[Second Edition.]

PATENT SPECIFICATION

Application Date : March 5, 1925. No. 6046/25. 252,786

Complete Left : Nov. 3, 1925.

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PROVISIONAL SPECIFICATION.

Production of Light Oils suitable for use as Fuel for Internal Combustion Engines from Water Gas or similar Gases.

I, MARKUS BRUTZKUS, Engineer, of rue St. Didier 25, Paris (France), a citizen of the Russian Republic, do hereby declare the nature of this invention to be as follows :—

It is known that starting with water gas at a high pressure and at temperatures of 400 to 450° C. with the use of suitable catalysts, it is possible to effect the synthesis of light oils adapted to be burnt as fuel in automobile and aeroplane engines. But in these known processes there are obtained such insignificant quantities of oil that these synthetic processes of producing light oils have not yet been carried out on a commercial scale.

The aqueous and oily liquids obtained by this synthesis contain a large amount of methyl alcohol and higher alcohols, aldehydes, ketones and acids but all these bodies have not more than 8 atoms of carbon. Non-oxygenated hydrocarbons have not been found in the products of this synthesis. It is admitted that this synthesis of these organic bodies commences by the formation of formate according to the following theoretical formulæ :

1. $CO+H_2=COH.H$
2. $2COH.H=CH_3HO+CO$
3. $CH_3.HO+CO=CH_3COOH$
4. $CH_3COOH+2H_2=C_2H_5.HO+H_2O$.

The combination of ethyl alcohol with carbonic oxide give the propylic acid etc. ; it is admitted that the aldehydes and the ketones which are found amongst the products are formed by the hydrogenation of the corresponding acids. Moreover the more complex alcohols may be formed of more simple alcohols particularly in the presence of strong alkalies. As this synthesis is effected with the assistance of a catalyst (usually this catalyst is formed by chips of iron impregnated with KHO) which is very sensible to combinations of sulphur it is necessary for the water gas to be well purified from any compound of sulphur.

The present invention has for its subject a synthetic process of producing light oils suitable for burning in automobile engines from water gas or other similar gases. This process is based on a new general chemico-technological principle and in the use for this purpose of an apparatus in the form of a compressor, this compressor apparatus being preferably in the form of a Diesel engine.

This chemico-technological principle has been established and demonstrated experimentally by the applicant on reactions of combustion in internal combustion engines. This principle consists therein :

" Any chemical reaction can be directed and accelerated in the desired direction by continuous and simultaneous external variations in pressure, temperature and concentration, these variations being produced from outside in a direction opposite to that of the variations produced by the desired reaction."

The production of chemical reactions according to this principle is possible by using for this purpose a compressor preferably in the form of a Diesel engine.

The reactions which take place in this synthesis are explained above :—

As is shown in the above diagram of the formation of organic bodies from water gas, all the reactions which take place have the following characteristic features.

[*Price* 1/-]

1. All these reactions, except reaction 2, are accompanied by a great reduction in the number of molecules. In accordance with the above chemico-technological principle, these reactions should be effected under the action of a continually increasing pressure.

2. As shown by the calculation of the heat of formation all the above reactions (except the first) are exothermic reactions. They should be effected simultaneously with energetic cooling.

3. These synthetic reactions are dependent on an absorption of the gases CO and H and consequently their partial pressures continually decrease. According to the above principle these reactions should be effected under the influence of a continuously increasing concentration of this gas.

It follows that the reaction of the synthesis of these light oils from water gas should be effected at a continually increasing pressure whilst continually cooling and with a continuous increase of the concentration of the gases H and CO.

The variations in the direction necessary for the three factors of chemical equilibrium during the reaction may be effected in the apparatus shown by way of example in Fig. 1 of the accompanying drawing.

The compressor 1 of this apparatus is constructed entirely as a Diesel engine. This apparatus only differs from a Diesel engine by the pulley 5 which is mounted on the main shaft of the compressor. The compressor can be driven by this pulley. This compressor like a Diesel engine, is provided with an auxiliary pump. This pump compresses the water gas to a pressure much higher than the pressure of the gas in the cylinder.

The auxiliary pump of the Diesel engine for the fuel may be omitted in this case but when working at a high temperature it is possible to use it for the injection of water into the cylinder for increasing the cooling of the gas.

These two pumps are entirely constructed like the corresponding pumps in a Diesel engine. They are not shown in Fig. 1.

The compressor 1, like a Diesel engine, is provided with four valves as follows :

1. The inlet valve 2 through which the gas from the tank 6 enters the cylinder.

2. The spraying valve 4 through which a jet of gas, at a pressure higher than the pressure of the gas in the cylinder enters well atomised into the cylinder for increasing the concentration of the gases in the compressor and for thoroughly cooling them. This gas is compressed by the auxiliary pump.

3. The exhaust valve 3 through which the products obtained escape into the tank 8.

4. The passage valve which corresponds with the starting valve in the Diesel engine. This valve is not shown in the annexed drawing.

By means of this valve, at the commencement of operation, the compressed gases can be passed directly into the tank 6 for raising the temperature therein. It is needed only if the communication between the reservoirs 6 & 8 is to be cut off.

All these valves are constructed like the valves of a Diesel engine and are actuated by distribution shafts so as to operate on the two or four stroke cycle.

The products obtained pass through the pipe 12 into the tank 8 where they are cooled and liquefied. The liquefied products leave the apparatus by the pipe 9 and the non-absorbed gases and the permanent gases formed by the process again enter by the tank 6 by the pipes 11 for the purpose of being again treated.

During the working of the compressor it is possible to regulate the temperature in the tank 6 by the flaps 13 and 14, allowing a portion of the gas to enter the tank directly without cooling. By means of the flap 17 it is possible to interrupt the communication between the tank 6 and the tank 8. It will be understood that the tank 6 should be filled continuously with gas in proportion as the gases are absorbed during the process. Care should also be taken that the gas in the tank is of constant composition. Sometimes it is more practicable to interrupt the communication of the tank 8 with the tank 6 directly. Then the gas not absorbed can be conducted from the tank 8 to the tank 6 by a special compressor which sucks the gases from the tank 8 and causes them to escape into the tank 6.

In Fig. 2 is shown a modification of the compressor 1. This modification differs from the compressor in Fig. 1 by a special reaction chamber, of which the walls may be cooled directly from the outside. If the operation is carried out with a catalyst this may be placed into this special chamber.

Fig. 3 shows a further modification of the compressor 1 particularly constructed for operation with a catalyst. In this modification the reaction chamber is terminated by the pipe 16 where the catalysts may be placed between metal films. The exhaust valve is placed in this compressor behind the pipe 16. The

gases are thus caused to pass through the layer of catalyst. In Figs. 2 and 3 the valves are indicated by the same references as the corresponding valves of the compressor in Fig. 1.

In the apparatus above described the synthesis of the organic body may be effected in two strokes in the following manner:

1. First stroke. The piston 7 moves towards the outside and the water gas which is in the tank 6 fills the cylinder 1 of the compressor. The gas has for example a temperature of 62° C. and a pressure of 4 atmos.

2. Second stroke. The piston moves towards the interior and compresses the gas enclosed for example during $\frac{3}{4}$ of its return stroke. At this moment the valve 4 opens and a jet of gas, quite cool, of a pressure of 160 atmos. enters the cylinder. This jet of gas in view of its low temperature and particularly by its expansion in the cylinder thoroughly cools the enclosed gas. At the same time this jet of water gas increases the concentration of the water gas in the cylinder. This gas at high pressure is supplied by the auxiliary pump of the compressor.

Without cooling and without the admission of the jet of gas the final temperature and pressure will be 700° C. and 140 atmos. calculated by the formulæ of adiabatic pressure, when the compression chamber is 8% of the cylinder.

When the piston approaches the dead point the valve 3 opens and the mixture of gases and vapours escapes into the tank 8 where this mixture is subjected to cooling by a jet of water or other liquid. The liquefied vapours leave the apparatus by the pipe 9. The gases which have not been absorbed pass into the tank 6 so as to be again treated.

In this manner is effected the synthetic process of producing light oils from water gas supplied continuously and simultaneously under the action of an increasing pressure, cooling and increase in the concentration of the hydrogen and carbon oxide in accordance with the above chemico-technological principle.

As a result of the operation above described there are obtained organic bodies indicated in the preamble to the present description. As the temperature in the preceding example was chosen sufficiently high, the products are more hydrogenated and less oxygenated bodies are obtained.

The final temperature and pressure fixed above are not the only ones under which this synthesis can be effected. This synthesis can be effected equally well at lower temperatures and at higher temperatures. The more the temperature of the operation is increased the higher should be the pressure chosen and on the contrary the lower the temperature chosen for the operation the lower will also be the pressure chosen for this operation. The higher the temperatures of the operation, more of the products obtained will contain hydrogenated products derived from the oxygenated hydrocarbons. The lower the temperature of the operation, the more products obtained will contain oxygenated products.

As all these synthetic reactions are exothermic reactions, the gases and vapours enclosed in the cylinder will contain a large quantity of kinetic energy which may be converted into mechanical energy. For this purpose the process above described may be effected not in two but in four strokes. The gases and the vapours obtained by the compressor at the end of the second stroke remain in the cylinder and the operation is continued in the following manner.

Third stroke. The piston moves towards the exterior and the gases and vapours obtained expand and convert their kinetic energy into mechanical work. By reason of the expansion and under the action of the cold walls the gases and the vapours are considerably cooled and there is thus avoided a recurrence of the reaction. If this cooling is not sufficient, it may be increased by the introduction into the cylinder of a jet of cooled gas or even a jet of water.

Fourth stroke. The piston moves towards the interior of the cylinder and the gases and the vapours already well cooled and expanded escape from the cylinder by the open exhaust valve 3 and enters the tank 8.

Instead of obtaining mechanical energy in the compressor itself the gas may be allowed to expand in a special cylinder interposed between the compressor and the tank 8. In this manner it is possible to obtain a quantity of mechanical energy sufficient to conduct the whole synthetic process with comparatively small or no expense of external mechanical energy.

In allowing the pressure, the temperature and the concentration to vary during the reaction in the opposite directions to those in which they vary by reason of the reaction the chemical system is continually extended from its state of equilibrium and an impulsion is given to the reaction in the desired direction.

This impulsion may replace the

impulsion given to the reaction by the catalyst.

In a certain temperature zone this impulsion will be sufficiently great for the reaction to take place in the desired direction without the use of a catalyst.

The carrying out of the reaction without a catalyst in accordance with the invention is preferable to other synthetic processes using a catalyst. In working without a catalyst there is no necessity of purifying the gases, and complications in operation are avoided which are always inherent to the use of a catalyst.

But if from a certain point of view it is desired to use a catalyst, it can be done in the same manner as above described in the compressor of Figs. 2 and 3.

The process above described may be used for the synthesis of light oils not only from water gas, but also from gases from coke furnaces, power from gas, of anthracite, of coke, and of lignite, illuminating gas, and blast furnace gases by adding the necessary quantity of hydrogen.

Dated the 26th day of February, 1925.

MARKUS BRUTZKUS.

COMPLETE SPECIFICATION.

Production of Light Oils suitable for use as Fuel for Internal Combustion Engines from Water Gas or similar Gases.

I, MARKUS BRUTZKUS, Engineer, a citizen of the Russian Republic, 25, rue St. Didier, Paris (16), France, do hereby declare the nature of this invention and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:—

It is known that starting with water gas at a high pressure and at temperatures of 400 to 450° C. with the use of suitable catalysts, it is possible to effect the synthesis of light oils adapted to be burnt as fuel in automobile and aeroplane engines. But in these known processes there are obtained such insignificant quantities of oil that these synthetic processes of producing light oils have not yet been carried out on a commercial scale.

The aqueous and oily liquids obtained by this synthesis contain a large amount of methyl alcohol and higher alcohols, aldehydes, ketones and acids but all these bodies have not more than 8 atoms of carbon. Non-oxygenated hydrocarbons have not been found in the products of this synthesis. It is admitted that this synthesis of these organic bodies commences by the formation of formaldehyde according to the following theoretical equations:—

1. $CO+H_2=COH.H$
2. $2COH.H=CH_3HO+CO$
3. $CH_3.HO+CO=CH_3COOH$
4. $CH_3COOH+2H_2=C_2H_5.HO+H_2O$.

The combination of ethyl alcohol with carbonic oxide give the propylic acid etc.; it is admitted that the aldehydes and the ketones which are found amongst the products are formed by the hydrogenation of the corresponding acids.

Moreover the more complex alcohols may be formed of more simple alcohols particularly in the presence of strong alkalies. As this synthesis is effected with the assistance of a catalyst (usually this catalyst is formed by chips of iron impregnated with KHO) which is very sensitive to the combinations of sulphur it is necessary for the water gas to be well purified from any compound of sulphur.

The present invention has for its subject a synthetic process of producing light oils suitable for burning in automobile engines from water gas or other similar gases. This process is based on a new general chemico-technological principle which was described in my prior Specifications Nos. 155,776, 217,747 and in the use for this purpose of an apparatus in the form of a compressor, this compressor apparatus being preferably in the form of a Diesel engine.

This chemico-technological principle has been established and demonstrated experimentally by the applicant on reactions of combustion in internal combustion engines. This principle consists therein:

"Any chemical reaction can be directed and accelerated in the desired direction by continuous and simultaneous external variations in pressure, temperature and concentration, these variations being produced from outside in a direction opposite to that of the variations produced by the desired reaction."

The production of chemical reactions according to this principle is possible by using for this purpose a compressor pre-

ferably in the form of a Diesel engine. This invention is a new application of this principle.

The reactions which take place in this synthesis are explained above:—

As is shown in the above diagram of the formation of organic bodies from water gas, all the reactions which take place have the following characteristic features.

1. All these reactions, except reaction 2, are accompanied by a great reduction in the number of molecules. In accordance with the above chemico-technological principle, these reactions should be effected under the action of a continually increasing pressure.

2. As it shows the calculation of the heat of formation all the above reactions (except the first) are exothermic reactions. They should be effected simultaneously with energetic cooling.

3. These synthetic reactions are accompanied by an absorption of the gases CO and H and consequently their partial pressures continually decrease. According to the above principle these reactions should be effected under the influence of a continuously increasing concentration of this gas.

It follows that the reaction of the synthesis of these light oils from water gas should be effected at a continually increasing pressure whilst continually lowering of temperature and with a continuous increase of the concentration of the gases H and CO.

The variations in the direction for the three factors of chemical equilibrium during the reaction may be effected in the apparatus shown by way of example in Fig. 1 of the drawings, accompanying the provisional specification.

The compressor 1 of this apparatus is constructed entirely as a Diesel engine. This apparatus only differs from a Diesel engine by the pulley 5 which is mounted on the main shaft of the compressor. The compressor can be driven by this pulley. This compressor like a Diesel engine, is provided with an auxiliary pump. This pump compresses the water gas to a pressure much higher than the pressure of the gas in the cylinder.

The auxiliary pump of the Diesel engine for the fuel may be omitted in this case but when working at a high temperature it is possible to use it for the injection of water into the cylinder for increasing the cooling of the gas.

These two pumps are entirely constructed like the corresponding pumps in a Diesel engine. They are not shown in Fig. 1.

The compressor 1, like a Diesel engine, is provided with four valves as follows:

1. The inlet valve 2 through which the gas from the tank 6 enters the cylinder.

2. The spraying valve 4 through which a jet of gas, at a pressure higher than the pressure of the gas in the cylinder enters as a spray into the cylinder for increasing the concentration of the gases in the compressor and for thoroughly cooling them. This gas is compressed by the auxiliary pump.

3. The exhaust valve 3 through which the products obtained escape into the tank 8.

4. The starting valve which corresponds with the starting valve in the Diesel engine. This valve is not shown in the annexed drawing.

By means of this valve, at the commencement of operation, the compressed gases can be passed directly into the tank 6 for raising the temperature therein. It is needed only if the communication between the reservoirs 6—8 is to be cut off.

All these valves are constructed like the valves of a Diesel engine and are actuated by distribution shafts so as to operate on the two or four stroke cycle.

The products obtained pass through the pipe 12 into the tank 8 where they are cooled and liquefied. The liquefied products leave the apparatus by the pipe 9 and the non-absorbent gases and the permanent gases formed by the process again enter by way of the tank 6 and the pipes 11 for the purpose of being again treated.

During the working of the compressor it is possible to regulate the temperature in the tank 6 by the valves 13 and 14, allowing a portion of the gas to enter the tank directly without cooling. By means of the valve 17 it is possible to interrupt the communication between the tank 6 and the tank 8. It will be understood that the tank 6 should be filled continuously with gas in proportion as the gases are absorbed during the process. Care should also be taken that the gas in the tank is of constant composition. Sometimes it is more practicable to interrupt the communication of the tank 8 with the tank 6 directly. Then the gas not absorbed can be conducted from the tank 8 to the tank 6 by a special compressor which sucks the gases from the tank 8 and causes them to escape into the tank 6.

In Fig. 2 is shown a modification of the compressor 1. This modification differs from the compressor in Fig. 1 by a special reaction chamber 15, of which the walls may be cooled directly from the outside. If the operation is

carried out with a catalyst this may be placed in this special chamber.

Fig. 3 shows a further modification of the compressor 1 particularly constructed for operation with a catalyst. In this modification the reaction chamber is terminated by the pipe section 16 where the catalyst may be placed between metal nets and also in the chamber as shown at 15. The exhaust valve is placed in this compressor behind the section 16. The gases are thus caused to pass through the layer of catalyst. In Fig. 2 and 3 the valves are indicated by the same references as the corresponding valves of the compressor in Fig. 1.

In the apparatus above described the synthesis of the organic body without using a catalyst may be effected in two stroke in the following manner:

1. First stroke. The piston 7 moves towards the outside and the water gas which is in the tank 6 fills the cylinder 1 of the compressor. The gas has for example a temperature of 62° C., a pressure of 4 atms. and concentrations 40% CO and 50% H_2.

2. Second stroke. The piston moves towards the interior and compresses the gas enclosed for example during $^3/_4$ of its return stroke. The gas will have then the pressure of 21 atm. and 270° C. At this moment the valve 4 opens and a jet of gas, of room temperature and of a pressure of 160 atmos. enters the cylinder. This jet of gas in view of its low temperature and particularly by its expansion in the cylinder thoroughly cools the enclosed gas. At the same time this jet of water gas increases the concentration of the water gas in the cylinder. This gas at high pressure is supplied by the auxiliary pump of the compressor. The gases injected make 20% of the gases included in the cylinder and are of the same composition.

Without cooling by the walls of the cylinder and without the admission of the jet of gas the final temperature and pressure will be 700° C. and 140 atm. calculated by the formulæ of adiabatic compression, when the compression space is 8% of the cylinder. Nearly 12% of the gases will be converted into hydrocarbons. The concentration of CO & H in the cylinder will not diminish during the reaction.

When the piston approaches the dead point the valve 3 opens and the mixture of gases and vapours escapes into the tank 8, where this mixture is subjected to cooling by a jet of water or other liquid. The liquefied vapours leave the apparatus by the pipe 9. The gases which have not been absorbed pass into the tank 6 so as to be again treated.

In this manner is effected the synthetic process of producing light oils without using a catalyst from water gas supplied continuously and simultaneously under the action of an increasing pressure, cooling and increase in the concentration of the hydrogen and carbon oxide in accordance with the above chemico-technological principle.

If for this process a catalyst is to be used the reaction is to be effected in a compressor of the form shown on Fig. 2 or 3, where the gases can be brought in contact with a catalyst.

As a result of the operation above described there are obtained organic bodies indicated in the preamble to the present description. As the temperature in the preceding example was chosen sufficiently high, the products are more hydrogenated and less oxygenated bodies are obtained.

The variations of temperature and pressure fixed above are not the only ones under which this synthesis can be effected. This synthesis can be effected equally well at lower zone of temperatures and at higher zone of temperatures. The more the zone of temperature of the operation is increased the higher should be the zone of pressure chosen and on the contrary the lower the zone of temperature chosen for the operation the lower will also be the zone of pressure chosen for this operation. The higher the temperatures of the operation, more the products obtained will contain hydrogenated products derived from the oxygenated hydrocarbons formed before, when the temperatures were low. The lower the zone of temperature of the operation, the more products obtained will contain oxygenated products.

In the described apparatus the variation of the zone of temperatures and pressures can be effected by a suitable choice of the initial temperature and pressure in the tank 6. The variation of concentrations can be effected by the variation of the initial pressure of the gases injected or by variation of the time of opening of valve 4.

As all these synthetic reactions are exothermic reactions the gases and vapours enclosed in the cylinder will contain a large quantity of heat which may be converted into mechanical energy. For this purpose the process above described may be effected not in two but in four stages. The gases and vapours obtained by the compressor at the end of the second stage remain in

the cylinder and the operation is continued in the following manner:

Third stage. The piston moves towards the exterior and the gases and vapours obtained expand and convert their heat energy into mechanical work. By reason of the expansion and under the action of cold walls the gases and the vapours are considerably cooled and there is thus avoided a reversal of the reaction. If this cooling is not sufficient, it may be increased by the introduction into the cylinder of a jet of cooled gas or even a jet of water.

Fourth stage. The piston moves towards the interior of the cylinder and the gases and the vapours already well cooled and expanded escape from the cylinder by the open exhaust valve 3 and enter the tank 8.

Instead of obtaining mechanical energy in the compressor itself the gas may be allowed to expand in a special cylinder interposed between the compressor and the tank 8. In this manner it is possible to obtain a quantity of mechanical energy sufficient to conduct the whole synthetic process with comparatively smaller or no expense of external mechanical energy.

The process above described may be used for the synthesis of light oils not only from water gas but also from coke-oven gas, producer gas, illuminating gas, blast furnace gases by adding the necessary quantity of hydrogen.

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. A process of synthesis of light oils, suitable to be burnt as fuel in automobile and aeroplane engines from water gas, producer gas, coke-oven gas, illuminating gas, blast furnaces gas and similar gases all of them mixed or not mixed with hydrogen, characterized by the fact that the synthesis is produced in the interior, of a compressor, whilst continually increasing the pressure whilst cooling and whilst continually increasing the concentration of the gases all these influences being exerted simultaneously, the cooling and the increasing of concentration of the gases being produced by a jet of the same gases injected in the cylinder by over pressure during the reaction.

2. A process according to Claim 1, characterized by the fact that a compressor in the form of a Diesel engine is used.

3. A process according to Claim 1, characterized by the fact that catalysts are placed in the reaction chamber.

4. A process according to claim 1, characterized by the fact that the heat energy of the gases after reaction is converted into mechanical work.

Dated the 1st day of November, 1925.

M. BRUTZKUS.

Abingdon: Printed for His Majesty's Stationery Office, by Burgess & Son.
[Wt. 56A.—50/2/1929.]

2nd Edition

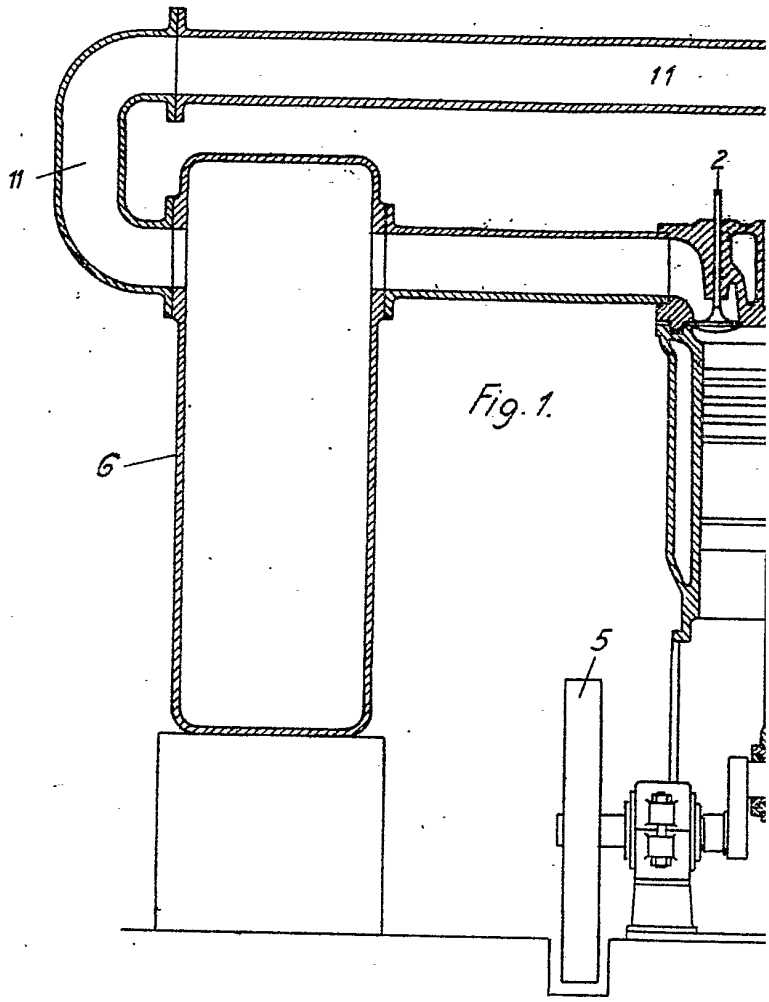


Fig. 1.

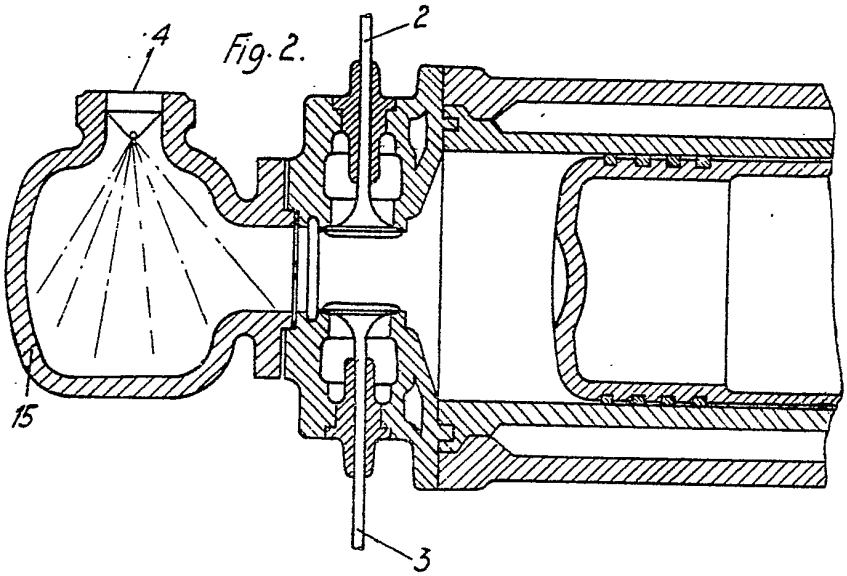


Fig. 2.

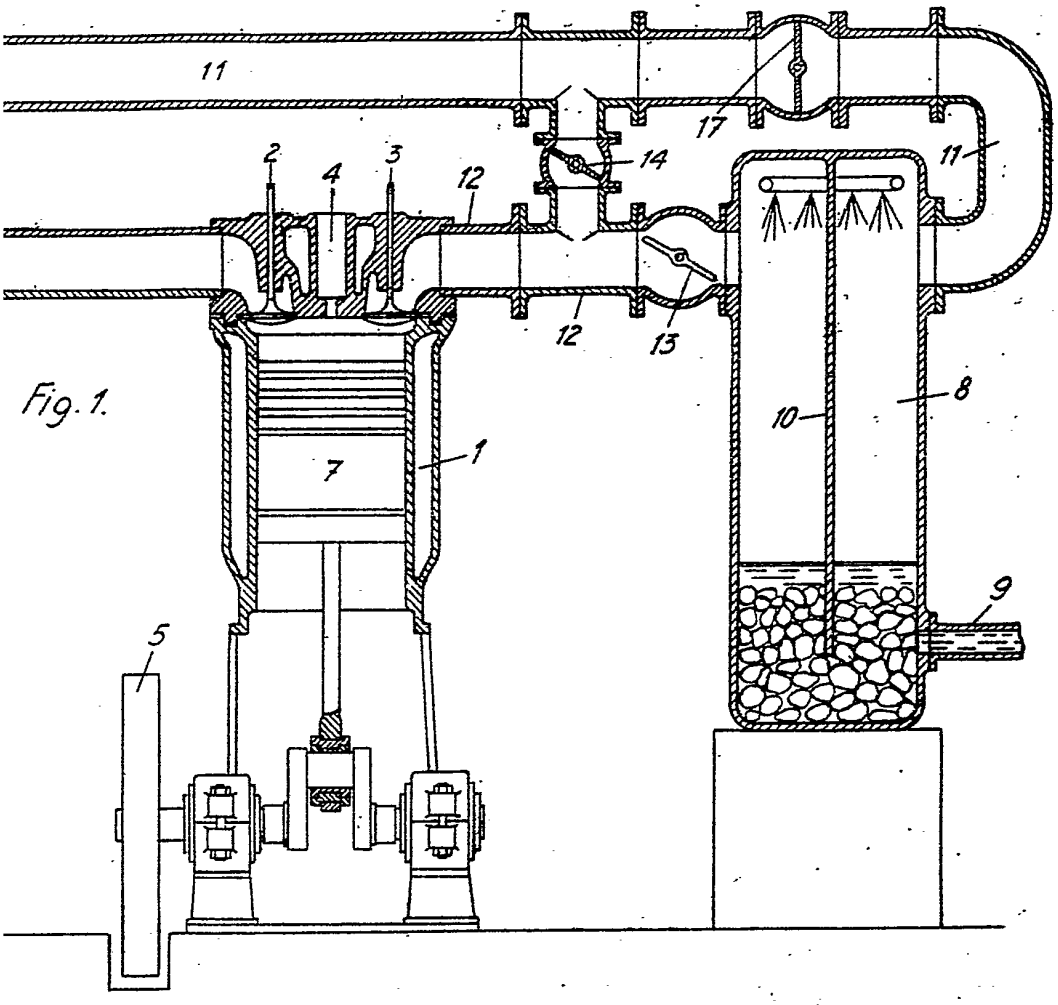


Fig. 1.

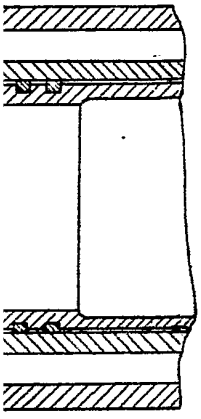
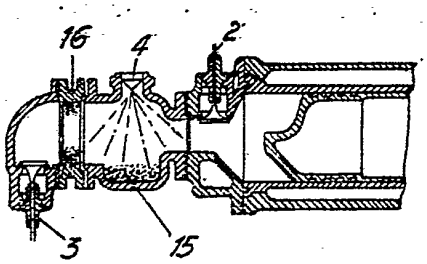


Fig. 3.



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252,786 PROVISIONAL SPECIFICATION

2nd Edition

1 SHEET

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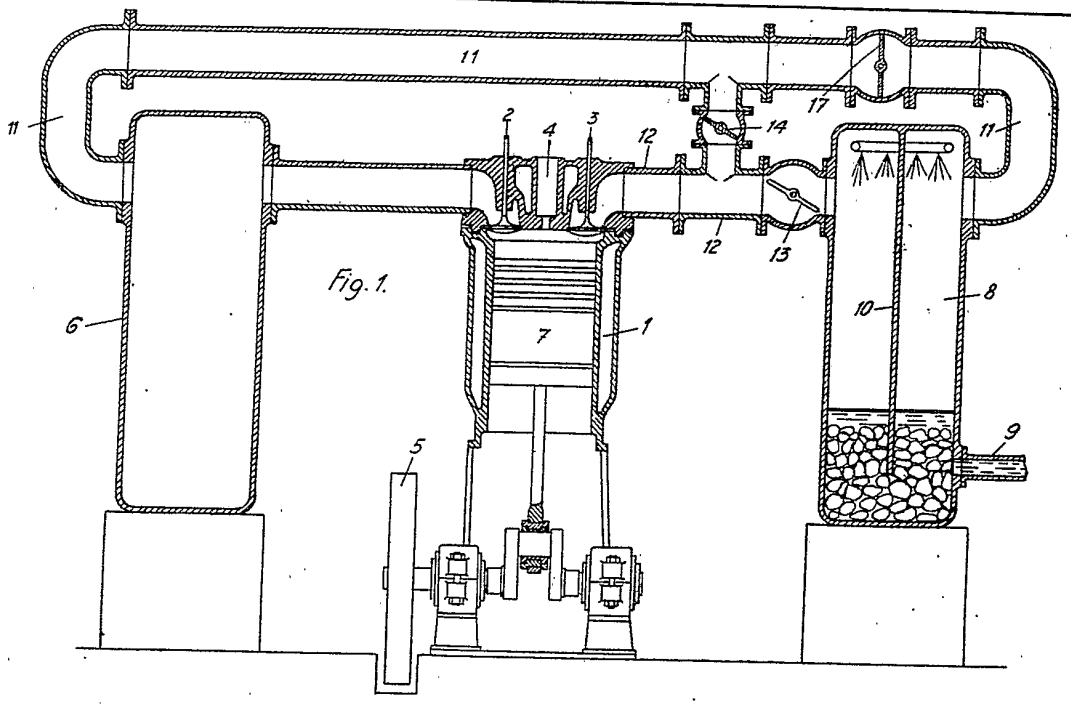

Fig. 1.

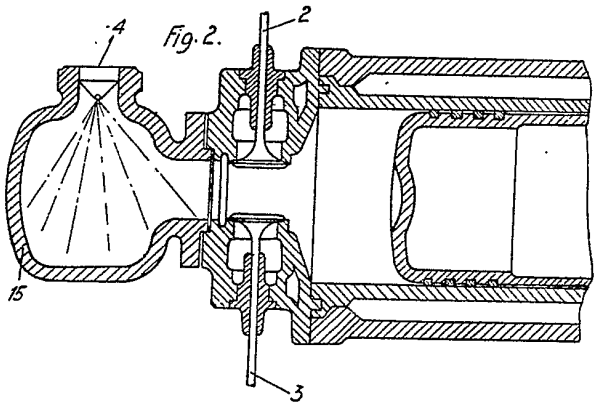

Fig. 2.

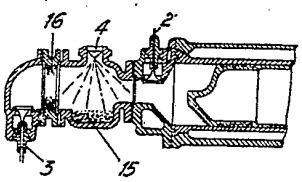

Fig. 3.

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