admission of treated material for its subsequent discharge from the machine.

The operation of the machine and its relation to the process already described will be

substantially clear from the foregoing description and may be briefly summarized as follows: Comminuted carbonaceous material, particularly comminuted carbonized lignin residues, generally indicated by reference character M, is continuously fed into the machine through the hopper 40 and the feeding means 42, and thus enters the heating chamber 10'', where it is brought into operative contact with the electrodes 20'. The electrodes 20', by the continuous rotation of the casing 10 carrying the same, are brought into intermittent contact with the material M and serve to intermittently and continuously stir and mix the same. This desired action is facilitated by the particular form which the members 20 possess. This form, resulting from the dished-out surfaces 25 with which these members are provided, serves to cause the members 20 intermittently to receive and subsequently to discharge a portion of the material M being treated within the furnace.

The course of such material is diagrammatically illustrated in Fig. 2 of the drawing, which approximates the position of the mass of material M within the casing for a particular speed of operation of the machine. The electrodes 20' serve, by reason of their connection with the conductor rings 30 and 31, and the lead wires 32 and 33 supplying current thereto, to discharge a current of regulated and controlled density through the mass M of material being treated in the furnace. The current should be such as to readily volatilize volatile matter contained in the mass of comminuted carbonized lignin residues being treated in the furnace, in the particular example given. The current should also be strong enough to fuse the fusible mineral substances present in the material being treated.

The time of treatment will, of course, depend upon the particular material being treated, but in general is about 15 to 60 minutes. The resulting product, in addition to the desirable properties already specified, possesses a high degree of neutrality, which is very desirable, and possesses also a decolorizing power substantially, and often as much as ten per cent, greater than the best activated carbons hitherto produced. The resulting product is described and claimed in a copending application filed by me of even date herewith.

It may here be stated that the carbonized lignin residues referred to herein are the residues remaining after leaching the residues obtained by carbonizing the spent or used cooking liquors obtained in the treatment of wood by the alkali (soda) process in the treatment of wood pulp; and that the terms "activation" and "decolorizing", or their equivalents, as used in the specification and claims, include also deodorization, purification and filtration.

What I claim as my invention is:

1. The method of treating carbonaceous material to obtain a highly activated, decolorizing carbon therefrom, which comprises heating a mass of such material to fuse a part at least of the mineral content thereof while passing collecting members through the mass to remove the fused material from the same.

2. The method of treating carbonized lignin residues to obtain a highly activated, decolorizing carbon therefrom, which comprises heating a mass of such material by passing an electric current therethrough to fuse a part at least of the mineral content thereof while passing collecting members through the mass to remove the fused material from the same.

3. The method of treating carbonaceous material to obtain a highly activated, decolorizing carbon therefrom, which comprises passing a plurality of electrodes through a mass of such material to heat the same sufficiently to fuse a part at least of the mineral content of the mass while stirring the same.

4. The method of treating carbonaceous material to obtain a highly activated, decolorizing carbon therefrom, which comprises passing a plurality of electrodes through a mass of such material to heat the same sufficiently to fuse a part at least of the mineral content of the mass while stirring the same to collect and remove therefrom the fused material.

5. The method of treating carbonaceous material to obtain a highly activated, decolorizing carbon therefrom, which comprises passing an electric current of regulated density through the mass, from electrodes extending into the same, to fuse a part at least of the mineral content of the mass, and removing the fused material therefrom.

6. The method of treating carbonized lignin residues to obtain a highly activated, decolorizing carbon therefrom, which comprises continuously passing an electric current of regulated density through the mass, from electrodes extending into the same, to fuse a part at least of the mineral content of the mass, and continuously removing the fused material therefrom.

7. The method of treating carbonaceous material to obtain a highly activated, decolorizing carbon therefrom, which comprises passing an electric current of regulated density through the mass, from electrodes passing into and through the same, to fuse a part at least of the mineral content of the mass, and removing the fused material therefrom by adhesion of said fused material to said electrodes.

8. The method of treating carbonized lignin residues to obtain a highly activated, decolorizing carbon therefrom, which comprises continuously passing an electric current of regulated density through the mass, from

electrodes passing through the same, to fuse a part at least of the mineral content of the mass, and continuously removing the fused material therefrom by adhesion of said fused material to said electrodes.

9. The method of treating carbonaceous material to obtain a highly activated, decolorizing carbon therefrom, which comprises passing an electric current of regulated density through the mass, from electrodes extending into the same and in the presence of foreign electrically conductive bodies, to fuse a part at least of the mineral content of the mass, and removing the fused material therefrom.

10. The method of treating carbonized lignin residues to obtain a highly activated, decolorizing carbon therefrom, which comprises continuously passing an electric current of regulated density through the mass, from electrodes extending into the same and in the presence of foreign carbonaceous bodies of comparatively great conductivity, to fuse a part at least of the mineral content of the mass, and continuously removing the fused material therefrom.

11. The method of treating carbonaceous material to obtain a highly activated, decolorizing carbon therefrom, which comprises passing an electric current of regulated density through the mass, from electrodes passing into and through the same and in the presence of foreign bodies of comparatively great conductivity, to fuse a part at least of the mineral content of the mass, and removing the fused material therefrom by adhesion of said fused material to said electrodes.

12. The method of treating carbonized lignin residues to obtain a highly activated, decolorizing carbon therefrom, which comprises continuously passing an electric current of regulated density through the mass, from electrodes passing through the same and in the presence of added carbonaceous bodies of comparatively great conductivity, to fuse a part at least of the mineral content of the mass, and continuously removing the fused material therefrom by adhesion of said fused material to said electrodes.

13. The process of continuously treating carbonaceous material, such as carbonized lignin residues, which comprises heating a mass of such material sufficiently to fuse a part at least of the mineral content thereof, continuously removing said fusible material, continuously adding untreated portions of material to the mass of material being treated, and continuously removing portions of treated material from the heating zone.

14. The process of continuously treating carbonaceous material, such as carbonized lignin residues, which comprises heating a mass of such material, by passing an electric current of regulated density therethrough, sufficiently to fuse a part at least of the mineral content thereof, continuously removing said fusible material, continuously adding untreated portions of material to the mass of material being treated, and continuously removing portions of treated material from the heating zone.

15. The process of continuously treating carbonaceous material, such as carbonized lignin residues, which comprises heating a mass of such material, by passing an electric current of regulated density therethrough from electrodes rotating through the mass of material being heated, sufficiently to fuse a part at least of the mineral content thereof, continuously removing said fusible material, continuously adding untreated portions of material to the mass of material beng treated, and continuously removing portions of treated material from the heating zone.

16. The process of continuously treating carbonaceous material, such as carbonized lignin residues, which comprises heating a mass of such material, by passing an electric current of regulated density therethrough from electrodes rotating through the mass of material being heated, sufficiently to fuse a part at least of the mineral content thereof, continuously removing said fusible material, continuously adding untreated portions of material to the mass of material being treated, and continuously removing portions of treated material from the heating zone by adhesion of said fused material to said electrodes.

17. The method of treating carbon lignin residues containing mineral impurities to obtain a highly activated carbon therefrom, which comprises heating a mass of such material in an electric furnace in the presence of an addition agent to thereby volatilize a part, at least, of the mineral impurities present in such material.

18. The method of treating carbon lignin residues containing silicious mineral impurities to obtain a highly activated carbon therefrom, which comprises heating a mass of such material in the presence of a fluoride to thereby volatilize a part, at least, of the silicious mineral impurities present in such material.

19. The method of treating carbonized lignin residues containing mineral impurities to obtain a highly activated decolorizing carbon therefrom, which comprises passing a heating electric current of regulated density through a mass of such material in the presence of an addition agent to thereby volatilize a part, at least, of the mineral impurities present in such material.

20. The method of treating carbonaceous material to obtain a highly activated carbon therefrom which comprises heating a mass of such material in an electric furnace to fuse a part at least of the mineral content thereof, in

the presence of an addition agent to thereby volatilize a part at least of the mineral content of the mass.

21. The method of treating carbonaceous material to obtain a highly activated carbon therefrom which comprises heating a mass of such material in an electric furnace to fuse a part at least of the mineral content thereof, in the presence of an addition agent to thereby volatilize a part at least of the mineral content of the mass while agitating said mass.

22. The method of treating carbonaceous material to obtain a highly activated carbon therefrom which comprises heating a mass of such material to fuse a part at least of the mineral content thereof, said heating being in the presence of a fluoride to thereby volatilize a part at least of the mineral content of the mass.

23. The method of treating carbonaceous material to obtain a highly activated carbon therefrom which comprises heating a mass of such material to fuse a part at least of the mineral content thereof, said heating being in the presence of a fluoride to thereby volatilize a part at least of the mineral content of the mass while agitating said mass.

24. The method of obtaining a highly activated carbon which comprises heating carbonaceous material in an electric furnace to fuse a part at least of the mineral content of the carbonaceous material while passing collecting members through the material to remove the fused material from the carbonaceous material.

In testimony whereof, I have signed my name to this specification this 30th day of March, 1923.

JOHN J. NAUGLE.