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AN IONISABLE ATMOSPHERE

3,421,804

Filed March 7, 1967

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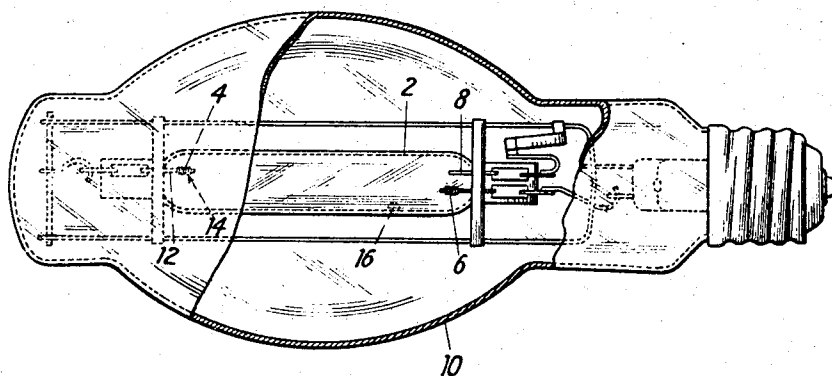


Fig. 1

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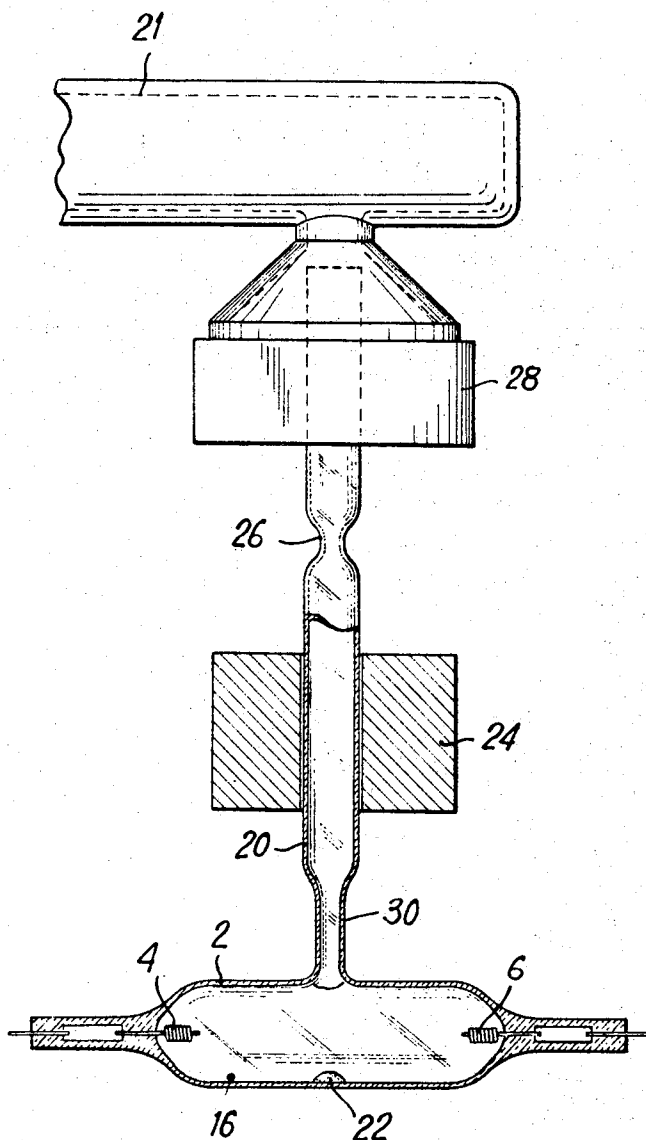
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Fig. 2



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## PROCESS FOR FILLING AN ELECTRIC DISCHARGE LAMP HAVING AN IONISABLE ATMOSPHERE

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Int. Cl. H01j 9/38

8 Claims

### ABSTRACT OF THE DISCLOSURE

The gas introduced into the discharge tube of the lamp contains hydrocarbons in order to increase the metal vapour content of the discharge atmosphere during functioning. In some cases, after electric discharges have been caused in the tube, the latter is pumped then the metals for the discharge atmosphere are sent back into the tube.

The present invention relates to a process for filling an electric discharge lamp having an ionisable atmosphere containing at least one rare gas and at least one metal vapour or metal-compound vapour. It is characterized by the fact that the said process comprises a step in which there is introduced into the lamp at least one organic compound which will produce, during the operation of the lamp under normal conditions, organic radicals capable of combining with the said metal or with a compound of the said metal, for example with a halogenated compound.

Preferably, the said organic compound contains no oxygen; for example, it is a hydrocarbon introduced under an absolute pressure of 0.1 to 10 mm. Hg.

In addition to the organic compound, hydrogen may be introduced, preferably under an absolute pressure of 0.1 to 50 mm. Hg.

When the said metal or metals, in compound form, the organic compound and where necessary the hydrogen have been introduced, but before the lamp is sealed, the latter may be successfully subjected to the following operations:

- (a) Electric discharge in the lamp;
- (b) Evacuation of the gases remaining after this discharge, with optional condensation of the metals, whether combined or not, but not of the organic compounds;
- (c) Filling with pure rare gas and optional reintroduction of the metals condensed in the previous operation.

The process of the invention is particularly useful in the case of discharge lamps in which it is desired to produce the radiation of certain low-volatility elements and which, although the temperature of the cold point in contact with the discharge atmosphere is at least 500° C., would only emit the radiation of these low-volatility elements in scarcely perceptible form or over only a fraction of the distance between the electrodes of the lamp under consideration, if this process were not employed.

It has been found that the organic radicals have the advantage of increasing the proportion of metal, or metal compound, in the atmosphere in which the discharge takes place, even if the metal, or its compound, is in the form of dry vapour during normal operation. Lamps operating by discharges in mercury under medium pressure, to which there are added inorganic metallic halides giving rise to the emission of the spectrum of the metals of these halides, are known, but these halides have low volatility and often tend to be deposited in solid form on the coldest parts of the lamps, in the form of opaque deposits.

On the other hand, it has been observed that, both at very high temperature and at relatively low temperature, certain metals, in the presence of organic compounds,

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notably halogenated organic compounds, change into the form of volatile compounds and no longer give rise to the formation of opaque deposits. It is probable that organometallic compounds are formed which have a much higher vapour pressure than the corresponding inorganic metallic halides.

In addition, it is found that, with appropriate quantities of the introduced substances, a lamp of this form gives, over the whole length of the arc, a uniform light emission due to the excitation of all the metals present in its ionisable atmosphere. Some of these metals, for example the alkali metals, would only have emitted their radiations over a limited part of the length of the arc if no organic compound had been introduced into the lamp.

One object of this invention is to provide electric discharge lamps emitting the spectra of the metals from substances which are in the form of dry vapour when the lamp is in normal operation, and thus, in the case of mixtures of several of these metallic compounds, to obtain a radiation of constant spectral composition which no longer depends upon the variations of vapour pressure as happens when certain constituents of the discharge atmosphere are in the form of dry vapour and others in the form of saturated vapour.

Another object is to obtain the spectra of the metals of those substances with a lower load per square centimeter of surface of the discharge tube.

Still another subject is to avoid more or less opaque deposits emanating from constituents of the discharge atmosphere.

For producing these radicals, it is possible to introduce into the lamp unoxidised organic substances: hydrocarbons, hydrocarbons halides, organometallic compounds, etc.

Further features and advantages of this invention will appear from a detailed description of the drawings in which:

FIGURE 1 shows a discharge lamp for which the process of the invention has been employed.

FIGURE 2 discloses an apparatus for carrying out the process of the invention.

The lamp of FIGURE 1 is of the general known type comprising a discharge tube 2 provided with two main electrodes 4 and 6 and with an auxiliary electrode 8, the said discharge tube being enclosed in a bulbous envelope 10, by which it is thermally insulated and on the inside face of which a fluorescent substance may be deposited. The said tube is, for example, a silica tube having an internal diameter of 20 mm., and a distance between its main electrodes of 60 mm. Each of the said electrodes is constructed, for example, as described in French Patent No. 1,464,066. A refractory support comprising a rod 12 of thoriated tungsten and a helix 14 of pure tungsten supports, in its zone consisting of the helix and the neighboring rod portions, notably the end of the electrode, a sintered mixture of thoria powder and of thorium powder in equal weights.

The tube 2 contains, while cold, before being put into operation:

- a drop 16 of 0.06 g. of mercury alloyed with 0.001 g. of indium;
- argon under pressure of 20 mm. Hg.;
- ethylene bromide under a pressure of 10 mm. Hg.

This lamp emits both mercury radiations and indium radiations for a power not exceeding 200 watts, although its dimensions are those of a usual 400 watt lamp.

The spectrum of the organic radicals appears only during the starting of the lamp, in the form of a spectrum of bands; these radicals, however, reduce the metallic deposits on the tube 2.

There may be employed in addition to or in place of

the ethylene bromide one or more other saturated or unsaturated halides of aliphatic carbons—or again one or more organometallic compounds for example those of mercury, of the alkali metals, or of calcium, barium, the rare-earth metals or the transition metals such as yttrium and scandium. The partial pressure under which the hydrocarbon halide is introduced is preferably between 0.1 and 50 mm. Hg.

The metal or metals contained in the ionisable atmosphere are introduced into the lamp in the free state or in the form of compounds, for example of halides.

The organic radicals may also be introduced in the form of one or more saturated or unsaturated hydrocarbons instead of an aliphatic hydrocarbon halide such as ethylene bromide.

By way of example, another discharge tube according to the invention may consist of a tube of transparent fused silica having an internal diameter of 17 mm., which is provided at its ends with two electrodes consisting of refractory metal, spaced 50 mm. apart. There have been introduced into this tube measured quantities of mercury, thallium iodide and indium iodide such that they are completely volatilised when this lamp is operated at 400 watts. There has also been introduced an iodide of low vapour pressure such as indium iodide. In addition, there has been introduced a gaseous filling consisting of argon under a pressure of 20 mm. Hg. and of ethylene under a pressure of 0.25 mm. Hg.

The said discharge tube is disposed in known manner, as illustrated in FIGURE 1, within a bulb which limits the heat exchanges with the exterior.

The spectral distribution and the intensity of the light produced by this lamp are uniform over the entire length of the arc. This effect may even be obtained if the diameter of the lamp is considerably reduced and even if its length is increased. In the presence of ethylene, a uniform colouration is obtained between the two electrodes, even in a tube having a diameter of 10 mm., with an inter-electrode spacing of 100 mm., which is filled with mercury, argon and halides of low volatility, such as those of the alkali metals.

The electrical characteristics of lamps of this form to which organic compounds have been added are appreciably modified as compared with those of lamps containing no organic compounds. Thus usual lamps which have, with a power of 400 w., a discharge voltage of about 135 v. with a current of 3.2 a. have, when ethylene is added thereto, a discharge voltage of the order of 400 v. for a current of 1 ampere. The same is the case with the other hydrocarbons.

The possibility of reducing the current intensity with a given power makes it possible to increase the power which may be employed in these lamps, because it is known that, in a silica lamp, the current increase is limited by the small cross-sectional areas of the current lead-in conductors of the molybdenum-strip type which are generally employed. Thus, by employing organic materials, and more particularly ethylene, the power of the lamp can be very greatly increased for the maximum current permitted in practice by the lead-in conductors.

This effect is particularly advantageous for lamps intended to be supplied with pulses in order to obtain high momentary powers. Such lamps may be employed for the excitation of lasers, by reason of the high luminous efficiencies and the high luminance obtained in the emission of certain intense spectral lines of metallic vapours such as the line 535 mm. of thallium which is situated in the excitation zone of ruby lasers.

On the other hand, in other cases, it may be advantageous to retain in the discharge lamp the electrical characteristics which it would have if ethylene or another organic compound had not been introduced into it.

This may be done by modifying the process described in the foregoing, namely by subjecting all of these substances to the action of an electric discharge, while con-

densing any products which may be formed, after introduction of the mercury, the other free or combined metals, the organic compounds and the rare-gas atmosphere. The excess of residual gaseous substances is thereafter pumped-out, and the final filling with rare gas is carried out.

FIGURE 2 diagrammatically illustrates by way of non-limiting example an apparatus for carrying out this modified process.

The illustrated lamp, which is rather similar to that of FIGURE 1, comprises two electrodes 4, 6 supported by lead-in conductors sealed in the ends of the lamp. Its stem 20 is not closed, but is connected to a vacuum duct 21. There has been passed through the lamp fixed to the apparatus a prolonged electric discharge, while the lamp contained mercury 16, iodides 22, argon and an organic compound, for example ethylene.

The discharge first occurred above all in the mixture of argon and organic compound and then, as the heating proceeded, there successively appeared the characteristic lines of mercury, and of the metals of the halides introduced.

A cold trap 24 was disposed around the stem 20 close to the junction between the stem and the lamp to condense the products vapourised under the action of the discharge. It is sufficient to cool the stem to about  $-10^{\circ}$  C. After a period of time whose order of magnitude will hereinafter be indicated, the electric discharge is interrupted, the lamp is allowed to cool until the volatilised products have completely condensed, the gaseous atmosphere of the lamp is pumped so as to eliminate the excess of gaseous organic constituent, and the lamp is refilled with argon under the usual pressure of about 20 mm. Hg. The stem is thereafter temporarily sealed at 26 between the cold trap and the junction 28 to the vacuum duct 21.

When the combined trap and lamp have been separated from the duct, the products which have condensed in the trap are sent into the lamp 2, the trap being heated. After sealing, the stem is cut at 30, in the neighbourhood of the discharge tube proper. The lamp thus formed is mounted in the usual way in a bulb to protect it from heat losses.

The intensity of the current for the electric discharge treatment before sealing is of the order of 1 minute, the tube having the dimensions and filling indicated in the foregoing example, in which the tube has an internal diameter of 17 mm.

If, after sealing, the lamp thus treated is subjected to a discharge of a power of 400 w., it is found that, instead of a high discharge voltage of the order of 400 v. as in the aforesaid example, there is obtained during normal operation a voltage and a current which are substantially equal to those of a similar lamp into which no organic compound has been introduced, that is to say, of the order of 135 v. The effect of uniformity of luminosity and of the spectral distribution and that of the absence of any production of halide deposits during operation of the lamp are maintained. It is also found that, at the commencement of the operation of the lamp, there immediately appears a band spectrum on which there are rapidly superimposed the spectrum of mercury and then the spectrum of the metals of the halides, at the same time as the band spectrum disappears.

It is possible, but this is not certain, that these phenomena are due to the formation of organometallic compounds. Thus, in the case of the above lamp, which was exhausted after discharges had passed therethrough, but which contains only mercury, sodium iodide and gas, it is found that if this lamp, after having been sealed, is set in operation with a current of 500 ma., the spectrum of mercury is almost exclusively obtained. If it is thereafter operated with a current of the order of 3 amperes, then after the mercury spectrum, the sodium spectrum is very rapidly obtained over the entire length of the arc, while at the same time the mercury spectrum decreases in in-

tensity. If then, after extinction and cooling, the lamp is again operated at 500 ma., the sodium spectrum is almost exclusively obtained, in contradistinction to what happens in the first operation. In the first phase at 500 ma., there has perhaps been formed an organometallic compound of mercury which, during the discharge at 3 a., has given rise to the formation of organometallic sodium compounds of higher vapour pressure than the mercury.

The treatment by the discharge before the evacuation of the residual gases must be limited in time in order to avoid the formation of carbonaceous products on the walls of the lamp. This time is of the order of one minute.

This time may be made less critical, and the said carbonaceous deposits avoided by carrying out this treatment in a highly hydrogenated atmosphere, for example by introducing, in addition to the aforesaid constituents, hydrogen having a partial pressure at least of the order of that of the organic compound introduced if it contains a double bond. The partial pressure of this hydrogen is preferably between 0.1 and 50 mm. Hg. It is possible that, in this way, the carbon due to the decomposition of the hydrocarbon reacts with the hydrogen to give volatile organic compounds which are thereafter evacuated.

The hydrogen may also be replaced by a compound of high hydrogen content, for example a saturated hydrocarbon such as methane, ethane, etc.

In the foregoing description, the use of ethylene as hydrocarbon has been referred to. In fact, the same results may be obtained with other hydrocarbons, more particularly with those having one or more double or triple bonds, such as vinylic or acetylenic hydrocarbons. For example, acetylene has proved very effective. The additional bonds favour the reaction.

The use of the process of the invention is particularly advantageous in discharge lamps whose atmosphere contains metals whose halides have a low vapour pressure, such as the alkali metals, the rare-earth metals, scandium and thorium.

What I claim is:

1. A process for filling an electric discharge lamp having an ionisable atmosphere containing at least one rare gas and at least one vapour of a metal or metal compound, which process comprises a step in which there is introduced into the lamp at least one organic compound which will produce during the operation of the lamp under normal conditions organic radicals which are ca-

pable of combining with the said metal or with a compound, for example with a halogenated compound of this metal.

2. A process as claimed in claim 1, wherein said organic compound contains no oxygen.

3. A process as claimed in claim 2, wherein said organic compound is a hydrocarbon which is preferably introduced under an absolute pressure of 0.1 to 10 mm. Hg.

4. A process as claimed in claim 3, wherein said hydrocarbon is ethylene or acetylene.

5. A process as claimed in claim 1, wherein said organic compounds is a halide of an aliphatic hydrocarbon preferably introduced under a pressure of 0.1 to 5 mm. Hg.

6. A process as claimed in claim 1 wherein said organic compound is an organometallic compound, for example an organometallic compound of mercury, of alkali metal, of alkaline-earth metal, of a rare-earth metal, of scandium or of yttrium.

7. A process as claimed in claim 1, wherein in addition to the organic compound hydrogen is introduced, preferably under an absolute pressure of 0.1 to 50 mm. Hg.

8. A process as claimed in claim 1, wherein, when the metal or metals in elementary form or in compound form, the organic compound and, where necessary, the hydrogen have been introduced, but before the lamp is sealed, the latter is successively subjected to the following operations:

- (a) electric discharge in the lamp;
- (b) evacuation of the gases remaining after this discharge, with optional condensation of the combined or uncombined metals, but not of the organic compounds introduced;
- (c) filling with pure rare gas and optional reintroduction of the metals condensed in the preceding operation.

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RICHARD H. EANES, JR., *Primary Examiner*.

UNITED STATES PATENT OFFICE  
CERTIFICATE OF CORRECTION

Patent No. 3,421,804

January 14, 1969

André Taxil

It is certified that error appears in the above identified patent and that said Letters Patent are hereby corrected as shown below:

In the heading to the printed specification, line 9, after "57,985" insert -- ; Nov. 16, 1966, 83,863 --.

Signed and sealed this 17th day of March 1970.

(SEAL)

Attest:

Edward M. Fletcher, Jr.

Attesting Officer

WILLIAM E. SCHUYLER, JR.

Commissioner of Patents