No. 829,874. PATENTED AUG. 28, 1906.

D. R. LOVEJOY.

METHOD OF EFFECTING THE COMBINATION OF GASES.

APPLICATION FILED JUNE 24, 1902. RENEWED JULY 5, 1906.

2 SHEETS—SHEET 1.

Fig. 1.

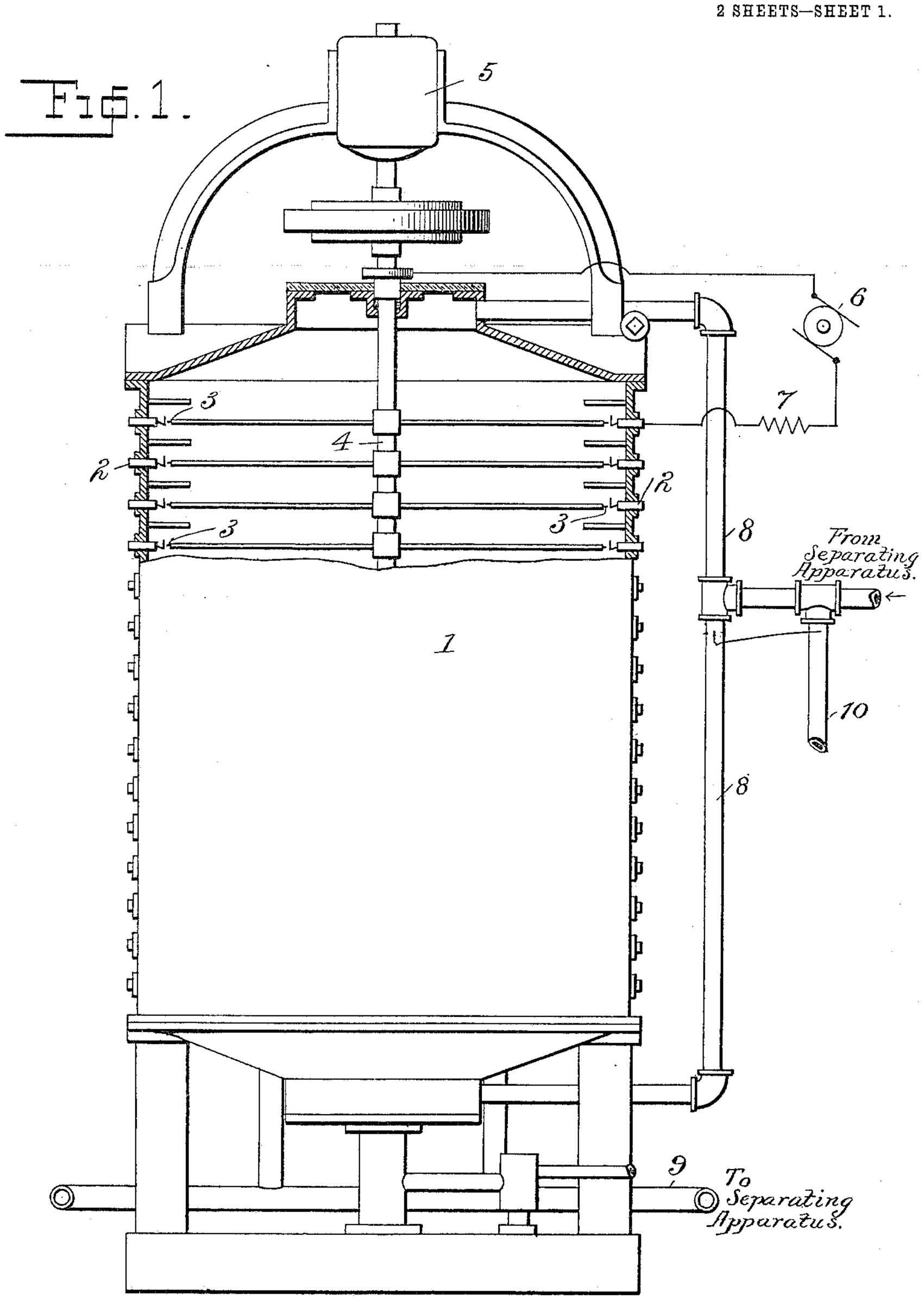

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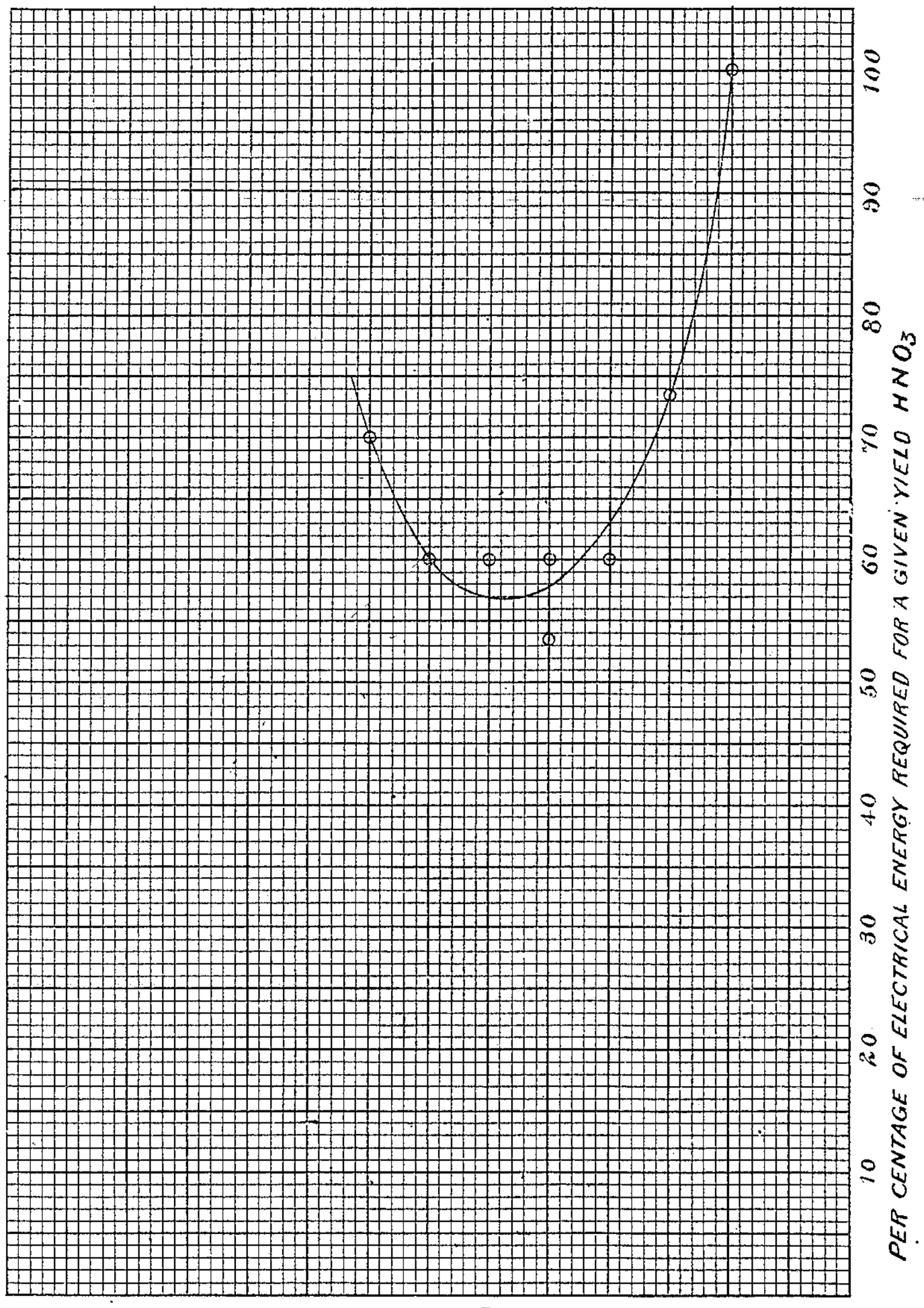
FIG. 2



Witnesses
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UNITED STATES PATENT OFFICE.

DIMMITT ROSS LOVEJOY, OF NIAGARA FALLS, NEW YORK, ASSIGNOR TO ATMOSPHERIC PRODUCTS COMPANY, OF NIAGARA FALLS, NEW YORK, A CORPORATION OF NEW YORK.

METHOD OF EFFECTING THE COMBINATION OF GASES.

No. 829,874. Specification of Letters Patent. Patented Aug. 28, 1906.

Application filed June 24, 1902. Renewed July 5, 1906. Serial No. 324,828.

To all whom it may concern:

Be it known that I, DIMMITT ROSS LOVEJOY, a citizen of the United States, residing at Niagara Falls, in the county of Niagara and State of New York, have invented certain new and useful Improvements in Methods of Effecting the Combination of Gases, of which the following is a specification.

The present invention is an improvement on the process described in United States Letters Patent of Charles S. Bradley and myself, serially numbered 709,868, dated September 30, 1902, in which oxids of nitrogen are formed by direct synthesis from the oxygen and nitrogen of the air by subjecting a current of air to the action of electric arcs. I have found that the yield of nitric oxids is greatly increased with a given expenditure of electrical energy if the atmospheric air is enriched by the addition of pure oxygen and that further efficiency is given by the addition of a small percentage of pure hydrogen to the ordinary mixture as found in atmospheric air or to mixtures in other proportions as formed by the addition of oxygen to air.

My present invention therefore consists, first, in the process of making compounds containing nitrogen by subjecting to the action of the electric arc nitrogen mixed with a larger porportion of oxygen than that found in the atmosphere, and, second, in modifying the process by adding hydrogen to said mixture or to the mixture of nitrogen and oxygen as found in air. I have found it convenient to carry out the process in an apparatus such as described in the application for United States Letters Patent made by me May 29, 1902, Serial No. 109,443, with certain additions adapting it to the present process.

In the accompanying drawings, Figure 1 illustrates in vertical section and partly in diagram the general component parts of such an apparatus; and Fig. 2 shows the curve of variation in the amount of power consumed for a given yield for different gaseous mixtures, ranging from twenty per cent. to ninety per cent. of oxygen.

The process is carried out in an apparatus such as illustrated in Fig. 1, in which 1 is a closed chamber having ranged around its walls a series of contacts 2, into sparking proximity with which a series of movable contacts 3 are carried by a shaft 4, driven by a motor 5. The current for the contacts is supplied by a suitable high-potential generator 6, and its effect is regulated at the several contacts by a series of inductances 7, inserted in the circuit, only one of which is indicated and which are fully described in the said application Serial No. 109,443. The air and the nitric compound which has been formed are led into the chamber by pipes 8 and led from the chamber by the pipes 9 to any suitable apparatus for separating out the nitric compound, and the uncombined air may be returned to the chamber 1 for further treatment.

The details of the construction of the mechanism and the general features of the process are not more fully described herein, reference being made to the said previous applications for same.

To increase the yield of nitric oxids with a given expenditure of electrical energy, I enrich the atmospheric air at or before the time of treating it with the electric arc—as, for example, by leading in at the pipe 9 a supply of pure oxygen. I have found that the maximum beneficial result is obtained when enough oxygen is added to bring the percentage of oxygen in the mixture up to approximately fifty per cent.—that is to say, equal volumes of oxygen and nitrogen. The variation in the amount of power consumed for a given yield for different gaseous mixtures of oxygen and nitrogen, ranging from twenty per cent. to ninety per cent. of oxygen, is shown by the annexed curve, Fig. 2, in which the ordinates represent the percentage of oxygen by volume in the mixture of the gases and the abscissa represent the percentage of electrical energy required for a given yield of HNO_3. From this it will be seen that the minimum amount of power is consumed when the mixture is one of approximately equal volume.

The following data are given as an example illustrating my invention. When the gases are in the proportion as found in air and with an energy of seventy-five watts per arc, a voltage of fifteen thousand volts, and with 3.75 cubic feet of air per arc per hour, a yield is obtained of 5.8 grams HNO_3 per arc per hour. When the air is enriched with oxygen so that the proportion of oxygen pres-

ent is fifty per cent., and with seventy-five watts per arc, a voltage of fifteen thousand volts, and 3.75 cubic feet of the oxygen mixture per arc per hour, a yield is obtained of 7.26 grams HNO_3 per arc per hour, thus showing a large gain in yield when the air is enriched with oxygen. The reactions occurring are the same as occur when the gases are in the proportion as found in air, except that more oxygen and nitrogen are converted. The probable reactions are:

$$N+O+\text{electrical energy}=NO$$
$$NO+O=NO_2$$

also

$$2NO+O=N_2O_3,$$

giving a yield of $NO_2+N_2O_3$ in varying proportions, according to circumstances, temperature, rate of draft, &c.

The efficiency of the process may also be increased by adding a small percentage of pure hydrogen. This may be led in also by the pipe 10 and may be added either to the ordinary mixture as found in atmospheric air or to a mixture substantially as just above described, in which the proportion of oxygen is increased over that found in atmospheric air. The percentage of hydrogen giving the greatest efficiency is less than ten per cent. of the whole volume. The mixture of nitrogen and oxygen or nitrogen, oxygen, and hydrogen is led into the chamber 1 and there subjected to the action of the electric arc and led away for the separation of the nitric compound in the manner described in the aforesaid prior applications.

In the present process where the uncombined gas is led back for re-treatment it will be observed that it is only necessary to add the atmospheric air and oxygen or oxygen and hydrogen in sufficient proportions to supply the loss occasioned by the removal of the combined gases. In such a system the combined and uncombined gases will be drawn or forced from pipes 8 to the separating apparatus, wherein the combined gases are removed and the uncombined gases are then led back through pipes 9 to the chamber 1, only enough oxygen or air, oxygen, and hydrogen being added to it by pipe to supply deficiency caused by the combination of a part of the gases. In this way great saving is effected, for a comparatively small amount of the gases are combined at each passage through the chamber, and if the mixed gas were thrown away after a single treatment there would be a considerable waste of the oxygen and hydrogen which has been especially prepared for addition to the air.

Having thus described my invention, what I claim as new therein, and desire to secure by Letters Patent, is—

1. The process of forming nitrogen compounds, which consists in producing an electric arc, attenuating said arc, and introducing air enriched with oxygen into the presence of said arc, substantially as described.

2. The process of forming nitrogen compounds, which consists in producing an electric arc, elongating said arc, and introducing into the presence of said arc air having the proportion of oxygen present increased above the normal.

3. The process of forming nitrogen compounds, which consists in producing an electric arc, elongating said arc, and introducing into the presence of said arc a mixture of gases containing nitrogen and oxygen in which the proportion of oxygen exceeds atmospheric proportions.

4. The process of effecting the combination of gases to form oxids of nitrogen and their compounds, consisting in mixing nitrogen and oxygen in substantially equal volumes and subjecting the mixture to the action of the electric arc.

5. The process of effecting the combination of gases to form oxids of nitrogen and their compounds, consisting in adding hydrogen to a mixture of oxygen and nitrogen and subjecting the resulting mixture to the action of the electric arc.

6. The process of effecting the combination of gases to form oxids of nitrogen and their compounds, consisting in adding hydrogen to a mixture of oxygen and nitrogen in such proportion as to form a mixture containing less than ten per cent. of hydrogen and subjecting this mixture to the action of the electric arc.

7. The herein-described process of forming nitrogen compounds, which consists in mixing oxygen with air until the total amount of oxygen in the mixture is substantially equal to that of the nitrogen and subjecting said mixture to the action of the electric arc.

8. The herein-described process of forming nitrogen compounds, which consists in adding to a given quantity of air a proportion of oxygen sufficient to bring the total quantity of oxygen in the mixture to an amount substantially equal to that of the nitrogen present, subjecting said mixture to the action of the electric arc, absorbing the nitrogen compounds formed, adding a proportion of oxygen to the uncombined gases sufficient to restore the relative proportions of oxygen and nitrogen to substantially equal amounts and resubjecting said uncombined gases and oxygen to the action of the electric arc.

9. The process of forming nitrogen compounds, which consists in simultaneously producing a plurality of electric arcs, simultaneously elongating said arcs, and introducing into the presence of said arcs a mixture of gases containing nitrogen and oxygen, in which the proportion of oxygen is substantially equal to that of the nitrogen.

10. The process of forming nitrogen com-

pounds, which consists in adding hydrogen to a gaseous mixture containing oxygen and nitrogen, introducing said mixture into the presence of electric arcs while elongating said arcs.

11. The process of forming nitrogen compounds, which consists in adding hydrogen to a gaseous mixture containing oxygen and nitrogen in substantially equal volumes, introducing said mixture into the presence of electric arcs while elongating said arcs.

12. The process of forming nitrogen compounds, which consists in adding hydrogen to a gaseous mixture containing oxygen and nitrogen until the proportion of hydrogen is less than ten per cent. of the whole volume, introducing said mixture into the presence of electric arcs while elongating said arcs.

13. The process of forming nitrogen compounds, which consists in successively and rapidly forming, elongating and interrupting an electric arc, and introducing into said arc a mixture of oxygen and nitrogen in which the oxygen is present in greater than atmospheric proportion.

14. The process of forming nitrogen compounds, which consists in successively and rapidly forming, elongating and interrupting an electric arc, and introducing into said arc a mixture of hydrogen, oxygen and nitrogen in which the hydrogen and oxygen are present in greater than atmospheric proportion.

DIMMITT ROSS LOVEJOY.

Witnesses:

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