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PROCESS AND APPARATUS FOR THE PRODUCTION OF METALLIC NITRIDS.

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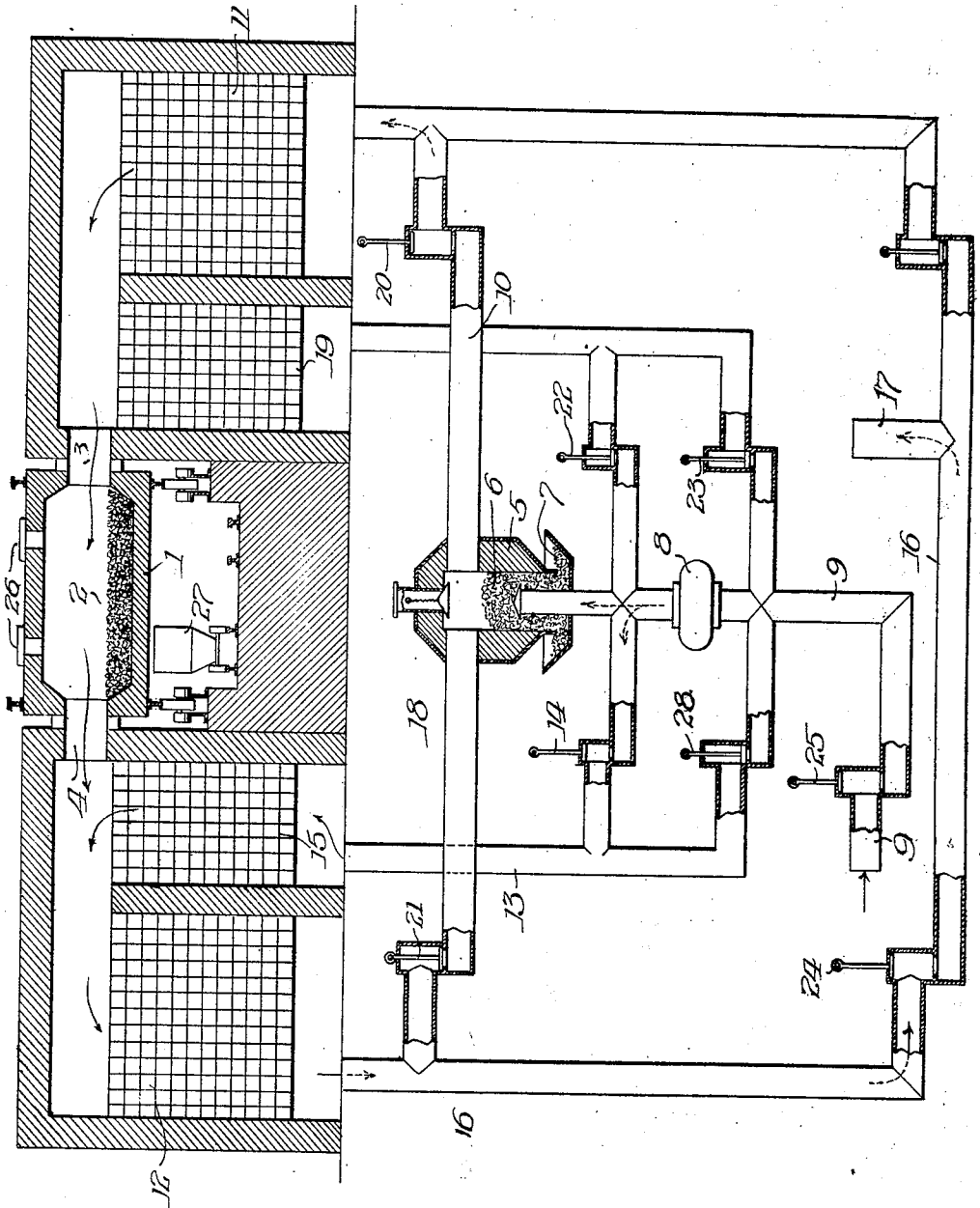

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UNITED STATES PATENT OFFICE.

FREDERICK T. SNYDER, OF OAK PARK, ILLINOIS.

PROCESS AND APPARATUS FOR THE PRODUCTION OF METALLIC NITRIDS.

1,305,414. Specification of Letters Patent. **Patented June 3, 1919.**

Application filed January 16, 1915. Serial No. 2,534.

To all whom it may concern:

Be it known that I, FREDERICK T. SNYDER, citizen of the United States, residing at Oak Park, in the county of Cook and State of Illinois, have invented a certain new and useful Improvement in Processes and Apparatus for the Production of Metallic Nitrids, of which the following is a full, clear, concise, and exact description.

My invention relates to a process and apparatus for the production of metallic nitrids, and its object is to overcome certain difficulties heretofore met with in the production of such nitrids.

It is well known that certain compounds of metal, especially the oxids, when heated to a high temperature under reducing conditions and simultaneously subjected to the action of nitrogen, will be largely converted into nitrids. Difficulty, however, has been met in maintaining the material under treatment at a suitable uniform temperature. To obtain a commercial percentage of nitrogen, it is necessary to heat the furnace charge to a high temperature. If the charge is heated much above the production temperature, the nitrid formed begins to break up again. As is well known, nitrogen acts as if it were very inert. This inertness is largely due to the strong hold which the nitrogen has for itself in the nitrogen molecule. Once the nitrogen molecule is broken up, the nitrogen combines readily with other available materials, especially the basic metals, with the formation of nitrids.

Heretofore nitrids have been formed by heating compounds of the basic metals to a high temperature, and then passing relatively cool nitrogen over the compounds. The compounds have been heated either by passing the heat through the walls of the retort, or internally, by the application of electricity. In either case, the metallic compounds are first heated, and then transfer their heat to the nitrogen gas. In my invention the entire quantity of heat required for the reaction is first introduced into the nitrogen gas by heating the gas up to a high temperature, and the basic metal compounds are then heated by passing this hot nitrogen gas over them.

The various features of my invention may be more readily understood by reference to the accompanying drawing, showing diagrammatically one embodiment of my invention, and in connection with a description of the manner of operating the structure to treat the material.

It will be understood that, except as defined in the claims, the invention is not limited to the treatment of any particular material. For the purpose of description, I will describe the process in connection with the production of aluminum nitrid from alumina.

Referring to the drawings, the furnace 1 is of the rotary type. Contained in the furnace is the oxid of the basic metal, in the case assumed, the oxid of aluminum Al_2O_3. Mixed with the alumina is the reducing carbon in a finely divided form. There is passed through this furnace a gas containing nitrogen, heated to a temperature of approximately 2000° C., which, in this case, is the gas temperature required to produce the reduction of the oxid by carbon and the simultaneous formation of metallic nitrid. The gas passes into the furnace through the opening 3, and out through the opening 4. After a short interval, the direction of the gas travel is reversed.

To maintain the gas passing into the furnace at a temperature of approximately 2000° C., a gas producer 5 is employed for converting air from a blower 8 into producer gas. The producer gas formed is largely composed of nitrogen; it, however, contains small amounts of hydrogen and hydro-carbons. This gaseous mixture passes from the producer 5 through a pipe 10 into a regenerator 11. During the previous period, the upper part of this regenerator has been heated by the passing gas to some 2300° C., and as the nitrogen gas passes through it, this gas is heated up to between 2000° and 2100° C. This gas in passing through the furnace 1 gives up heat to the charge 2 in the furnace, and leaves the furnace at about 1800° C. The blower 8, in addition to supplying air for the producer 5, also blows air through a valve 14 and pipe 13 into an air regenerator 15. Passing through the regenerator 15, the air is heated up to some 1600° C., and, meeting the producer gas as it leaves the furnace 1, mixes with the producer gas and burns it. The resulting flame reaches a high temperature and passes out through a regenerator 12, heating the upper bricks of this regenerator 12 to about 2500° C. The outgoing gases leave the bottom of regenerator 12 at about 600° C. and pass out through a pipe 16 to a

stack 17 where they are discharged into the air.

After an interval of about ten minutes, the brick work in the regenerator 12 becomes so highly heated that it does not economically absorb further heat from the gases. Furthermore, such excessive heating might damage or destroy the apparatus. At this time valves 14, 24 and 25 are closed, and a valve 28 opened. This results in the flow of air through the regenerator 15 being stopped and the gas from the furnace 1 being caused to flow down through the air regenerator 15, through the valve 28 and the blower 8 into the producer 5, and then, by the pipe 10 and valve 20, into regenerator 11. Part of this further heat is given up to the material in the furnace 1, and the remainder goes to heating the regenerator 15. Any CO_2 gas or moisture which is formed in the furnace, or in the circulating pipes, is destroyed by passing the gas through the white hot carbon in the producer 5, so that the gases which go to the regenerator 11 and thence to the furnace are at all times entirely free from CO_2 gas and moisture. After a further interval of about ten minutes, the valve system is reversed so that the same cycle of operations take place from the other end of the furnace.

The reduction of the basic metals absorbs large quantities of heat. This heat is supplied by the hot nitrogen. In order to do this, it is necessary that the nitrogen should be heated much more than the charge 2 in the furnace 1. Heating the nitrogen to this high temperature has a tendency to break up the nitrogen molecules, and in this way make the nitrogen more active in entering into combination with the basic metal as reduced by the carbon. It is this greater activity of the nitrogen, due to its temperature being higher than that of the material on which it is acting, which contributes largely to the successful operation of my invention. After the gas has been passed alternately back and forth through the furnace 1 a sufficient length of time to transform most of the available basic metal into nitrid, the passage of the gas is stopped, and the furnace 1 is emptied by opening the doors 26 and allowing the charge to discharge into the car 27 by slowly revolving the furnace 1. A fresh charge is then placed in the furnace 1, the doors 26 closed, and the operation repeated.

It will be noted that the gas is passed alternately over the furnace charge first in one direction and then in the other. By this means all parts of the charge are brought uniformly up to the reduction temperature, and the raising of any portion of the charge to the temperature of decomposition of the nitrids formed is largely avoided. A considerable part of the advantage gained by my invention is due to the fact that the gases are passed alternately in opposite directions over an isolated charge, which can be retained for treatment until the proper result has been reached. This is of great importance commercially, as it permits of retaining the charge in the furnace until the maximum conditions of nitrification are secured.

What I claim is:

1. The process of making compounds of nitrogen, which consists in passing nitrogen bearing gas over a mixture of an oxid of a basic of metal and carbon, the temperature of the gas being above the reduction temperature of the oxid, subsequently burning the gas with air to raise the temperature of the gas, then passing the gas through regenerators used to heat further amounts of incoming gas, then circulating further amounts of the nitrogen bearing gas in a closed cycle through a regenerator, a gas producer containing carbon and the original regenerator to transfer further amounts of heat from the original regenerator to the furnace charge.

2. The process of producing aluminum nitrid, which consists in directing air from a blower through a gas producer to make producer gas and directing said producer gas alternately through two regenerators and over an intermediately located charge of alumina and carbon, permitting air from said blower to unite with said gas after it leaves said furnace to burn said gas and maintain the regenerators at a temperature considerably above the nitrification point of the charge and continuously rotating the charge containing furnace during said operation.

3. The process of making compounds of nitrogen, which consists in passing over a mixture of an oxid of a basic metal and carbon, a great excess of superheated producer gas, the temperature of said gas being above the reduction temperature of the oxid of the metal, subsequently burning the gas with air to raise the temperature of the gas and then passing the products of combustion through a regenerator used to heat the subsequent incoming gas, then shutting off the air supply and passing the unburned nitrogen gas through the gas producer to maintain the circulation of the gas over the charge without further heating of said regenerators.

4. High temperature apparatus for the production of aluminum nitrid comprising a rotatable furnace for the charge, a pair of gas regenerators each connected with one end of said furnace, a gas producer connected with said regenerators, a blower connected to said gas producer, valves permitting a reversal of flow of reducing gas through said furnace, means for supplying

air from said blower to said gas after it passes through said furnace whereby it burns in one regenerator after being superheated in the other, and means for permitting the gas to be drawn from the furnace by said blower and repassed through said blower, said gas producer, and the regenerator on the inlet side of said furnace.

5. High temperature apparatus for the production of aluminum nitrid comprising a rotatable furnace for the charge, means for passing nitrogen gas through said furnace alternately in opposite directions, means for burning said gas after it passes the furnace to maintain a regenerative action at a high reduction temperature, means for discharging the waste products of combustion, and means for passing heated gas through said furnace in a closed cycle during each reversal.

6. High temperature apparatus for metallurgical purposes comprising a rotatable furnace for the charge, means for passing combustible gas through said furnace alternately in opposite directions, means for burning said gas after it passes the furnace to maintain a regenerative action at a high temperature, and means for subsequently passing such regeneratively heated gas alternately in opposite directions through said furnace without combustion.

7. The process of producing nitrids which consists in directing air from a blower through a gas producer to make producer gas and directing said producer gas alternately through two regenerators and over an intermediately located charge of the oxid of a metal and carbon, permitting air to unite with said gas after it leaves said furnace to burn said gas and maintain the regenerators at a temperature considerably above the nitrification point of the charge and continuously rotating the charge-containing furnace during said operation.

8. The process of producing nitrids which consists in directing air from a blower through a gas producer to make producer gas and directing said producer gas alternately through two generators and over an intermediately located charge of the oxid of a metal and carbon, permitting air to unite with said gas after it leaves said furnace to burn said gas and maintain the regenerators at a temperature considerably above the nitrification point of the charge, continuously rotating the charge-containing furnace during said operation and passing the regeneratively heated gases alternately in opposite directions through said furnace without combustion.

9. In combination, a rotary furnace, a pair of air regenerators connected to opposite sides thereof, a pair of gas regenerators connected thereto beyond said air regenerators, a gas producer, a blower for supplying air thereto, connections from said gas producer to said gas regenerators and two sets of connections from opposite sides of said blower to said air regenerators to permit reversal of flow through said furnace and also to permit flow in a closed cycle.

10. In combination, a furnace, two pairs, of regenerators, one pair being connected to each side of said furnace, means for passing a combustible gas through the regenerators on one side, through said furnace and through a regenerator on the other side, means for passing air through the remaining regenerator on the other side whereby said gas burns as it passes through said regenerator, means for reversing the flow of gases through said respective regenerators, and means for shutting off said air supply and permitting said gas to flow in a closed cycle without burning.

11. In combination, a furnace, two pairs of regenerators, one pair being connected to each side of said furnace, means for passing a combustible gas through the regenerators on one side, through said furnace and through a regenerator on the other side, means for passing air through the remaining regenerator on the other side whereby said gas burns as it passes through said regenerator, means for reversing the flow of gases through said respective regenerators, and means for shutting off said air supply and permitting said gas to flow in a closed cycle without burning, the flow in a closed cycle taking place through said air regenerators in a direction the reverse of that through which the air flows.

In witness whereof, I hereunto subscribe my name this 9th day of January, A. D., 1915.

FREDERICK T. SNYDER.

Witnesses:
GEORGE E. FOLK,
MCCLELLAN YOUNG.

Corrections in Letters Patent No. 1,305,414.

It is hereby certified that in Letters Patent No. 1,305,414, granted June 3, 1919, upon the application of Frederick T. Snyder, of Oak Park, Illinois, for an improvement in "Processes and Apparatus for the Production of Metallic Nitrids," errors appear in the printed specification requiring correction as follows: Page 2, line 79, claim 1, strike out the word "of"; same page, line 117, claim 3, strike out the article "the"; and that the said Letters Patent should be read with these corrections therein that the same may conform to the record of the case in the Patent Office.

Signed and sealed this 17th day of June, A. D., 1919.

[SEAL.]

J. T. NEWTON,

Commissioner of Patents.

Cl. 23—13