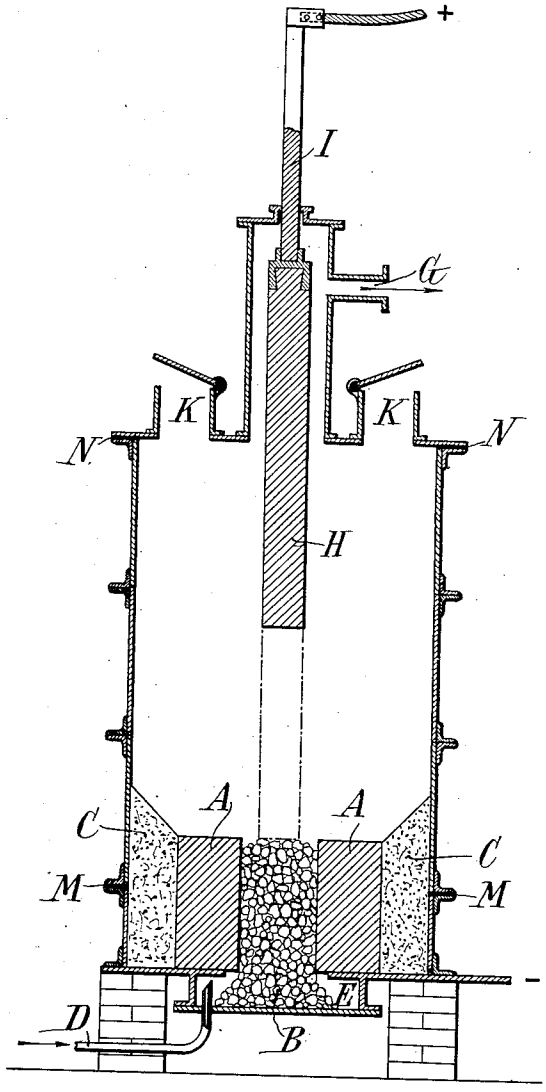
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ELECTRIC FURNACE INTENDED CHIEFLY FOR THE MANUFACTURE OF NITRIDS.

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1,158.899.

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UNITED STATES PATENT OFFICE.

GEORGES COUTAGNE, OF LYON, FRANCE.

ELECTRIC FURNACE INTENDED CHIEFLY FOR THE MANUFACTURE OF NITRIDS.

1,158,899. Specification of Letters Patent. Patented Nov. 2, 1915.

Application filed May 21, 1914. Serial No. 839,963.

To all whom it may concern:

Be it known that I, GEORGES COUTAGNE, a citizen of the Republic of France, residing in Lyon, Rhône, France, have invented certain new and useful Improvements in Electric Furnaces Intended Chiefly for the Manufacture of Nitrids, of which the following is a specification.

Thomas Leopold Willson patented in England as No. 21755 of November 15, 1895, the manufacture of aluminium nitrid by heating in an electric furnace in the presence of nitrogen a mixture of carbon and aluminium or an oxygen compound of aluminium. The aluminous material to be used with best advantage is obviously natural alumina, *i. e.* bauxite. The source of nitrogen that is most economical is evidently the gas called "generator gas" which consists chiefly of nitrogen and carbon monoxid and that is obtained by passing air over incandescent coal. Generator gas has besides already been proposed as a source of nitrogen for the manufacture of aluminium nitrid in the above mentioned patent to Willson and in American Patent No. 741396 of October 13, 1903 to Guillaume de Chalmot.

On heating in a small electric furnace, arranged as described and shown by Willson, a mixture of bauxite and coal such as coke or anthracite and by blowing generator gas into the incandescent mass, it is indeed possible to obtain a more or less abundant amount of aluminium nitrid. If one tries to use this process of manufacture on an industrial scale however by using the one or the other of the four devices shown by Willson, on having enlarged the same and even on having considerably modified the relative proportions of the various parts, no satisfactory result is obtained.

A great number of devices that differ considerably from those of Willson and most of which are very ingenious have been contrived, patented and tried during these last years. The difficulties to be overcome are such however that the manufacture of aluminium nitrid has not been practicable hithereto on an industrial scale. It appeared preferable thereafter to go back to Willson's process and simply improve the furnace with intermittent working proposed by the American inventor in 1895, without trying to contrive a furnace with continuous working. After a great many tests really practical results have at last been obtained with the furnace described and shown in the present specification. This furnace is derived from the device of Figure 1 of Willson's patent comprising essentially as an upper electrode a cylindrical solid vertical and movable electrode, as a lower electrode a mass of agglomerated carbon in the form of a crucible and for the introduction of the generator gas an axial channel in the lower electrode. In these original arrangements that have been retained four modifications or new features have been made that are all very important and absolutely essential. The same are as follows:

1. The lower electrode is made of a large tube A (see the accompanying figure of the present specification), which is vertical, cylindrical or polygonal, very thick and constitutes an electrode of agglomerated coal. The inside space of this tube is filled with coke B, and a layer C of refractory material forming a bad conductor of electricity, for instance ordinary crushed bauxite reduced to sand, surrounds the tube A. The tube A must be fairly high so that the hearth of steel which supports the tube and through which the current is fed is never heated too strongly. The inside of tube A must be large enough that the generator gas which is supplied under pressure through tube D and first spreads out in the annular space E, can rise in the mass of coke in fairly large quantity and without too high a speed.

2. The furnace is entirely surrounded by a metal casing composed of cast iron or cast steel parts or of iron or steel plates with air tight joints, so that the nitrogenized gas may on the one hand be injected under pressure and can overcome the resistance caused by the mass of material treated in the furnace and in order that the residual gases consisting chiefly of carbon monoxid and a little nitrogen that has not been combined can on the other hand be collected, passed out and used any distance away from the furnace. These gases pass out at the opening G. The latter may be arranged at any other place instead of the one shown, provided it is always at the upper part of the furnace. The movable electrode H is suspended on a steel rod I which slides in a stuffing box. Openings K are arranged in a suitable number all around the electrode on the metal lid of the furnace and serve for introducing the mixture of bauxite and coal. These openings must be provided with

hinged lids or any other contrivance that will prevent the gases from passing out. Insulating joints are provided between the several shell rings of sheet iron that are built up to form the casing of the furnace. It has been found for instance that a good arrangement is obtained by making two insulating joints, one at the bottom of the furnace between the first and the second shell ring, at M M, and the other at the top where the lid is joined onto the last shell ring, at N N.

3. The whole side wall of refractory material is suppressed in the part of the furnace above tube A. This modification in Willson's furnace seems paradoxical at first sight, but experience has shown its efficiency and by this means one of the greatest difficulties in the manufacture of aluminium nitrid in the electric furnace has been overcome. On carefully studying the conditions of working of the new furnace described in the present specification it will be understood moreover that by localizing the work of the electric current in the axis of the furnace, the mixture of bauxite and coal which is next to the sheet iron casing is not penetrated by the current and does not undergo the reaction, so that it remains in the state of a pulverulent mixture or a mixture that is slightly agglomerated and only conducts heat to a slight extent, so that it is perfectly apt to act as a non-conducting material.

4. The upper shell rings of the furnace have to be arranged so that they can be taken down and removed easily and quickly, so as to be able to remove the mass of aluminium nitrid formed after each operation and take it over to another work shop. In industrial furnaces of high power each shell ring can be made of several parts. Instead of using superposed cylindrical shell rings the metal casing may be made in any other manner, for instance of walls, all of which or part only can readily be removed.

This furnace is used in the following manner: When the movable electrode has been brought right down and even engaged a little in the central space inside tube A, the current passes from one electrode to the other through the coke and this condition is maintained for some time, so as to thoroughly heat the annular hearth and the coke. The supply is then started which is made thereafter at regular intervals through the upper openings K, and at the same time the injection of the generator gas through tube D is begun. An operation lasts several days and is finished when the whole inner cavity or recess of the furnace is filled. The weights of each charge are obtained by experience as well as the interval between two consecutive charges that is most suitable for the capacity and power of each furnace. Care must be taken particularly at the beginning of the operation to avoid feeding the furnace too quickly with too large quantities of the mixture of bauxite and coal, as otherwise a large metallic mass of an alloy of iron, silicium and aluminium is obtained on the tubular hearth that is not easily penetrated by the nitrogen, so that this mass is only partly transformed at its periphery into aluminium nitrid. If the charges are supplied on the contrary at longer intervals and are reduced to an adequate extent, each charge is partly brought to the temperature of the reaction and even partly transformed into nitrid when the following charge is fed, and as the particles of metallic alloy contained in the mass are small and disseminated in a mass permeable to gas, they gradually allow their aluminium to combine with the nitrogen.

The furnace could evidently be supplied also in a continuous manner, but this would not be so simple. The example of blast furnaces shows that working on a large scale can quite well be effected with discontinuous charges.

When the operation is finished the electric current and the current of gas containing nitrogen are interrupted and on having cooled down and opened the furnace a large cake of aluminium nitrid is found that is surrounded by a slightly agglomerated layer of a mixture of bauxite and coal, that can easily be separated from the nitrid and can be put aside to be introduced again in the following operation together with the mixture of new material.

The crude aluminium nitrid also contains grains or small clusters of metallic alloy *i. e.* ferro-silicium with a varying amount of aluminium. On having reduced and crushed the mass these metallic parts can easily be separated by any suitable means, for instance by an electromagnetic separator. For certain purposes however such as the manufacture of ammonia in which the alumina is returned to the nitrid furnace, this separation is not necessary. These particles of metallic alloy and particularly the larger masses of this same alloy that are sometimes found in the cake, principally in its lower part, moreover fall into dust after some time, particularly in moist air, probably owing to the action of small quantities of aluminium carbid disseminated in the substance of these masses or particles. This property can therefore be used even together with the addition of water or a treatment with water vapor at atmospheric pressure; this treatment does not decompose the aluminium nitrid. The reduction to dust can therefore be accelerated, as the same largely facilitates the separation of the various products mixed with the nitrid in the crude cake taken out of the furnace.

In the parts of the cake that are nearest to the electrode geodes are found that are covered with crystals of nitrid and carborundum, which give an idea of the temperature attained in the furnace. It is known indeed that according to the experiments of Tucker and Lampen (*Journal of the American Chemical Society*, 1906), carborundum only begins to assume a crystalline form toward 1950° C.

When using ordinary red bauxite an excess of coal, in the mixture of bauxite and coal; with regard to the amount calculated according to the chemical formulæ as being necessary for completely deoxidizing the bauxite, is not necessary either for increasing the permeability for the gases or for increasing the electric conducting properties of the mixture. If another kind of bauxite is used however or other aluminous material an excess of coal is of advantage, as proposed already in 1896 by G. de Chalmot.

Generator gas containing about 35% of carbon monoxid produces very satisfactory results; by using pure nitrogen however the yield would of course be improved. According to the cost prices at which generator gas or nitrogen can be had in each case, it will be of advantage to use either the one or the other of these gases. As the nitrid remains subjected to a high temperature after the moment it has been formed till the end of each operation, the retrogression recently pointed out and studied by Fraenkel (*Zeitschrift für Elektrochemie*, April 15, 1913, p. 362–373) and that is particularly important at low temperatures (1500 to 1600°), does not take place in this furnace; this is a very considerable advantage of the new process.

The furnace and process described in this specification can be used for the manufacture of other nitrids than aluminium nitrid.

What I claim is:

1. An electric furnace for the production of nitrids comprising a chamber having electrodes therein, the lower one of said electrodes being in the form of a tubular carbonaceous block and forming a part of the base of the furnace, and a filling of coke in said tubular block.

2. An electric furnace for the production of nitrids comprising a chamber having electrodes therein, the lower one of said electrodes being in the form of a tubular carbonaceous block surrounded at its sides by refractory material and forming a part of the base of the furnace, and a filling of coke in said tubular block.

3. An electric furnace for the production of nitrids comprising a chamber having electrodes therein, one of said electrodes being in the form of a tubular carbonaceous block, a filling of loose carbonaceous material in said tubular block, and an inlet for a supply of nitrogen in communication with said furnace through said loose filling.

4. An electric furnace for the production of nitrids comprising a chamber, a pair of electrodes therein, one of said electrodes being in the form of a stick or pencil and the other in the form of a tubular carbonaceous block, a filling of loose carbonaceous material in said tubular block, and an inlet for a supply of nitrogen in communication with said furnace through said loose filling.

5. An electric furnace for the production of nitrids comprising a chamber having electrodes therein, one of said electrodes being in the form of a tubular carbonaceous block, a loose filling of coke in said tubular block, and said chamber above said tubular block being of relatively large capacity and the inner surface being of iron and unlined with refractory material.

6. An electric furnace for the production of nitrids comprising a chamber having electrodes therein, one of said electrodes being in the form of a tubular carbonaceous block, a loose filling of carbonaceous material in said tubular block, and an inlet for the supply of nitrogen in communication with said furnace through said loose filling, the tubular space in said tubular block being relatively large, the gas being capable of spreading out therein and passing through at a relatively low speed.

In witness whereof, I have hereunto signed my name in the presence of two subscribing witnesses.

GEORGES COUTAGNE.

Witnesses:

JEAN MILLION,

VINCENT MILLION.