C. ELLIS.
ELECTRICAL OXIDATION OF NITROGEN.
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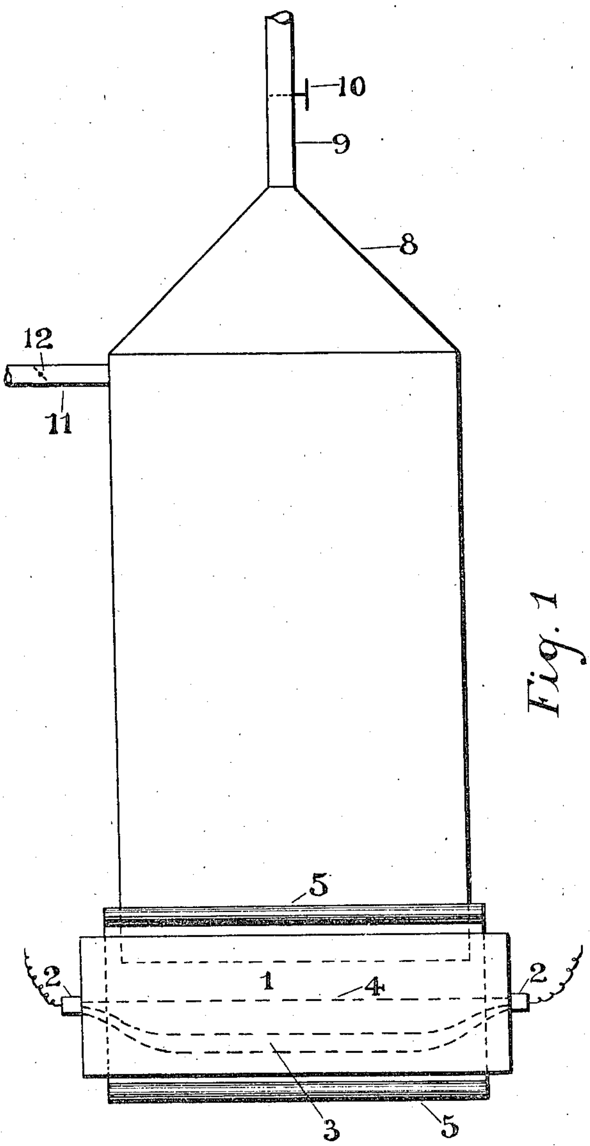

Fig. 1

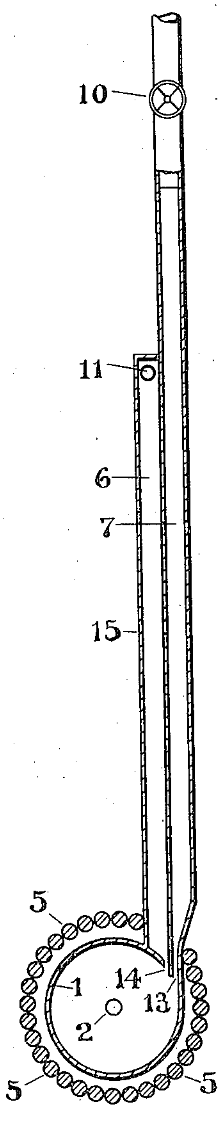

Fig. 2

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ELECTRICAL OXIDATION OF NITROGEN.

1,002,249. Specification of Letters Patent. Patented Sept. 5, 1911.

Application filed June 6, 1911. Serial No. 631,533.

To all whom it may concern:

Be it known that I, CARLETON ELLIS, a citizen of the United States, and a resident of Montclair, in the county of Essex and State of New Jersey, have invented certain new and useful Improvements in Electrical Oxidation of Nitrogen, of which the following is a specification.

This invention relates to the oxidation of the nitrogen of the air by electrical means, and relates in particular to a specific method of bringing air into contact with an electric arc, whereby a sufficient exposure is secured to said arc to produce a maximum content of oxids of nitrogen, all as more fully hereinafter described and claimed.

At the enormously high temperature of the electric arc nitrogen and oxygen apparently dissociate into atoms, and then combine to form nitric oxid as follows:—

$$N_2=2N \text{ and } O_2=2O$$
$$2N+2O=2NO$$

This is probably a pure thermal effect and not due to any specific electric effect. That thermal action is sufficient for this reaction is indicated by the work of Bunsen who found that in the explosion of air with hydrogen oxids of nitrogen were formed. Unfortunately this reaction does not progress to completion. On the contrary, it is very incomplete.

As a purely thermal effect, the simple physico-chemical laws obtain. The law of mass action determines equilibrium and from the above equation we have

$$K=\frac{(NO)_2}{(O_2)(N_2)}$$

K. is a constant depending upon the temperature and is independent of the relative amounts of gas at the beginning of the reaction. It is of course important to determine the most favorable temperature for a maximum concentration of nitric oxid. It has been found that at temperatures of 3,000 to 4,000 degrees, combination is extremely rapid. Since however, the reaction is reversible, it is very important that the gases be quickly cooled after being exposed to such a high temperature. It has been found that at a temperature of 1700° the reaction velocity is so slow, that reversion to nitrogen and oxygen is not to be feared.

It has been observed that the output of nitric oxid is smaller, the greater the energy which produces the arc. The electric arc consists of an interior portion which is very hot and an exterior portion, superposed layer, or aureola which is at a lower temperature. Apparently the nitrogen and oxygen combine in the interior portion of the arc and in passing from thence through the aureola are cooled in so gradual a way that a reversal of the reaction occurs. Hence the greater the energy supplied to the arc, the greater the aureola and the more extensive the decomposition of the previously formed nitric oxid. Theoretically therefore, it would be better to use spark discharges, but the energy of a spark discharge is so small that the method is not feasible. Of course with the aid of a system of condensers the current of a spark discharge might be greatly increased, but there are difficulties in the way of carrying out such a plan commercially. Therefore it is necessary to make use of the arc in spite of its disadvantage of aureola decomposition.

In the treatment of air by its passage across a flaming arc, great difficulty is experienced in preserving the continuity of said arc, as the air tends to extinguish or "blow out" the arc. In order to secure the rapid cooling effect desired when an intensely hot electric arc, with an extensive aureola is employed, a great excess of air is used, so that the oxids of nitrogen, as soon as formed, will be swept as rapidly as possible out of the aureola zone of decomposition, and cooled by contact with the surrounding air. Hence it follows that the content of oxids of nitrogen at the present time is usually not more than 1% or so of the total air passing through the electrically heated apparatus. This means not only the handling of a great volume of air in the subsequent process of absorption, but there is also a tremendous heat loss, due to employing so costly an agent as electric energy for heating up the 98 or 99%, or so of air which does not enter into reaction. The sensible heat absorption in this manner is very great and the arc therefore does not work to good advantage under such circumstances. Probably not more than 3% or 4% of the electric energy is absorbed in the actual process of making nitric oxid. Of course a certain portion of this heat is recovered in various ways, as for example, passing the gases from the electric furnace under steam boilers or evaporating pans, but the generation of heat for such purposes in this way is of course costly, and undesirable.

Besides the flaming arc mentioned above, in which the air passes transversely across the arc, there also exists the method of oxidizing nitrogen in which air is passed spirally about an elongated electric arc and in which case aureola contact is of course pronounced.

The present invention has for its object, the production of an arc of great length, so that each furnace or unit of apparatus is capable of handling a great volume of air and in which there is a minimum of aureola contact with respect to the length of the arc. Preferably for this purpose, I form a long electric arc between two electrodes, or a plurality of electrodes, and cause the air to pass in a circular path in a plane at right-angles to the axis of the normal arc passing between the electrodes, and I cause said arc to be deviated from its normal axis into the path of said current of air. Obviously, if the arc is stationary in respect to the travel of the gaseous current, it is liable to be extinguished by the blast of air against it, and to overcome this difficulty I cause the arc to revolve about its normal axis in the path of the current of air and at a speed either slightly greater than the rate of travel of the air current, or preferably at a slightly lower speed, so that a lag effect is obtained. Hence, as the arc is traveling practically at the same speed as the air current, it is prone not to be extinguished by the said current and I thus secure the advantage of a greatly elongated transverse arc with a minimum of aureola contact. That is to say, the extinguishing effect of the air blast is minimized by causing the arc to travel in this manner, and the aureola contact is greatly reduced over methods which involve the travel of the air longitudinally with the arc. The process therefore yields a large output with simplicity of furnace construction, small number of units and therefore reduced labor cost and maximum electrical efficiency.

In the accompanying drawing, means are shown for carrying out my invention.

In Figure 1, is shown a plan view of a furnace adapted to carry out the method above indicated, and Fig. 2 is a longitudinal section of Fig. 1.

In the drawings, 1 is a cylindrical shell or furnace, having at each end the electrodes, 2; an arc 3, is formed between the electrodes, 2. The normal axis of this arc is indicated by 4. The furnace shell is surrounded by electromagnets 5, so arranged that the magnetic effect may be caused to travel in a wave or circular path about the furnace shell. An inlet, 13, is provided for the incoming air which enters the furnace in tangential fashion, so that the air current is caused to pass around the normal axis of the arc in a concentric way. An outlet 14, is provided for the heated gases. The passages 6 and 7, contain respectively, the outgoing and incoming gases and form a continuous regenerator or heat recuperator. Both the inlets and outlets have the form of slits, so that the air is caused to travel in a concentric path in a plane at right angles to the normal axis of the arc.

The initial air enters the regenerative system through the pipe 9, the amount introduced being controlled by the valve 10. It is supplied under pressure. A distributing expanded portion 8 leads from pipe 9, to the flat recuperator. An outlet for the heated gases is shown at 11, and this is provided with the regulating damper 12.

The operation of the process with the illustrative apparatus described is as follows:—An arc is formed between the electrodes 2, using any suitable means for starting said arc. A pressure of 4,000 to 5,000 volts may be used and in the case of a polyphase system several furnaces may be connected to utilize the phase effects economically. An electric current is sent through the control magnet system 5, and by means of a commutator or other distributing system or method of connecting, magnetism is locally established and caused to travel about the furnace shell, thereby causing the arc to be deviated from its normal axis and revolve in a position between the normal axis and the furnace shell. Air is admitted through the pipe 9, passed along the recuperator passage 7, enters the furnace tangentially at slit 13, passes concentrically about the normal axis 4, and leaves at the slit exit 14, passing through the recuperator passage 6, and to the absorbing towers, etc., by the conduit 11. The rate of movement of the arc with respect to the speed of travel of the air is regulated in a predetermined manner; the arc being speeded either to keep slightly in advance of the air current or preferably to lag slightly, so while it is traveling substantially at the same rate as the current of air, its lag is sufficient to permit of the passage of the major portion of the air current through the said arc. In so doing, the oxygen and the nitrogen are heated to the dissociating temperature and combine to form nitric oxid. They are quickly however, removed from the zone of reverse decomposition, as the present arrangement, it will be seen, provides a minimum of aureola contact. In passing through the recuperator the heated gases are gradually cooled and further oxidation may take place to some extent, depending upon the length of the recuperator. When nitric oxid is cooled below 600° it takes up further oxygen, and becomes nitrogen dioxid, NO_2. The hot gases leaving by conduit 11, may be passed under a steam boiler for the generation of steam, or may be sent under evaporating pans containing liquors from absorption operations. From thence the gases may be conducted to a tower through which lime or other alkalis in solution or suspension is passed, so as to form a mixed nitrate or nitrite. Or the gases may go to water absorption towers where nitric acid is formed and recovered.

As stated, the air which enters the apparatus is under pressure, which pressure may be merely sufficient to propel the air through the system and this movement of the gaseous current may be facilitated by an exhaust pump placed in the exit conduit 11, which pump is not here shown. I prefer, however, to maintain a plenum in the furnace 1, and for this purpose I preferably introduce air under several pounds pressure. By having all the joints of the furnace air tight, this pressure may be increased to 2 or 3 atmospheres in which case the concentration of the nitrogen and oxygen becomes greater as it were, that is to say, the "partial" pressure of the nitrogen, for example, seemingly increases, and the action of the arc is more effective. The presence of water vapor in the air current is detrimental to the extent that variations in the humidity of the air causes variations from time to time in the character of reactions and I therefore prefer to have the air enter the apparatus either dried to a certain definite or constant condition of dryness, or else moisten the air so as to preserve it at a constant degree of humidity. Reactions between gases are generally facilitated by the presence of a small amount of moisture, but this amount does not need to be large and in fact, a large amount of water vapor in the air current would be detrimental, as the sensible heat of water is very great and increases rapidly with increasing temperature. In the preferred form of my invention, I therefore enter the air in the apparatus under constant condition of humidity.

The interior of the furnace shell may be lined with difficultly fusible bodies having a catalytic action, and such catalysts as thorium oxid, vanadium oxid, titanium oxid, uranium oxid, boron oxid, aluminum oxid and the like may be applied to the interior of the furnace shell. The exterior of the shell may be water-cooled if desired in the manner well known in the furnace art.

In preheating the air supplied to the furnace, it will be evident from the foregoing that it is undesirable to use a preheater of so effective a character that the entering gases would have a temperature above the critical temperature of reversion, which as stated is commercially speaking, 1700°. It is however, desirable to heat the air as near as possible to this temperature without causing retrogressive changes of a material character. The present process thus secures a large output with a maximum electrical efficiency and a sufficient time contact is secured to derive the necessary heating effect by the proper advance or lag of the arc and this differential acceleration or retardation is regulated in a predetermined manner by magnetic control or otherwise.

With a long arc and an air current traveling from one end of the arc to the other, there is as stated, unavoidable aureola contact so that the union and decomposition of oxygen and nitrogen may take place an infinite number of times, during the long period of travel, resulting in the imperfect application and waste of electric energy. The present process overcomes this difficulty while taking advantage of the commercially valuable features of the long arc.

The illustrative method herein given depicts the invention in its preferred form, but it is evident that various modifications may be made in the manner in which the air is caused to circulate about the normal axis of an arc, and the manner in which said arc is caused to travel through the air current, and I do not wish to limit myself to the exact procedure herein described, but may invoke the doctrine of equivalency, so far as same is herein applicable. For example, the apparatus shown in the drawings may be inverted thus impelling the circular current of air in the furnace 1, in a reverse direction, or anti-clock-wise. Under such circumstances, the relatively cool air enters near the top of the furnace and becomes heated more and more as it passes forward and downward in its reverse circular path finally coming in contact with the hottest portion of the arc and forming nitric oxid.

To preclude the possibility of disturbing eddies, care should be taken to avoid too close a juxtaposition of the inlet and outlet slits. If these slits are then not properly directed a slight eddy is set up which affects the arc somewhat if the latter is revolving, but not appreciably if the arc is stationary. The best method of reducing exit-eddy to a minimum is to have the exit passage or slit, set nearly in the line of a tangent at the point of exit. Further stability can be given the flame in the inverted type by using air of constant moisture content under pressure. With an alternating current it often becomes desirable to introduce an inductive resistance into the circuit, usually directly in series with the arc. With very large chambers a plurality of air inlet slits may be required to secure the necessary momentum to the air current and preserve its substantially circular path in the furnace chamber. The inverted type also has another advantage in that the recuperative passage may be more readily constructed to give a rapid transfer of heat. Preferably in a continuous regenerator the hotter flue should be beneath the cooler one and this arrangement is better carried out in the inverted apparatus.

Other modifications will now suggest themselves to those skilled in the art and I recapitulate by stating that my invention relates to the method and its equivalents of forming an electric arc, or plurality of arcs, preferably greatly elongated, in passing about the arc, or its normal axis or the plurality of arcs or their normal or common axis a current of air, or mixture containing essentially oxygen and nitrogen, in causing the current to take a circular or substantially circular path thereabout in planes substantially at right angles thereto; in withdrawing the air after making such circuit, at a point preferably adjacent to the initiation of the circular flow, in preferably causing the arc to deviate from the normal axis by magnetic action or otherwise, and to revolve about said axis at a speed preferably slightly less than the speed of the current of air, etc., and to introduce the air or other gaseous mixture into the electrical heating zone, preferably in a preheated state, preferably under pressure and preferably of constant moisture content.

What I claim is:

1. The process of oxidizing nitrogen which consists in forming an elongated electric arc, in passing a current of oxygen and nitrogen concentrically in a plane at right angles to the normal axis of said arc and in magnetically deviating said electric arc into the path of the gaseous current.

2. The process of oxidizing nitrogen which comprises forming an electric arc, in passing a current of air at a predetermined speed concentrically about and substantially in a plane at right angles to the normal axis of the arc and in causing said arc to be deviated from its normal axis and to revolve about said axis at a predetermined speed in the gaseous current.

3. The process of oxidizing nitrogen which comprises forming an electric arc, in passing a current of preheated air at a predetermined speed concentrically about and in planes substantially at right angles to the normal axis of said arc and in causing said arc to be deviated from its normal axis, and to revolve in the said gaseous current and at a rate of speed slightly less than that of the current of the gases.

4. The process of oxidizing nitrogen which comprises forming an electric arc, in passing a current of preheated air concentrically about and in planes substantially at right angles to the normal axis of said arc and in causing said arc to be diverted from its normal axis by magnetic action and to revolve about said normal axis in the path of the gaseous current at a speed less than that of said gaseous current.

5. The process of oxidizing nitrogen which comprises forming an elongated electric arc, in passing a current of preheated air of constant moisture content concentrically about and in planes substantially at right angles to the normal axis of the arc and in causing the arc to be deviated from its normal axis and revolve thereabout at a speed slightly less than that of the rate of travel of the gaseous current.

6. The process of oxidizing nitrogen which comprises forming an electric arc, in passing a current of preheated air at a predetermined speed concentrically about substantially the entire length of the arc and in planes substantially at right angles to the normal axis of the arc and in causing said arc to revolve about its normal axis by magnetic action, at a speed slightly less than that of the said gaseous current in withdrawing the said gaseous current after it has made a substantial circuit of the normal axis of the arc, and in transmitting a portion of its heat to the incoming cool air.

Signed at Montclair, in the county of Essex and State of New Jersey this 25th day of May A. D. 1910.

CARLETON ELLIS.

Witnesses:
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SYDNEY M. SPEDON.