J. E. BUCHER.

PROCESS OF FIXING NITROGEN AND ELECTRIC FURNACE FOR EFFECTING THE SAME.

APPLICATION FILED OCT. 2, 1914.

1,174,668.

Patented Mar. 7, 1916.

2 SHEETS—SHEET 1.

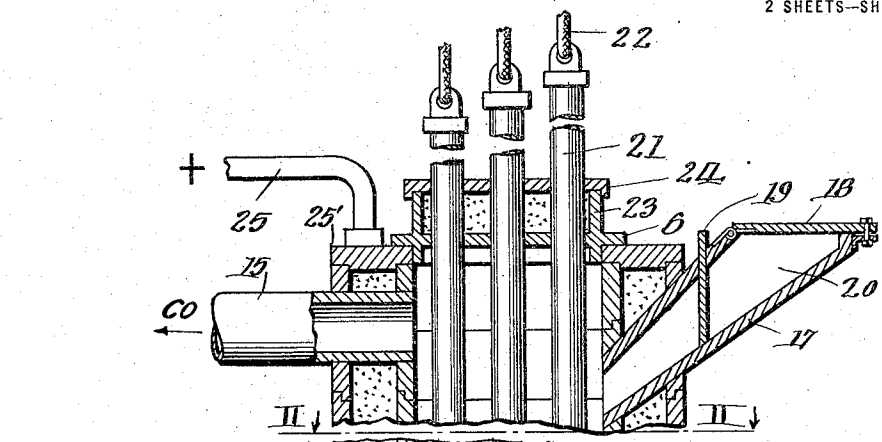


Fig. 1.

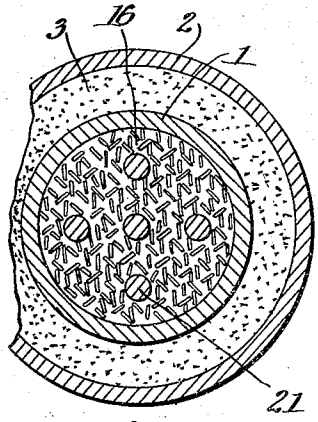


Fig. 2.

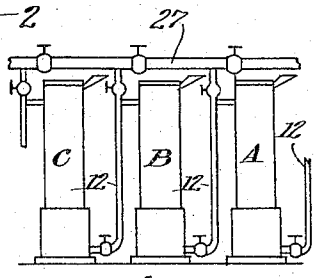
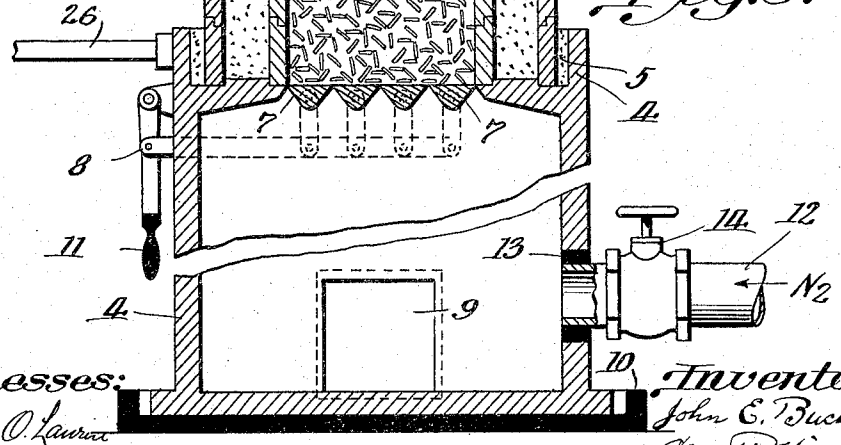


Fig. 3.



Witnesses:
Charles O. Laurence
E. M. Jordan.

Inventor
John E. Bucher
By Chas. F. Hidden
his Atty.

THE COLUMBIA PLANOGRAPH CO., WASHINGTON, D. C.

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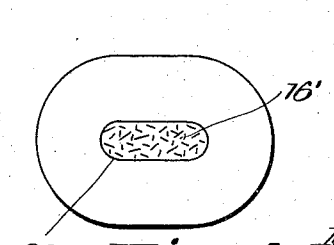


Fig. 5.

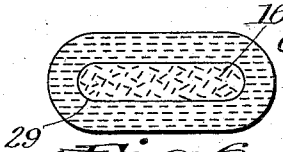


Fig. 6.

Fig. 4.

Witnesses:
Charles O. Lourie
E. M. Jordan

Inventor:
John E. Bucher
By Chas. F. Hoban
his Atty.

THE COLUMBIA PLANOGRAPH CO., WASHINGTON, D. C.

UNITED STATES PATENT OFFICE.

JOHN E. BUCHER, OF COVENTRY, RHODE ISLAND, ASSIGNOR TO NITROGEN PRODUCTS COMPANY, OF PROVIDENCE, RHODE ISLAND, A CORPORATION OF RHODE ISLAND.

PROCESS OF FIXING NITROGEN AND ELECTRIC FURNACE FOR EFFECTING THE SAME.

1,174,668. Specification of Letters Patent. Patented Mar. 7, 1916.

Original application filed June 25, 1914, Serial No. 847,337. Divided and this application filed October 2, 1914. Serial No. 864,690.

To all whom it may concern:

Be it known that I, JOHN E. BUCHER, a citizen of the United States, residing at Coventry, in the county of Kent and State of Rhode Island, have invented certain new and useful Improvements in Processes of Fixing Nitrogen and Electric Furnaces for Effecting the Same, of which the following is a specification.

This invention relates to a process and means for the fixation of atmospheric nitrogen; the free nitrogen being caused to combine with carbon and with a suitable base forming element such as an alkali or alkaline earth metal, to form a cyanogen compound.

This combination is effected through the intermediacy of a suitable catalyzer which is capable of rendering the carbon reactive, and the cyanogen-compound-forming reaction is in the hereindescribed process effected in a suitable receptacle or furnace, heated either directly, or in part at least directly, by passing electric current through the mass of material which supplies preferably all of the constituents of the cyanogen compound to be formed other than the nitrogen.

The preferred process is especially characterized by the fact that the electrical heating is employed in such a manner that the charge itself acts, either entirely, or in part, as the electrical resistor.

A charge of the preferred material, or rather mixture of materials, conducts heat rather slowly and, if it be attempted to heat the mass thereof exteriorly, heat has to penetrate the interior portions by being conducted through the walls of the retort and the outer layers of the charge.

Owing to the extremely efficient mixture and the rapidity with which the nitrogen may be supplied, especially to briqueted materials of the character in question, the rapidity of the process is very seriously retarded by this relatively slow heat penetration; and it has been one of the principal objects of the present invention to surmount this difficulty.

Furthermore, where external heating is resorted to the retorts and furnaces must be so designed that the heat has to penetrate but a comparatively short distance into the charge and even then the temperature outside has to be kept much higher than that necessary for economical cyanid formation, to the end that a reasonably rapid heat penetration may be assured. This involves the additional serious disadvantages that the retorts, if of iron as they preferably are, soon become highly carburized and are more likely to melt or become injured by oxidation; that the exterior portions of the charge may be heated so as to melt or sinter the catalyzer, which is preferably pulverulent, at least initially; and that in the hotter exterior portions there may occur such a volatilization of the alkali compounds or their equivalents as to completely disintegrate the briquets with consequent tendency to flood other portions of the charge with such an excess of alkali material as to make such portions partially or entirely inactive. I have found that these disadvantages, which are practically inherent in substantially all methods of external heating can be overcome completely by internal electrical heating through the use of the charge itself as a resistor.

Nitrogen fixation, by the use of an electrical resistor made up of such materials as a mixture of finely divided iron, carbon and sodium carbonate has not heretofore been accomplished, and its accomplishment presented some apparently serious difficulties. For example, while such a resistor is, largely by reason of the presence of the solid alkali carbonate, virtually an insulator, I have found that its resistivity may be so enormously diminished upon heating, as to convert it to a fairly good conductor which becomes plastic while at the same time ingredients thereof participate powerfully in the endothermic cyanid-forming reaction. This marked change in conductivity is probably due in large part to the presence of the finely divided catalyzer which is preferably a conductor of the first class, whose particles while at first, for the most part, are out of direct contact with each other or with the carbon, are, as the resistor heats up, caused to make better contact both with each other and with the carbon through the partial melting of the insulating binder.

By way of exemplifying the preferred mode of conducting my process, it may be

here stated that iron and carbon, both finely divided, may be mixed with, for example, alkali carbonate and briqueted under pressure after the addition of suitable quantities of water; or, the mixture may be heated slightly above the melting point of the alkali carbonate while substantially excluding air, and briqueted under suitable pressure. In the latter case the briquets are already at the temperature of the cyanid formation and this preheating is of great advantage.

If the briquets are prepared moist, they are best dried quickly so that they may be firm and even this slight heating is of great advantage, not only because of the heat thus supplied to the briquets, but also on account of the increase in electrical conductivity due to this preheating.

The briquets may be made in many shapes but the ones preferably used in this process were pressed through a die and were made cylindrical in shape, being each one-quarter of an inch in diameter and about one inch long. A resistor of such briquets presents large interstices through which the nitrogen current can flow to all portions of the resistor, after which it quickly penetrates by diffusion through the very small interstices or pores between the particles or iron and carbon.

The briquets are in general only in contact with each other at points and hence at first offer very poor electrical contact; and the same must be true to some extent of the very small irregular fragments of iron and carbon of which the briquets are formed, which are, in addition, more or less separated by the powdered sodium carbonate; the latter being an insulator when cold. The entire resistor is hence almost a non-conductor when cold and even 500 volts E. M. F. may send but a very slight current through a five-foot column in a one-inch tube. The very slight current soon reaches a maximum when the heat loss by radiation counterbalances the heating effect of the current and it would hence require an excessively high voltage to bring the heating action to the critical point and a still more impractical voltage to do this expeditiously. Increasing the pressure on the mass increases the conductivity somewhat but not to the desired extent. This initial resistance can be overcome by using a shorter column which permits of the use of a more reasonable voltage and when the current flows freely, the electrodes may be partly withdrawn and more briquets added after which the current may be again allowed to act until it flows freely. This procedure may be repeated easily and quickly in a furnace of the character shown in Fig. 1 of the accompanying drawings until the resistor is of the desired length. The current is then allowed to flow until the entire mass shows a uniformly bright incandescence and the conductivity of the resistor has increased over a thousandfold, in some cases. It is also possible to avoid the use of currents of high voltage by preheating the briquets or using them hot as they come from the drier in which their moisture is removed. When a mass of briquets is used as a resistor, the conductivity of the carbon and soda ash increases while that of the iron decreases, and, as the temperature rises heat is developed at the points of poorer contact, but as the temperature continues to rise the briquets soften at first at such points and the weight of the column then presses them together so as to produce a surface contact with comparatively good conductivity whereupon excessive heating at such surfaces ceases automatically and other portions in turn, become heated; thus equalizing the heating of the resistor. Also if the resistor is of such a shape that the current may flow in channels, producing excessive heating, this may be equalized by reducing the voltage of the current or shutting it off entirely until the heat has been conducted away to the cooler parts; thus again equalizing the resistance and consequently the heating.

Injurious local heating in such a resistor is further counteracted by the latent heat of fusion of the alkali carbonate which is present, and even more effectively by the very rapid volatilization of such alkali compounds, where the temperature temporarily gets too close to the melting point of the carburized iron. The condensation of this volatilized matter on the neighboring colder parts heats such neighboring parts; and thereby again tends to equalize heat throughout the mass. The gas current through the coarse interstices of the resistor greatly increases this favorable heat distributing action both by convection and increasing volatility. The heat changes due to vaporization and condensation not only act directly as a safety device to secure uniform heat distribution in the resistor, but, also effectively, by thus controlling the heat generation itself, effect a consequential automatic adjustment of resistance.

In addition to the equalization and control of temperature, effected by reason of the nature of the resistor employed, the powerfully endothermic reaction which takes place within the resistor mass;

(1) $Na_2CO_3+4C+N_2=2NaCN+3CO-138{,}500$ cal.

affords another very effective means for aiding in the equalization of temperature through the resistor mass. For example, when a portion of the resistor becomes hotter than the rest, cyanid formation will occur much more rapidly at such places; thus caus-

ing rapid heat absorption with simultaneous readjustment of resistance to equalize heat production.

The enormous change of conductivity with temperature makes such a resistor an excellent thermostat for the process, as it is only necessary to determine for given conditions what amperage is necessary and then insert the proper fuses, circuit breakers and transformers into the circuit. Thus the current may be safely and automatically controlled even when heat is generated very rapidly in the charge. A 100 ampere current at 104 volts can readily be passed through a cylindrical column of briquets 1½ inches in diameter by 10 inches in length, which current will supply enough heat to complete the above reaction in about thirty seconds. On a small scale with properly prepared briquets I have actually converted the carbonate into cyanid in less than one minute. Such a very rapid generation of electrical energy in the resistor allows comparatively little time for loss by radiation and hence gives very high heating efficiency. This resistor contains an extremely active catalytic solution surface, provides for very free access of nitrogen to the reacting surface, and permits rapid heating of the interior portions of the charge. The preferred resistor, further, permits of very free circulation of alkali metals or compounds through its interstices by reason of its porosity, and of the absence therefrom of injurious binders and supports. Because of these and other advantages it permits cyanid formation more rapidly and efficiently than has heretofore been possible.

The resistor may be made in many different forms. For example, it may be pressed through a die in one piece, with suitable perforations to conduct the gases therethrough. The iron-carbon-sodium carbonate mixture may even be used in the powdered form without briqueting; but in any case the resistor should be so prepared and used as to provide and maintain an enormous catalytic solution surface at which the cyanid-forming reaction may take place, while at the same time providing for free access of alkali metal or compounds thereof or equivalents of such metal or compounds, together with nitrogen, thereto.

In the accompanying drawings which form a part hereof I have exemplified a preferred furnace; but as I am aware of very many changes and modifications which may be made herein without departing from the spirit of my invention and while yet affording means suitable for the effectuation of the herein described process, I desire in no way to be restricted to the form of apparatus shown nor indeed to the specifically described steps of my process other than as set forth in the claims appended hereto.

Referring to the drawings: Figure 1 is a vertical median section of a furnace in which my process may be carried out. Fig. 2 is a section through said furnace taken on line II—II of Fig. 1. Fig. 3 is a diagrammatic representation of a group of furnaces. Fig. 4 is a somewhat diagrammatic section of a modified construction adapted for continuous operation of the process. Fig. 5 is a section taken on line V—V of Fig. 4, and Fig. 6 is a section taken on line VI—VI of said figure.

In the apparatus shown in Fig. 1 the furnace comprises inner and outer cylindrical portions respectively designated 1 and 2. The inner one may be built of superposed magnesia brick and the space 3 therebetween may be filled with kieselguhr, or even ashes or some like refractory material. These telescoped chimney-like sections may be sustained by a base 4 of cast-iron or the like and the base may further be sealed, as at 5, by kieselguhr or other suitable material; the top of the furnace being provided with a cover 6, preferably of cast-iron which is capable of being rotated so that, if desired, the electrodes may thereby be made to contact with the resistor at different places to counteract the tendency of the current to flow in channels when the length of the resistor is too short as compared with its cross-section. This method of working and applying the electrodes also insures positive contact and suitable pressure on the resistor, under all conditions, thus preventing the formation of arcs.

The base 4 has pivotally mounted therein a series of grate-bars 7 provided with suitable means 8 for rotating these bars to permit of a discharge of the whole or of a portion of the material within the furnace, into the chamber provided below said bars in the base 4. This chamber may have an air-tight closure 9 at an opening therein through which the treated and cooled material deposited in said chamber, may be removed. The base 4 is insulatedly mounted, as at 10, and the operating parts of the grate are preferably provided with an insulated or insulating handle 11.

Nitrogen may be introduced through a pipe 12 which leads into the chamber below the grate, and said pipe may be insulated from the base, as at 13, in any suitable manner, and is further preferably provided with a valve 14 for regulating the inflow of nitrogen. The gaseous products of the reaction effected in this furnace, which are principally carbon monoxid, pass off from the upper portion of the furnace through a pipe 15, which may be of magnesia brick or other suitable material; and the briqueted charge 16, which in the preferred construction constitutes the resistor for the furnace, may be introduced through a hopper or

chute 17. The latter has an air-tight cover or closure 18, and is also preferably provided with a slide or gate 19 so that when gate 19 is closed a determined quantity of briquets can be introduced into the space or chamber 20 between gates 18 and 19; cover 18 being thereupon closed and the gate 19 raised to permit of this charge sliding down into the interior of the furnace.

In the present instance, in the furnace shown, there are provided five electrodes 21, of iron or other suitable material, which are insulatedly suspended by wire ropes 22 and pass downwardly through apertures in the cover 6, so that their lower extremities rest upon the upper surface of the briqueted charge. The apertures in said cover through which these electrodes extend may be sealed by means of asbestos 23, or in any other suitable manner, a cap 24 being provided to retain the asbestos against upward displacement when the electrodes are elevated. Electrical connections are made to these electrodes either directly, or, through the cover 6 which engages said electrodes, and a lead 25 from a suitable source of current is connected to the frame 25′ of the cover 6. In the present case the five electrodes 21 are of course in parallel.

Assuming that the lead 25 is positive, current passes down through the electrodes and enters the resistor mass 16 at the top thereof, traverses the same downwardly and passes out through the grate-bars 7, base 4 and returns to negative lead 26.

As above intimated, unless the briquet charge or resistor be preheated, its resistivity is so great that it is a comparatively good insulator and hence in starting a furnace of the type shown, it is preferable to preheat the briquets, at least moderately, and to further introduce but a portion of the mass, initially, into the furnace, so that for example, but a few inches of charge may rest upon the bars 7; the electrodes 21 being allowed to descend well into the furnace until they make contact with this shallow charge. Thereupon current is allowed to flow through the resistor which gradually heats up and as its temperature rises its conductivity is enormously increased, permitting additional briqueted matter to be introduced through the hopper 20 from time to time. The electrodes are then successively raised, with or without rotation, by turning the cover through a suitable angle, and allowed to drop back into contact with the upper surface of the resistor, until a charge of sufficient depth has been introduced into the furnace, after which the nitrogen current is turned on and flows up through the interstices between the bars 7 and between the briquets, effecting the cyanid-forming reaction according to the previously given equation.

Of course the nitrogen may be turned in, if desired, even when the column of briquets is short or shallow and with substantially equally good results. In practice the current is allowed to become of sufficient density to heat the resistor to from about 800° up to 1000° C.; although the cyanid-forming reaction may take place at a materially lower temperature if suitable provision be made for effecting adequate volatilization, or in some cases liquefaction, of the alkali compound or its equivalent which constitutes the source of the metal which is to become the base of the cyanogen compound formed. For example, by mixing potassium and sodium carbonate, in lieu of using but a single one of these substances, the operating temperature may be materially lowered and if other similar compounds be judiciously mixed the operating temperature may be made as low as 650° C. or thereabout. However, on account of the cheapness of the alkaline compounds, above specified, I prefer to use sodium carbonate alone, or in certain cases to use a mixture of sodium carbonate and potassium carbonate. If the temperature of the operation be sufficiently high the cyanid formed will be vaporized and will pass off together with the carbon monoxid through the tube 15, to condense as soon as the rapidly lowering temperature permits. The cyanid formed may thus be separated from the resistor by electrical heating either with or without the passage of a gas current along with the cyanid vapor, or, it may even be distilled under diminished pressure. This use of the resistor for distilling cyanid from the reaction mixture, therein, materially aids in avoiding the great difficulty of supplying heat from the outside of the retort to the interior of the poorly conducting charge. As the charge settles fresh briquets may be introduced from time to time through the hopper 20 and when the mass has been sufficiently treated, the current is turned off and the briquets are allowed to cool and harden, after which the grate bars are rotated and the now no longer plastic briquets will hence drop through readily.

By reason of the peculiar properties of the resistor above referred to, the electric heating of the mass may be made economically effective and far more uniform throughout than is possible when a similar mass of like bulk is heated by external means.

The rather remarkable change in the resistivity of such a resistor as the temperature approximates that at which the cyanid is formed, is one of the most striking features of the present process and in certain cases the difference in resistance between the briqueted charge when cold and the same charge when heated, let us say, to about

1000° C. may even reach a thousandfold or more. This permits of the use of a current of such low voltage that an ordinary lighting circuit may be used. Indeed it is possible by properly preheating the mass to employ a current of 50 volts or even lower E. M. F. It will be found advantageous in many cases, and particularly in this latter, to also preheat the nitrogen which is supplied to the furnace.

The electrical heating of the above described resistor which contains metal (resistivity 12) carbon (resistivity 1200 or more) and soda ash which is practically a fusible insulator, was at first considered impossible of practical accomplishment; but I have proven that it can be done very efficiently when the above described measures are observed. In the carrying out of my present process there is involved, therefore, a movable chemical resistor, which is in large measure automatically self-regulating by enormous change in conductivity by volatilization of one of its constituents, by internal endothermic chemical change, and by fusion of its internal insulator (soda ash, etc.); this fusion of the soda ash, or the equivalent thereof, producing a tremendous change in contact conditions between the metallic and carbon portions of the mass, as well as relatively great electrical conductivity as compared with the conductivity of the resistor when comparatively cold, and, hence, also, a correspondingly great increase in heat conductivity at high temperatures. I believe that it is hard to overestimate the value of these changes which so intimately concern the heat regulation of the reacting mass and which in effect, and in combination with suitable fuses and circuit breakers, constitute a means for practically limiting the rise in temperature at any one point in the mass beyond that temperature at which it is undesirable to conduct the process.

In this connection it may be noted that when a catalyzer such as iron is used in the solid phase, it is preferably very finely divided and the eutectic point of the carbon saturated catalyzer should, in such cases, never be exceeded since the catalytic solution surface would be thereupon seriously impaired by fusion or sintering of the iron particles, which would result in a material decrease in the extent of the catalytic surface and a consequent decrease in the effectiveness of the chemical resistor at such points therein as might be subject to such excessive temperatures.

The catalyzer used is preferably iron or a carbon dissolving metal; but it may also form nitrids to aid in the process.

I may here point out that it is highly desirable in preparing the briquets, that the catalyzer be most intimately associated with the carbon, and that during the cyanid-forming operation the current of nitrogen be directed into contact with the catalytic material while the base-forming element, or source of the same, *e. g.* sodium carbonate, is also in contact therewith, whether said element or compound be in the gaseous phase or otherwise.

Free oxygen should be carefully excluded from the reaction zone in the furnace and indeed from contact with the cyanid, after formation of the latter, so long as said cyanid or equivalent cyanogen compound is hot. Obviously also the escape of cyanogen compound fumes should be prevented, these latter being poisonous; while any material waste of such material or of the volatilized sodium carbonate is costly.

It should here be noted also that the columnar disposition of the mass of porous briquets is greatly to be preferred to any horizontal arrangement of the same, owing to the tendency of the mass under the influence of gravity, to compact itself, to prevent the formation of any large channel or channels therein through which the free nitrogen might pass without effectively participating in the cyanogen-compound-forming reaction. Again, it is by no means necessary that all of the heating current should be passed through the briqueted mass as a resistor, since I am aware of various other electrical heating arrangements which may be made reasonably effective for the effectuation of my process. I prefer that shown, however, on account of its simplicity and economy.

A series of furnaces of the type shown in Fig. 1 may be arranged as diagrammatically indicated in Fig. 3, so that a hot charge in furnace A, for example, which has been treated to form cyanid therein, may be cooled by passing cold nitrogen from the gas main 27 up through the treated charge in furnace A before delivering it through pipes covered with kieselguhr or the like to furnace B or C; suitable valves being provided to permit of any desired disposition of the gaseous current. Thus the treated briquets may be rapidly cooled preparatory to dropping them out of the reaction zone of the furnace while at the same time the nitrogen to be delivered to some other furnace in the series is highly preheated and heat is conserved. This also serves to preheat the briquets in furnaces B and C as well as to condense the poisonous alkili and cyanid fumes. It, further, permits the maintenance of a heat zone in the resistor at such temperatures that a deposit of carbon may occur therein under the influence of the catalyzer according to the equation:

(2) $2CO \rightleftarrows CO_2 + C + 38{,}960$ calories.

In so far as this takes place, it produces ashless carbon together with the simulta-

neous internal heating in the resistor itself, and saves both carbon and grinding of the same.

In Fig. 4 I have diagrammatically represented a furnace which may be continuously operated. Herein, a briquet resistor column 16′ is fed more or less continuously through a preferably flattened channel 28, portions of which, not between electrodes, may be of wrought iron pipe or the like, which further may flare for a portion of the length thereof, as at 29. The channel walls between the electrodes may be of magnesia brick or some like non-conductive material.

I prefer to water-cool the flared part 29 so as to quickly reduce the plasticity of the descending briquets before the latter reach the revolving table 30 or equivalent charge shifting or conveying device.

A screw conveyer 31 or the like may deliver the charge in a continuous stream, if desired, from a suitable hopper or receptacle 32, and the briquets before entering the reaction zone may be preheated as by burning the carbon monoxid, formed during the course of the reaction, in the pre-heater indicated.

The ascending gaseous reaction products help to internally preheat the moving resistor and may emerge from the furnace at 33. Electrodes 34, connected to bus bars 35, are suitably disposed around the channel 28 to establish the location of the reaction zone therein; said electrodes being preferably annuli of iron or other conductive material; and in the present case the electric current flows down through the chemical resistor from the upper to the lower of these electrodes. Around the pipe 28 at the reaction zone and respectively above and below the electrodes, are rings or blocks of magnesia brick, and a layer of kieselguhr is suitably retained around the outside of these rings and around the electrodes to insulate the furnace as far as practicable against loss of heat. The channel 28 is flattened as indicated in Fig. 5 to favor the preheating of the briquets, while the subsequent cooling of the charge is greatly favored by the flattening of the pipe at 29, as indicated in Fig. 6.

In operating a furnace of this type, or one of the form previously considered, it may at times be found desirable to shut off or reduce the electric current in order to enable the heat in the charge to more uniformly distribute itself; but the novel resistor employed in very large measure obviates the necessity for so doing and usually permits of a practically continuous operation of the furnace. Also, when external preheating is used, it is desirable to heat the briquets in such a manner that the hotter briquets become mixed with the cooler ones to get an average uniform heat distribution in the resistor. This may be done in a rotary furnace, or even by having a preheater at 36 around the horizontal pipe through which the briquets are conveyed to the furnace, or in other suitable manner.

If the part of the furnace between *a* and *b* is not filled with briquets, this empty space will serve as a mixing chamber as the briquets fall to the bottom. In this way, the initial formation of a comparatively good conducting annular layer of briquets on the outside of the resistor, with a subsequent tendency to conduct too large a part of the current (thus leaving a comparatively cold core on the inside), may be avoided in continuously acting furnaces.

I particularly desire to point out the value of the columnar disposition of the moving charge, not only in preventing wasteful gas channels such as I have found are apt to form when the charge is stationary and horizontally disposed, but further in preventing arcing at the electrodes; since gravity causes the briquets in the vertical column to always flow into proper contact with the electrodes and gaps across which arcing might occur are thus obviated.

If the furnace is to be operated with gases such as those containing nitrogen mixed with carbon monoxid, they are introduced, preferably highly preheated, into the reaction zone in the hot resistor at 37. By thus causing a more or less continuous advance of the resistor column through the heat or reaction zone, to the already mentioned advantages of the stationary resistor, we add those of returning the upwardly escaping volatilized (but subsequently condensed) alkali metal and compounds to the heat zone of the moving resistor, to thus produce circulation without flooding the colder portions with alkali compounds, while at the same time introducing extra heat and ash-free carbon from without according to equation (2) and avoiding the destructive action, on the cooling cyanid containing mass, of the CO_2 produced coincidentally with the deposition of said carbon in said mass. Finally on account of the high resistance of the charge when cold, in starting a continuous furnace of the type shown, the preheating of the charge is very desirable unless some such provision as that of the movable electrodes shown in the first described construction be resorted to; and whether the furnace be of the continuous or discontinuous type ash free carbon may, if desired, be deposited from carbon monoxid in the preheating zone.

In the appended claims the term "cyanogen compound" is to be regarded as of sufficient breadth to cover not only cyanid but other cyanogen containing substances,

such, for example, as cyanamid, etc.; since I am aware that, for example, by providing insufficient carbon for the cyanid forming reaction, cyanamid will be formed.

The present case is a division of my pending application, Serial Number 847,337, entitled—Process of fixing nitrogen and electric furnace for effecting the same, and filed June 25, 1914; certain of the claims herein having been originally presented in this parent application.

Having thus described my invention what I claim is:

1. The process of operating a series of furnaces to fix nitrogen therein, which comprises catalytically effecting a reaction in one of said furnaces, in which participate carbon, nitrogen, a substance containing oxygen and an element capable of acting as the base of a stable cyanogen compound to be formed by said reaction, said reaction yielding carbon monoxid in addition to said cyanogen compound, then passing the carbon monoxid, so formed, through a second of said furnaces at a temperature which permits of the conversion of said carbon monoxid to carbon dioxid, depositing carbon in a mass of reactive material in said second furnace through said conversion, and using the deposited carbon for the further production of said cyanogen compound.

2. The process of operating a series of furnaces to fix nitrogen therein which comprises effecting a cyanogen compound forming reaction in one of said furnaces while preheating another preparatory to the formation of said compound therein, by passing fumes containing said compound and also carbon monoxid, both resulting from said reaction, through said second furnace, and condensing said fumes while converting said carbon monoxid in part at least to carbon dioxid to deposit carbon for the further production of said cyanogen compound.

3. An electric cyanogen-compound-forming furnace having a resistor the resistance of which to the passage of electric current is many times as much as the resistance offered by said resistor to said current when a cyanid forming temperature has been reached in said furnace, said resistor being initially of relatively short length and said furnace having provisions to accommodate an extension of the resistor in length as its decreasing resistivity permits of such lengthening of the same, the resistor comprising a catalytic material of relatively low resistivity, which is present in said resistor in finely divided condition, and fusible material which initially tends to partially insulate portions of said catalytic material from one another, said fusible material being adapted to actively participate in a cyanogen-compound-forming reaction to be effected through the intermediacy of said catalytic material and said furnace having connections for electrically heating said resistor and for directing nitrogen into contact with said catalytic surface.

4. The process of operating a series of furnaces to fix nitrogen therein which comprises effecting a cyanogen compound forming reaction in one of said furnaces while preheating another preparatory to the formation of said compound therein, by passing fumes containing the said compound which results from said reaction in the first mentioned furnace through said other furnace, and condensing said compound to prevent the escape thereof.

5. The process of operating apparatus which includes a plurality of receptacles, to fix nitrogen in at least one of said receptacles, which comprises effecting a cyanogen compound forming reaction through the intermediacy of catalytic material present in said one of said receptacles, and then preheating an additional quantity of said catalytic material, while it is in contact with at least one of the constituents of the cyanogen compound to be formed by the process, by passing fumes of said compound, resulting from said reaction in said first mentioned receptacle, through a second receptacle in which is present said additional quantity of catalytic material, whereby to both preheat said additional material preparatory for participation in said reaction and condense said fumes.

6. The process of operating an electrically heated apparatus which includes a plurality of receptacles in one at least of which is an electrical resistor within the mass of which a cyanogen compound forming reaction may be effected, which process comprises producing said compound in said mass in vaporous condition and passing fumes of said compound through additional resistor material to preheat the latter and decrease the electrical resistance thereof preparatory for use in the process while condensing said fumes therein.

7. The process of operating an electrically heated apparatus which includes a plurality of receptacles in one at least of which is an electrical chemically reactive resistor within the mass of which is present catalytic material containing carbon in solution therein, which process comprises combining said dissolved carbon with the remaining elements of a cyanogen compound to be formed, while said elements are in contact with said catalytic material, and subjecting an additional amount of said catalytic material, while it is in contact with carbon and in a second of said receptacles, to the action of heat obtained from the condensation of gaseous reaction products resulting from the

combination of said dissolved carbon with said remaining elements, whereby to dissolve the carbon in contact with said additional catalytic material in the latter and preheat said material preparatory to the formation of an additional quantity of said cyanogen compound.

8. An electric furnace comprising a chemically active resistor containing a metallic catalyzer for the reaction to be effected, which catalyzer is in finely divided condition and acts to reduce the electrical resistance of said resistor, movably mounted electrodes to contact with said resistor, and means to displace said electrodes with respect to said resistor.

9. An electric furnace comprising a chemically active resistor containing a catalyzer which is an electrical conductor of the first class, disseminated throughout at least a large part of said resistor, electrodes to contact with said resistor, and means to effect relative movement between said resistor and at least one of said electrodes.

10. An electric furnace comprising a normally substantially stationary chemically active resistor having at least one constituent thereof adapted to actively participate in a reaction to be effected within said furnace, and having a second constituent the presence of which in said resistor tends to materially reduce the resistance thereof to the passage of electric current therethrough, said second constituent being further adapted to catalytically participate in said reaction.

In testimony whereof I have affixed my signature, in the presence of two witnesses.

JOHN E. BUCHER.

Witnesses:

H. F. HYLAND,

HELEN M. BARNBROOK.

Copies of this patent may be obtained for five cents each, by addressing the "Commissioner of Patents, Washington, D. C."