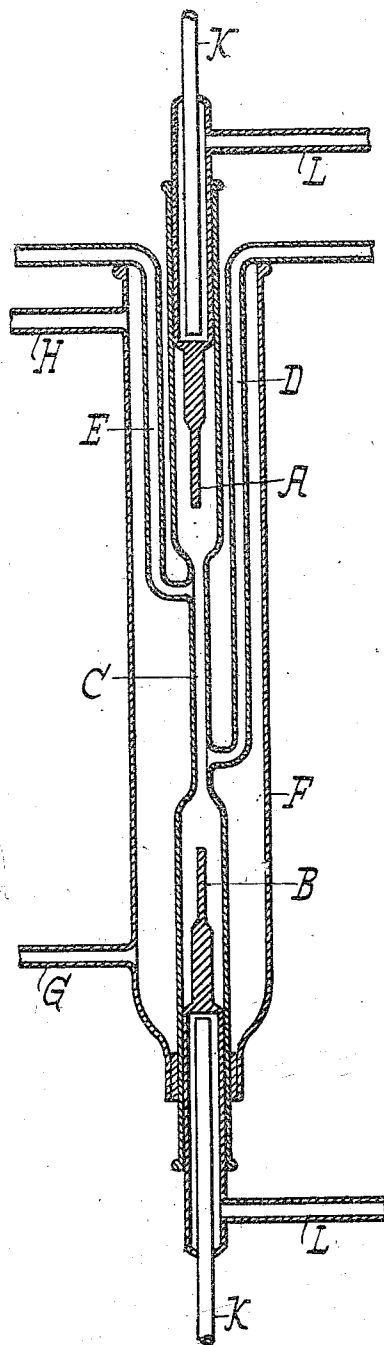


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PROCESS AND APPARATUS FOR THE PRODUCTION OF COMPOUNDS OF OXYGEN AND NITROGEN.
APPLICATION FILED JULY 15, 1908.

938,316.

Patented Oct. 28, 1909.



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UNITED STATES PATENT OFFICE.

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PROCESS AND APPARATUS FOR THE PRODUCTION OF COMPOUNDS OF OXYGEN AND
NITROGEN.

938,316.

Specification of Letters Patent.

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To all whom it may concern:

Be it known that we, FRITZ HABER and ADOLF KOENIG, subjects, respectively, of the King of Prussia and the Emperor of Austria-Hungary, residing at Karlsruhe, Germany, have invented new and useful Improvements in Processes and Apparatus for the Production of Compounds of Oxygen and Nitrogen, of which the following is a specification.

Our invention relates to a process and apparatus for the production of compounds of oxygen and nitrogen from mixtures thereof such for instance as air by means of an electric arc.

In carrying out the process of our invention we subject the gas mixture to the action of an electric arc while at a pressure not exceeding that of half an atmosphere and in an apparatus of such shape that the inside thereof is practically filled with arc flame, while the apparatus is cooled either by the natural escape of heat or by additional means of cooling. By this means the concentration of the nitric oxide in the gases leaving the apparatus is increased and it is easy to produce gases containing from nine and one-half to ten per cent. nitric oxide when using air, and from thirteen and one-half to fourteen per cent. when a mixture of equal quantities of oxygen and nitrogen is employed.

Accounts are given in the literature of experiments which have been made to subject mixtures of nitrogen and oxygen, preferably air, to the action of the electric discharge at pressures other than that of the atmosphere, in particular Lord Rayleigh (*Journal Chem. Soc.*, 1897, vol. 71, p. 181) carried out experiments at pressures both above and below that of the atmosphere. At a total pressure equal to half an atmosphere he obtained somewhat less favorable yields than those obtained at ordinary pressure; he remarks, however, that the results point to the yield being almost independent of the pressure. This conclusion is as a matter of fact correct in so far as mere alteration of the pressure alone does not produce results substantially different from those obtained at ordinary pressure.

The process of our present invention differs from the previously described processes

in that whereas hitherto attempts have been made to maintain as high a temperature as possible in the arc and cool the products of the reaction as quickly as possible, in the process of the present invention the arc is kept comparatively cool by withdrawing heat and an extremely high temperature is avoided. The reduced pressure also assists this cooling since the discharge, which at ordinary pressure possesses the shape of a thin rod, expands and approaches the walls of the apparatus, which being cooled help to remove heat from the arc. In this way discharge flames can be produced which practically no longer possess the well-known brown zone in which the nitric oxide is believed to decompose partially back again to nitrogen and oxygen. The wall of the tube, or the like, containing the arc should be maintained so cool that the arc does not spring across to it and thereby disturb the discharge. If the natural escape of heat is not sufficient for this purpose, special cooling means must be provided and this is advisable even if the tube, or the like, be made of material which at ordinary temperature is a non-conductor of electricity, since at sufficiently high temperatures such material assumes the properties of a conductor.

The arc is preferably formed between metallic electrodes, or electrolyte electrodes, under which term we include, for instance, electrodes of magnesium oxide, calcium oxide and strontium oxide, and is allowed to burn in a long cylindrical vessel of which the walls can, if necessary, be cooled from the outside. The establishment of the discharge can easily be brought about even when the electrodes are at a considerable distance from one another, for instance by greatly reducing the pressure for the time being. When once the arc is created the pressure can be allowed to rise again, whereupon the electrodes, unless specially cooled (which in some cases may be desirable) become red-hot and the passage of the current is very much facilitated. In this way a long arc column, which appears to be quite stationary, is produced, and when the diameter of the tube, or the like, and the pressure of the gas are suitably chosen, the arc practically fills the tube, or the like. Electrolyte electrodes have generally the advantage of producing less dust

than metallic electrodes. The mixtures of nitrogen and oxygen are passed through the cylindrical tube, or the like, which is filled by the flame column and the gases are constantly maintained at a pressure not exceeding that of half an atmosphere. The aforementioned yields of nitric oxid are obtained without it being necessary to cool the gases rapidly by removing them quickly from the sphere of reaction, for instance by withdrawing them through metallic coolers. For instance the aforementioned yields were obtained when the arc was burned inside a tube of quartz which was cooled with water and the air or mixture of oxygen and nitrogen entered and left the tube at a pressure of 90 mm. mercury, and with the low velocity of one-fifth meter per second. In order to prevent decomposition of the nitric oxid at the red-hot electrodes, the entry and exit tubes for the gases can be so placed that the layer of gases around the electrodes is not renewed. The sectional current density, *i. e.* the strength of the current per unit area of the cross-section of the tube should preferably be as high as possible.

It is difficult to define sharply the boundaries within which the pressure can be varied. No advantage, however, is obtained by reducing the pressure considerably under 80 mm. mercury. With regard to higher pressures, it must be remembered that the higher the pressure the greater will be the velocity of decomposition of the nitric oxid. Experiments which were carried out at a pressure of 200 mm. mercury showed, however, that the upper boundary at which the usual small concentrations are obtained had not been reached.

The drawing accompanying this specification will assist in illustrating the nature of our invention, but we do not limit our invention to the apparatus there shown.

The drawing represents in section a suitable apparatus for carrying out the process.

A and B are the two electrodes and the arc which springs from one to the other is inclosed by and practically fills the tube C. Air or other mixture of nitrogen and oxygen is applied to and drawn away from the tube C through the tubes D and E, which are so arranged that the gases surrounding the electrodes A and B are practically not renewed.

F is a wider tube surrounding the tube C and provided with entry and exit openings G and H through which a supply of cooling fluid can be introduced and removed.

K K and L L are tubes leading to and from the bases of the electrodes A and B, and through these tubes a cooling fluid can be passed and assists in preventing the electrodes from becoming too hot.

Now what we claim is:

1. The process of producing compounds

of nitrogen and oxygen from mixtures of nitrogen and oxygen by subjecting the gas mixture to the action of an electric arc while at a pressure not exceeding that of half an atmosphere, and while causing the arc flame to practically fill the vessel containing it substantially as described.

2. The process of producing compounds of nitrogen and oxygen from mixtures of nitrogen and oxygen by subjecting the gas mixture to the action of an electric arc while at a pressure not exceeding that of half an atmosphere, and while causing the arc flame to practically fill the vessel containing it and cooling the vessel substantially as described.

3. The process of producing compounds of nitrogen and oxygen from mixtures of nitrogen and oxygen by subjecting the gas mixture to the action of an electric arc while at a pressure not exceeding that of half an atmosphere and while causing the arc flame to practically fill the vessel containing it, and cooling the external wall of the vessel, and while causing the layer of gases around the electrodes to be practically not renewed, substantially as described.

4. Apparatus for producing compounds of nitrogen and oxygen from mixtures of nitrogen and oxygen comprising a cylindrical vessel in combination with electrodes situated in and parallel to the axis of the said cylindrical vessel, entry and exit openings for the gases and means for maintaining the gases in the vessel under reduced pressure.

5. Apparatus for producing compounds of nitrogen and oxygen from mixtures of nitrogen and oxygen comprising a cylindrical vessel in combination with electrodes situated in and parallel to the axis of the said cylindrical vessel, entry and exit openings for the gases and means for maintaining the gases in the vessel under reduced pressure and means for cooling the external walls of said vessel.

6. Apparatus for producing compounds of nitrogen and oxygen from mixtures of nitrogen and oxygen comprising a cylindrical vessel in combination with electrodes situated in and parallel to the axis of the said cylindrical vessel, entry and exit openings for the gases arranged so that the layer of gases around the electrodes is practically not renewed means for maintaining the gases in the vessel under reduced pressure and means for cooling the external walls of said vessel.

In testimony whereof we have hereunto set our hands in the presence of two subscribing witnesses.

FRITZ HABER.
ADOLF KOENIG.

Witnesses:

A. REUTLINGER,
JOS. H. LEULÉE.