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METHOD AND APPARATUS FOR SYNTHESIZING NITROGEN COMPOUNDS

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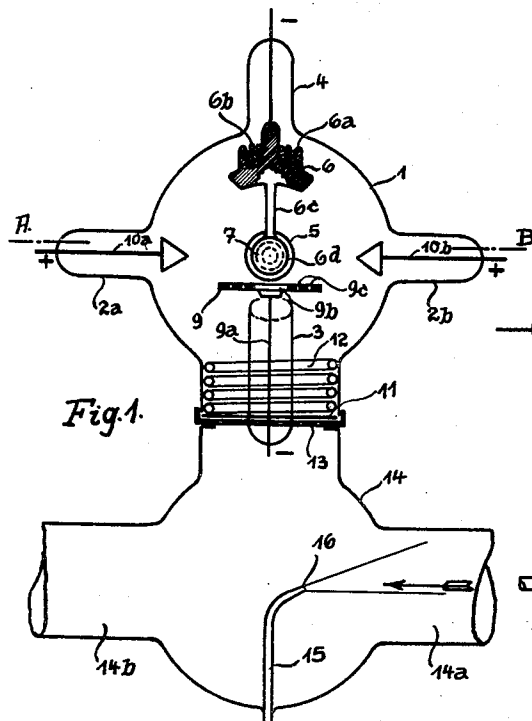


Fig. 1.

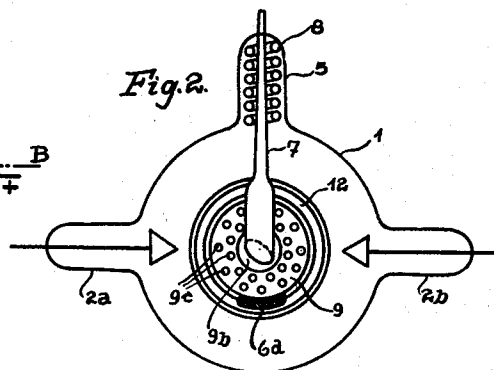


Fig. 2.

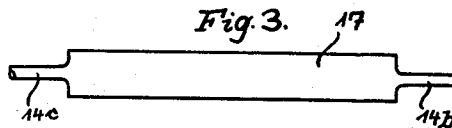


Fig. 3.

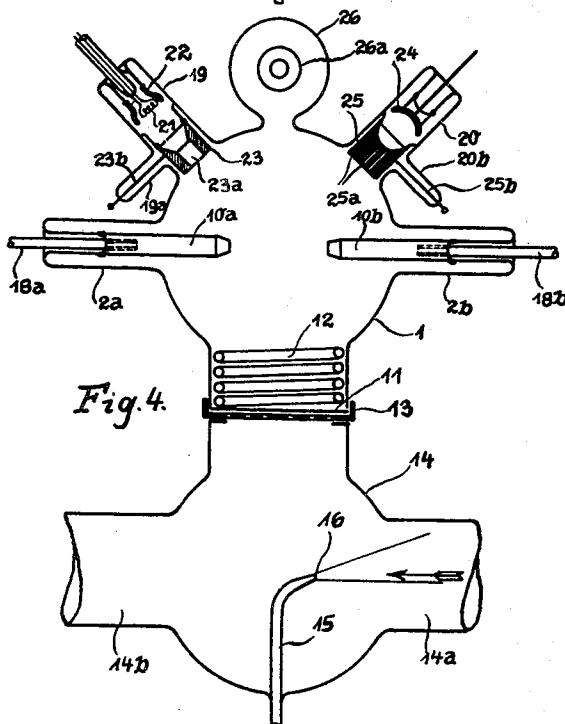


Fig. 4.

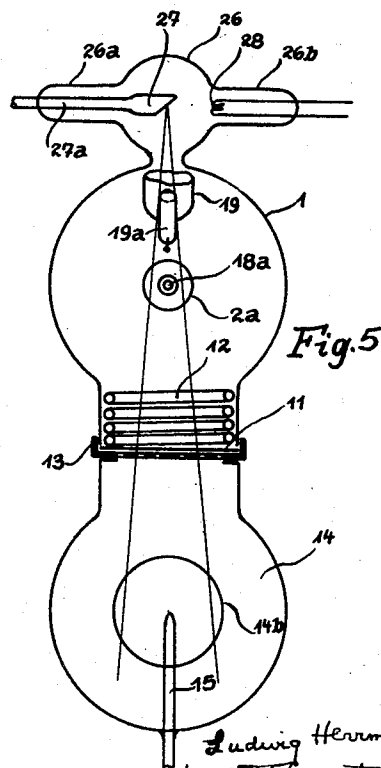


Fig. 5.

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UNITED STATES PATENT OFFICE

2,064,260

METHOD AND APPARATUS FOR SYNTHESIZ-
ING NITROGEN COMPOUNDS

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23 Claims. (Cl. 204—31)

My invention has reference to the carrying out of chemical processes especially of the production of certain nitrogen compounds and means serving therefor.

5 In making my experiments I started from the so-called Lenard-tube. A Lenard-tube is known to be a vacuum-tube which permits the exit of the electrons or cathode-rays produced therein. For this purpose the vacuum-tube or vessel is
10 provided with a window consisting of a thin metal-foil. Such tubes or vessels had been already used for influencing chemical reactions by the electrons or cathode-rays emitted therefrom.

In such a vacuum-tube X-rays also are pro-
15 duced in larger or smaller quantities.

My invention is based upon the observation that new and unexpected effects are produced if the tube is so constructed that positive rays also strike against its window. Positive rays are
20 understood to be both the canal-rays or diacathode-rays discovered by Goldstein and the anode-rays discovered by Gehrcke and Reichenheim. I have ascertained the unexpected fact that the effects I had observed are not produced
25 if two vacuum-tubes are joined to a common reaction chamber, one of which tubes emits only cathode-rays through its window into the reaction chamber whereas in the other tube only canal-rays are directed against the window sepa-
30 rating the last mentioned tube from the reaction chamber. It is on the contrary necessary for obtaining the effects characterizing my invention to produce in the same vacuum-tube or bulb both the cathode-rays and the positive rays
35 and to direct them both against a window provided at the tube.

In the accompanying drawing some embodiments of apparatus constructed according to my invention are illustrated which I used in carry-
40 ing out chemical syntheses.

Figure 1 is a plan view of one embodiment partly in section.

Figure 2 is a section on line A—B of Fig. 1.

Figure 3 shows schematically a reaction-
45 chamber for the production of ammonia sulphate.

Figure 4 shows a further embodiment of an apparatus for the production of positive and negative rays, partly in section.

50 Figure 5 shows a side elevation of the apparatus according to Fig. 4 partly in section.

As illustrated in Figures 1 and 2 the tube 1 is provided with five tubular extensions 2a, 2b, 3, 4 and 5 for introducing electrodes. In the
55 upper extension 4 the leading-in conductor for

the cathode 6 is arranged. The cathode 6 has a pocket 6a in which a heating-coil for heating the cathode is inserted. A circular cathode-disc 6d is fixed by an arm 6c to the cathode-body 6. This disc forms the cathode for the anode 7 or target serving for the production of X-rays and shaped in such a manner as the anticathodes or targets of X-ray tubes for medical purposes. The bushing of the anode 7 passes the tubular extension 5. The anode 7 is cooled by the cooling-coil 8. In the oblique tubular extension 3 is arranged the leading-in conductor 9a of a second cathode 9 which is a pierced-cathode. This has a large aperture 9b in its center and small openings 9c towards its periphery. The large aperture 9b serves for letting pass the electrons or cathode-rays emitted by the cathode 6. These electrons are accelerated by the two auxiliary electrodes 10a, 10b having a positive potential. The leading-in conductors for these two auxiliary electrodes are arranged in the tubular extensions 2a, 2b. Canal-rays pass through the small openings of the pierced electrode 9. The bulb 1 is provided with a window 11 which is formed by a thin metal-foil for instance of an aluminium-alloy. Because of the rays passing through the window 11 excessive heating of the window would tend to occur. For preventing this it is cooled by a cooling-device with liquid-circuit. The cooling effect of the coil 12 is sufficient for preventing the temperature of the window 11 from rising to an unpermissible degree. This window is covered by a pierced metal cap 13 cemented to a circular flange of the reaction-chamber 14. This chamber is provided with an inlet pipe 14a and an outlet pipe 14b. A nozzle 15 is introduced into the reaction-chamber 14 with its mouth 16 opened against the direction of the gas-current and serving for the injection of
40 liquids.

The tube 1 being in operation and air being conducted to the reaction-chamber through the inlet pipe 14a a highly explosive gas is produced in the reaction-chamber which leaves this chamber by the outlet pipe 14b. Preferably water or hydrogen peroxide is injected through the nozzle 16.

I have not yet analyzed exactly the chemical combination of the gas produced by these experiments. I was able, however, to determine with certainty that this combination is a compound or a mixture of compounds of nitrogen. This gas has been found to be an excellent fuel for inter-
55

nal combustion engines. Before feeding the combustion chamber of the engine with the gas it should be admixed with air.

If the gas produced in the reaction-chamber 14 by irradiating air with positive and negative rays is led according to the Figure 3 into a second reaction-chamber 17 provided with a leading-out pipe 14c and containing ferrous sulphide (FeS) and water acidified with sulphuric acid, ammonia sulphate is produced.

For producing anode-rays in addition to canal-rays the anticathode or target 7 is provided with a recess filled with a salt causing the generation of anode-rays. Such salts are known to be salts such as sodium-, potassium- or strontium-salts.

I cannot give an explanation of the kind of chemical influence the positive rays exert, but I presume that the cathode-rays preactivate the gases flowing through the reaction chamber 14 and that the positive rays act as a catalyst. I do not desire to limit the invention by this theory. It may be imagined that the rays themselves exercise no direct influence upon the chemical reaction but produce accompanying secondary phenomena.

Moreover it is not at all necessary to provide in the tube 1 a special anticathode 7 for producing X-rays. In each vacuum-tube X-rays are produced to larger or smaller extent by the cathode-rays of the tube. These X-rays are sufficient for the carrying-out of the syntheses according to my invention. In view of this I suppose that X-rays are of less importance as to the scope of my invention. On the other hand, the addition of positive rays to the cathode-rays is an essential feature of my invention whether the positive rays be merely canal- or diacathode-rays or such rays and anode-rays. In making my experiments I have evacuated the bulb or vessel 1 to an extremely high degree, e. g. to 10^{-5} mm. Hg, and for this purpose I leave the pump permanently joined to the bulb.

The bulb 1 shown in Figures 4 and 5 has two lateral tubular extensions 2a, 2b provided with re-entrant portions, to which the auxiliary electrodes 10a, 10b are hermetically sealed. Metal bolts 18a, 18b serving as conductors are screwed to the electrodes 10a, 10b. The upper part of the glass bulb 1 has two oblique tubular extensions 19 and 20. An incandescent cathode 21 surrounded by a metal cap 22 is arranged within the upper part of the extension 19. An anode 23 with an opening 23a for the passage of the electrons or cathode-rays emitted from the incandescent cathode 21 is sealed to a circular re-entrant portion of the under part of the tubular extension 19. These electrons are deflected by means of the auxiliary electrodes 10a, 10b in such a manner that they strike the window 11. The extension 19 carries a small tubular extension 19a containing the leading-in conductor 23b for the anode 23. An anode 24 is sealed to the re-entrant portion of the tubular extension 20, the lower part of this extension carrying the pierced cathode 25. The openings 25a in this cathode are of small diameter, but they are many in number. The cathode 25 is put under tension by means of a leading-in conductor 25b, traversing the small tubular extension 20b of the inclined extension 20.

A third extension 26 is arranged between the extensions 19 and 20 having a globular shape and two tubular ends 26a, 26b. The extension 26 is

constructed as an X-ray-tube common in the art. In the tubular end 26a the leading-in conductor 27a for the anticathode or target 27 is placed and the filament of the incandescent cathode 28 is arranged near the shoulder of the tubular end 26b. The X-rays produced on the surface of the target 27 are directed vertically downwards on the window 11, consisting of a thin foil of an aluminum-alloy. In addition to the electrons or cathode rays emitted from the cathode 21 and accelerated by the field of the auxiliary electrodes 10a, 10b also the canal or diacathode rays leaving the openings 25a of the pierced cathode 25 are directed against this window. The cooling coil cools the window 11. A similar cooling device may be provided for the target 27. A metal cap 13 is arranged below the window 11, said cap having a large number of small openings. The reaction chamber 14 is shaped in the same manner as the reaction chamber according to the embodiment of the invention shown in Figures 1 and 2. It has a leading-in pipe 14a and a leading-out pipe 14b. A nozzle 15 traverses the wall of the reaction chamber 14 the opening of which is directed against the leading-in pipe 14a, i. e. against the direction of the gas current.

Experiments carried out with canal-rays and anode rays only showed no change of irradiated air or other irradiated substances within the limits of 10^{-5} to 2×10^{-6} mm. Hg and anode tensions between the limits of 1.5×10^5 to 3.5×10^5 volts. In all experiments I chose the tensions for the heating of filaments of the cathodes to be about 4 to 6 volts.

In other experiments made under the same conditions I investigated the influence of X-rays on atmospheric air and other substances but I could not ascertain any chemical change of the substances irradiated only by X-rays. They had been only ionized by the X-rays.

Further experiments I carried out with cathode rays only, but with these experiments the occurrence of X-rays naturally could not be prevented. They showed a yellow flash up of the irradiated air i. e. its nitrogen, continuing about thirty seconds after irradiation and then disappearing. This phenomenon could be ascertained with a vacuum of 8.3×10^{-6} mm. Hg and a tension of 1.5×10^5 volts.

After canal rays and/or anode rays were added to the cathode rays and X-rays the flashing up disappeared and the substances irradiated produced a chemical compound, the constitution of which I have not yet been able to determine but which is characterized by a great explosive force.

Although I have referred to the electrodes in the apparatus herein-before described as anode and cathode I wish to state that this has been done only for the sake of simplicity. In fact some of the electrodes are entitled to have a multiple function. For avoiding any misunderstanding I have inserted in the following schedules the voltages that might be by way of example applied to the electrodes:

Embodiment according to Figs. 1 and 2

Electrode	Tension
6	0 volt
7	200 kv.
9	0 kv.
10a	200 kv.
10b	200 kv.

Embodiment according to Figs. 4 and 5

Electrode	Tension
21, 22	0 volt
23	150 volts
24	0 volt
25	200 kv.
10a	200 kv.
10b	200 kv.
27	200 kv.
28	0 volt

Having now particularly described and ascertained the nature of my said invention, and in what manner the same is to be performed, I declare that what I claim is:

1. In an apparatus for carrying out chemical reactions, in combination, a reaction chamber, means for driving gas through this chamber, means for producing X-rays, means for producing cathode rays, means for producing positive rays, all said ray producing means being included in a common evacuated bulb, and means for subjecting the said gas to the action of all of said rays.
2. In an apparatus for carrying out chemical reactions, in combination, a reaction chamber, means for driving gas through this chamber, means for producing X-rays, means for producing cathode rays, means for producing canal rays, all said ray producing means being included in a common evacuated bulb, and means for subjecting the said gas to the action of all of said rays.
3. In an apparatus for carrying out chemical reactions, in combination, a reaction chamber, means for driving gas through this chamber, means for producing X-rays, means for producing cathode rays, means for producing canal rays, means for producing anode rays, all said ray producing means being included in a common evacuated bulb, and means for subjecting the said gas to the action of all of said rays.
4. In an apparatus for carrying out chemical reactions, in combination, a reaction chamber, means for driving gas through this chamber, an evacuated bulb, a window of sheet metal between the bulb and the reaction chamber, electrodes enclosed in said bulb and adapted to produce X-rays, cathode rays and positive rays, all the said rays being directed to the metal window.
5. In an apparatus for carrying out chemical reactions, in combination, a reaction chamber, means for driving gas through this chamber, an evacuated bulb, a window consisting of a thin metal foil separating the bulb from the reaction chamber, and electrodes in said bulb including a plurality of anodes and cathodes adapted to emit X-rays, cathode rays and positive rays to said window.
6. In an apparatus for carrying out chemical reactions, in combination, a reaction chamber, means for driving gas through this chamber, an evacuated bulb, a window consisting of a thin metal foil separating this bulb from the reaction chamber, cathodes and anodes in the said bulb adapted to direct a beam of X-rays, cathode rays and positive rays to the said window, one of said cathodes being pierced for the passage of canal rays, and auxiliary electrodes in the said bulb adapted to accelerate the electrons striking the said window.
7. In an apparatus for carrying out chemical reactions, in combination, a reaction chamber, means for driving gas through this chamber, an evacuated bulb, a window consisting of a thin

metal foil separating this bulb from the reaction chamber, electrodes within this bulb, adapted to direct a beam of cathode rays to the said window, auxiliary electrodes adapted to accelerate the said electrons and a pierced cathode, the apertures of which, opposite the electrode in said bulb which acts as the cooperating anode, are facing the said window.

8. In an apparatus for carrying out chemical reactions, in combination, a reaction chamber, means for driving gas through this chamber, an evacuated bulb, a window consisting of a thin metal foil separating this bulb from the reaction chamber, means for cooling this window and electrodes within this bulb adapted to emit X-rays, cathode rays and positive rays to said window.

9. In an apparatus for carrying out the chemical reactions, in combination, a reaction chamber, means for driving gas through this chamber, an evacuated vessel, a window consisting of a thin metal foil separating this vessel from the reaction chamber, means for cooling this window and electrodes within this vessel adapted to emit electrons, canal rays and anode rays to the said window.

10. A method of synthesizing nitrogen compounds which consists in influencing nitrogen of atmospheric air in presence of water by X-rays, cathode rays and positive rays produced in a common evacuated vessel.

11. A method of synthesizing nitrogen compounds which consists in influencing nitrogen of atmospheric air in presence of water by X-rays, cathode rays, canal rays and anode rays produced in a common evacuated bulb.

12. As an explosive gas the product obtained by influencing the nitrogen of atmospheric air in presence of water by X-rays, cathode rays and positive rays produced in the same evacuated bulb.

13. In a fuel generator for internal combustion machines, in combination, a reaction chamber, means for driving atmospheric air through this chamber, means for producing X-rays, means for producing cathode rays, means for producing positive rays, all said means being enclosed in the same evacuated bulb, said bulb being separated from the reaction chamber by a window consisting of a metal foil.

14. Apparatus for carrying out chemical reactions consisting of an evacuated vessel, electrodes therein for the production of cathode rays and canal rays so arranged that the rays cooperate to produce a radiation which is able to penetrate parts opaque to canal rays, and means outside the vessel for directing a stream of gas past the portion of the tube at which the radiation produced by cooperation of the cathode and canal rays emerges from the vessel.

15. Apparatus for the production of strongly active rays consisting of an evacuated vessel, anode and cathode arranged therein for the production of an electron current, an anode which cooperates within said vessels with a perforated cathode so that behind the perforated cathode a current of canal rays is produced, all the electrodes being so arranged that through the cooperation of the electron current and the current of canal rays a radiation is produced which is able to emerge from the vessel through parts that are opaque to canal rays.

16. Apparatus for carrying out chemical reactions consisting of an evacuated vessel, electrodes therein for the production of cathode rays

and canal rays, one of said electrodes being an incandescent cathode and another of said electrodes being a pierced cathode, a reaction chamber associated with said evacuated vessel and means for driving gases through said chamber so that said gases are subjected to the action of said rays.

17. Apparatus for carrying out chemical reactions consisting of an evacuated vessel, electrodes therein for the production of negative and positive rays, one of said electrodes being an incandescent cathode and another of said electrodes being an anode containing a substance adapted to produce anode rays, a reaction chamber associated with said evacuated vessel and means for driving gases through said chamber so that said gases are subjected to the action of said rays.

18. The method of preparing an explosive gas comprising the activation of the nitrogen of atmospheric air in presence of water by X-rays, cathode rays and positive rays produced in the same evacuated vessel and mixing the irradiated gas with atmospheric air.

19. A method of synthesizing nitrogen compounds which consists in influencing nitrogen and another substance capable of reacting therewith by X-rays, cathode rays and positive rays produced in a common bulb, said substance being one of a group consisting of H_2O and H_2O_2 .

20. A method of synthesizing nitrogen com-

pounds which consists in influencing nitrogen of atmospheric air and another substance capable of reacting therewith by X-rays, cathode rays and positive rays produced in a common bulb, said substance being one of a group consisting of H_2O and H_2O_2 .

21. A method of synthesizing ammonia sulphate which consists in influencing nitrogen in presence of another substance by X-rays, cathode rays and positive rays produced in the same evacuated container, said other substance being one of a group consisting of water and hydrogen peroxide, and bringing the irradiated gas into contact with ferrous sulphide and water acidified with sulphuric acid.

22. A method of synthesizing ammonia sulphate which consists in influencing nitrogen of atmospheric air in presence of another substance by X-rays, cathode rays and positive rays produced in the same evacuated container, said other substance being one of a group consisting of water and hydrogen peroxide and bringing the irradiated gas into contact with ferrous sulphide and water acidified with sulphuric acid.

23. As an explosive gas the product obtained by influencing the nitrogen of atmospheric air in presence of another substance selected from a group of compounds consisting of H_2O and H_2O_2 by X-rays, cathode rays and positive rays produced in a common bulb.

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