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Silver

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[54] **ELECTRIC DISCHARGE LAMP HAVING A FILL INCLUDING NIOBIUM PENTAIODIDE COMPLEXED WITH AN INORGANIC OXO-COMPOUND AS THE PRIMARY ACTIVE COMPONENT**

[75] Inventor: **H. Graham Silver**, Kings Point, N.Y.

[73] Assignee: **General Telephone & Electronics Laboratories Incorporated**, New York, N.Y.

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[51] **Int. Cl.** **H01j 61/18**

[58] **Field of Search** 313/225, 229, 184, 313/227

[56] **References Cited**

UNITED STATES PATENTS

3,234,421	2/1966	Reiling	313/225 X
3,514,659	5/1970	Gungle et al.	313/184
3,521,110	7/1970	Johnson	313/229 X

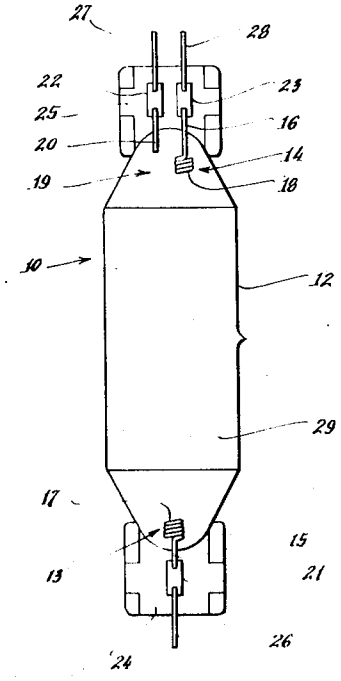
Primary Examiner—Palmer C. Demeo

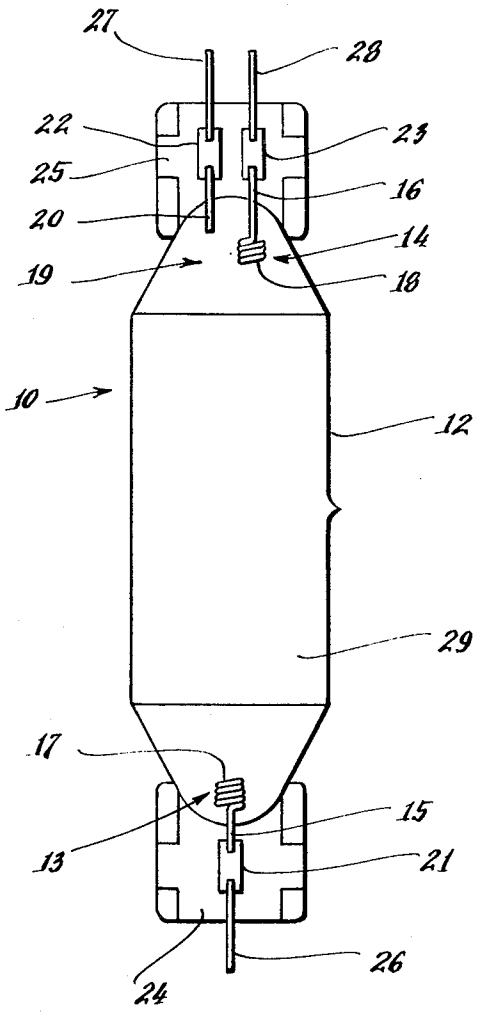
Attorney—Irving M. Kriegsman

[57] **ABSTRACT**

An electric discharge device comprising a sealed light-transmissive envelope; and a fill within the envelope, the fill including, as the primary light-emitting material, niobium pentaiodide complexed with an inorganic oxo-compound, particularly an oxytriiodide of a Group VA element or an oxydiiodide of a Group VIA element.

22 Claims, 1 Drawing Figure

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ELECTRIC DISCHARGE LAMP HAVING A FILL INCLUDING NIOBIUM PENTAIODIDE COMPLEXED WITH AN INORGANIC OXO-COMPOUND AS THE PRIMARY ACTIVE COMPONENT

BACKGROUND OF THE INVENTION

This invention relates to electric discharge devices, particularly of the type wherein the fill includes inorganic adduct molecules of niobium pentaiodide and inorganic oxo-compounds.

In copending patent application Ser. No. 229,933 filed Feb. 28, 1972, and assigned to the assignee of the present invention, there is described electric discharge devices which include, as the primary light-emitting component of the fill, a volatile oxytrihalide of a Group VB element, such as niobium. These devices have been shown to be exceptionally fine light generators, producing light of excellent quality and high efficiency. The molecular species Nb-O, when dissociated from the halogen of the niobium oxytrihalide in the arc of a discharge lamp, emits a dense line spectrum which lies predominantly in the visible portion of the spectrum, thus producing a light source with extremely desirable color characteristics. Niobium oxytrichloride and niobium oxytribromide have sufficiently high vapor pressures (at temperatures regularly achieved in such an arc lamp running under normal conditions) that there is no need either for external heating of the sealed envelope or for the mounting of the sealed envelope within an outer glass envelope. However, in order to minimize electrode transport problems, the moderately volatile niobium oxytriiodide is the presently preferred fill component for generating the molecular species Nb-O. The use of niobium oxytriiodide, however, has a drawback in that it decomposes to form relatively involatile niobium oxydiiodide and molecular iodine at slightly elevated temperatures. An arc lamp containing niobium oxytriiodide, therefore, has to be heated strongly to around 900°C to produce the desired Nb-O molecular emission. This necessitates the use of external heating of the sealed envelope and, optionally, the mounting of the discharge device within an outer glass envelope.

It would, therefore, be desirable to provide an electric discharge device which makes use of an oxyiodide fill which yields the molecular species Nb-O, but does not require external means to heat the fill to higher than normal operating temperatures while generating light of a desired and acceptable spectral quality.

OBJECTS OF THE INVENTION

It is, therefore, an object of the present invention to provide a novel electric discharge device.

It is a further object of the present invention to provide an electric discharge device having a volatile active component which yields, during operation of the device, the molecular species Nb-O.

It is a further object of the present invention to provide an electric discharge device having a volatile active component which yields the molecular species Nb-O at normal operating temperatures whereby external means for increasing the temperature of the device is not needed.

It is a further object of the present invention to provide an electric discharge device which includes, as the primary active component, adduct molecules of niobium pentaiodide and an inorganic oxo-compound of a triiodide of a Group VA element or an oxo-compound of a diiodide of a Group VIA element.

It is still a further object of the present invention to provide an electric discharge device which includes, as the primary active components of the fill, niobium pentaiodide, an oxygen yielding material, and a triiodide of a Group VA element or a diiodide of a Group VIA element.

These and still further objects, features and advantages of the present invention will become apparent upon consideration of the following detailed disclosure.

BRIEF SUMMARY OF THE INVENTION

These and still further objects of the present invention are achieved, in accordance therewith, by providing an electric discharge device having a sealed light-transmissive envelope and a fill within the envelope; the fill including, as the primary active component, adduct molecules of niobium pentaiodide with an inorganic oxo-compound. Of particular interest are the oxytriiodides of Group VA elements and the oxydiiodides of Group VIA elements. Such adduct molecules can be conveniently generated within the sealed envelope by an in-situ technique by mixing together, within the sealed envelope, equimolar quantities of niobium pentaiodide, an oxygen yielding substance, such as mercuric oxide, and a triiodide of a Group VA element or a di-iodide of a Group VIA element. Also of interest are the adducts of niobium pentaiodide with sulfur dioxide or sulfur trioxide. This in-situ technique produces, during operation of the device, adduct molecules having the formula $NbI_5 \cdot XO_mI_n$ where X is a Group VA or Group VIA element, m is 1, 2 or 3 and n is 0, 2 or 3. In operation, after an initial warm-up period of a few seconds, the discharge assumes an intense pink-white color which is characteristic of the emission of the molecular species Nb-O, thus demonstrating that a highly volatile compound has been synthesized in-situ. As indicated above, it is unnecessary to provide external heating or an outer glass envelope to maintain the discharge device at operating temperatures whereat the desired molecular emission is produced.

The envelope contains, in addition, to the adduct herein described, or the components which will yield adduct molecules during operation of the device, a small quantity of a noble gas, such as argon, to facilitate starting. In addition, mercury can be added in a conventional amount if it is desired to have a higher pressure discharge device with a lower operating voltage. Where mercuric oxide is utilized as the oxygen donating material, mercury will inherently be provided, during operation of the device since the mercuric oxide will dissociate to yield the desired molecular oxygen and will simultaneously yield mercury. Furthermore, other well known and conventional materials, such as thallium, sodium or cesium halides, such as, for example, thallium iodide, sodium iodide, and cesium iodide, as well as other materials including cesium oxide, thallium oxide, mercurous iodide, etc. can be added to the fill for their known purposes, for example, to lower operating voltages, adjust spectral output, etc. During operation, the pressure within the envelope is principally generated by the volatilization of the mercury with only a small contribution to the total pressure being made by the volatile complex herein described. It is presently contemplated that total pressure, at operational tem-

peratures, will normally be in the range of about one-half to about 10 atmospheres, generally from about 1 to about 3 atmospheres. At these pressures, the partial pressure of the niobium complex material herein described will generally be on the order of about 2 torr to about 100 torr.

Because of the volatility of the primary active component (i.e. the niobium pentaiodide adduct with an oxo-iodide), the discharge devices of the present invention do not require external wall heaters to maintain the adduct in a volatile state. This is a distinct advantage which not only reduces production cost and chance of earlier operational failure, but is achieved with excellent light output having excellent spectral characteristics. For example, efficiencies on the order of about 60 to 80 lumens per watt have been attained with these devices, while light of excellent quality is generated.

The sealed light-transmissive envelope is generally made of quartz, although other types of glass may be used, such as alumina glass or Vycor, the latter being a glass of high silica content. It is essential, of course, that the material selected for the enevelope and the materials utilized in the fill not adversely react with one another, or with reaction products that are produced during operation. The lamp further includes energizing means located either inside or outside of the envelope, depending on the type of energy applied for producing an electrical discharge inside the envelope.

DESCRIPTION OF SPECIFIC EMBODIMENTS

Referring to the drawing, discharge device **10** includes sealed light-transmissive envelope **12** having main discharge electrodes **13** and **14** at opposite ends thereof. Electrodes **13** and **14,** made of a suitable material such as thoriated tungsten, are supported by lead-in wires **15** and **16,** respectively and have a tungsten helix **17** and **18,** respectively, at their interior ends. An auxiliary starting electrode **19,** generally prepared of tantalum or tungsten, is provided at one end of envelope **12** adjacent to main discharge electrode **14,** and comprises an inwardly projecting end of another lead-in wire **20.** Each of the three current lead-in wires **15, 16** and **20** have their ends welded to intermediate foil sections **21, 22** and **23,** respectively, of molybdenum which are hermetically sealed within pinched sealed portions **24** and **25** of envelope **12.** The foil sections are very thin, for example, approximately 0.0008 inch thick, and go into tension without rupturing or scaling off when the heated envelope cools. Molybdenum or tantalum wires **26, 27** and **28** are welded to the outer ends of foils **21, 22** and **23,** respectively, and serve to convey current to the electrodes inside envelope **12.** Inside envelope **12,** there is volume **29** in which the adduct molecules herein described are volatized, etc., during operation of device **10.**

In operation, the adduct molecules of the present invention dissociate completely into Nb atoms, NbO molecules, I atoms, oxygen atoms, etc. in the arc. Such dissociated components most probably exit only for fractions of a second in the hottest portions of the sealed envelope but recombine in the cooler sections of the envelope to regenerate the volatile adducts. The adducts diffuse into the discharge and dissociate, thereby beginning, once again, this cyclic dissociation and regeneration process. Unfortunately, the spaced electrodes are slowly transported due to the atmosphere containing both oxygen and halogen atoms. Thus, these devices can be operated on the order of up to about 100 hours prior to electrode failure and, accordingly, are limited in the uses to which they may be put. During their lifetime, however, these devices are exceptionally fine light generators, producing light of excellent quality at moderate efficiencies. The electrode failure problem can be eliminated, however, by eliminating the spaced electrodes and replacing them with radio frequency or microwave discharge activators or energizers (not shown). With such energizers, the electrode transport problem is eliminated, whereby the devices can be operated for greater periods of time.

The following Examples are given to enable those skilled in this art to more fully understand and practice the present inventon. They should not be considered as a limitation upon the scope of the invention but merely as being illustrative and representative thereof.

In Examples I – X, the envelope utilized is a 400 watt "shallow-press" (S.P.) envelope made of quartz, having a diameter of about 2.3 cm, a length of about 6.0 cm, a length between electrode tips of about 5.3 cm, and a volume of about 17.8 cc.

The lamps of Examples I – X have been "aged" by running at their full-rated power for short periods of times, on the order of 2 to 18 minutes, after which period the characteristics identified in the Examples were measured without a cooling period.

When given in the following Examples, "strike potential" is defined as the peak AC voltage (with reference to the zero potential) required for lamp ignition, and "peak potential" is defined as the peak AC voltage (with reference to the zero potential) after lamp stabilization when operated at its rated power.

EXAMPLE I

A 400 watt S.P. envelope is filled with 8.1 mg NbI_5, 4.6 mg PI_3, 2.4 mg HgO, 50 mg Hg, and 23 torr argon. These components correspond to the chemical adduct formula $NbI_5 \cdot POI_3$. When operated at 397 watts and 1.55 amps, the lamp had a strike potential of 2,100 volts, a peak potential of 4,000 volts and an efficiency of 45 lumens per watt (lm/w). After an initial warm-up period of a few seconds, the discharge assumed an intense pink-white color.

EXAMPLE II

A 400 watt S.P. envelope is filled with 8.1 mg NbI_5, 5.1 mg AsI_3, 2.4 mg HgO, 51 mg Hg, and 23 torr argon. These components correspond to the chemical adduct formula $NbI_5 \cdot AsOI_3$. When operated at 399 watts and 1.58 amps, the lamp had a strike potential of 2,400 volts, a peak potential of 3,200 volts, and an efficiency of 31 lm/w. After an initial warm-up period of a few seconds, the discharge assumed an intense pink-white color.

EXAMPLE III

A 400 watt S.P. envelope is filled with 8.1 mg NbI_5, 5.6 mg SbI_3, 2.5 mg HgO, 50 mg Hg and 23 torr argon. These components correspond to the chemical adduct formula $NbI_5 \cdot SbOI_3$. When operated at 400 watts and 1.45 amps, the lamp had a strike potential of 4,000 volts, a peak potential of 3,700 volts, and an efficiency of 60 lm/w. After an initial warm-up period of a few seconds, the discharge assumed an intense pink-white color.

EXAMPLE IV

A 400 watt S.P. envelope is filled with 8.1 mg NbI_5, 6.6 mg BiI_3, 2.4 mg HgO, 50 mg Hg and 23 torr argon. These components correspond to the chemical adduct formula $NbI_5 \cdot BiOI_3$. When operated at 401 watts and 1.49 amps, the lamp had a strike potential of 4,000 volts, a peak potential of 3,550 volts, and an efficiency of 38 lm/w. After an initial warm-up period of a few seconds, the discharge assumed an intense pink-white color.

EXAMPLE V

A 400 watt S.P. envelope is filled with 8.1 mg NbI_5, 0.36 mg S, 5.1 mg HgI_2, 2.4 mg HgO, 48 mg Hg, and 23 torr argon. These individual components correspond to the chemical adduct formula $NbI_5 \cdot SOI_2$. When operated at 345 watts and 1.53 amps, the lamp had a strike potential of 1,650 volts, a peak potential of 4,000 volts, and an efficiency of 28 lm/w. After an initial warm-up period of a few seconds, the discharge assumed an intense pink-white color.

EXAMPLE VI

A 400 watt S.P. envelope is filled with 8.1 mg NbI_5, 3.6 mg S, 5.1 mg HgI_2, 4.8 mg HgO, 46 mg Hg and 23 torr argon. These individual components correspond to the chemical adduct formula $NbI_5 \cdot SO_2I_2$. When operated at 400 watts and 1.50 amps, the lamp had a strike potential of 2,200 volts, a peak potential of 4,000 volts, and an efficiency of 60 lm/w. After an initial warm-up period of a few seconds, the discharge assumed an intense pink-white color.

EXAMPLE VII

A 400 watt S.P. envelope is filled with 8.1 mg NbI_5, 3.1 mg HgSe, 5.1 mg HgI_2, 2.4 mg HgO, 46 mg Hg and 23 torr argon. These individual components correspond to the chemical adduct formula $NbI_5 \cdot SeOI_2$. When operated at 400 watts and 1.83 amps, the lamp had a strike potential of 1,400 volts, a peak potential of 2,300 volts, and an efficiency of 48 lm/w. After an initial warm-up period of a few seconds, the discharge assumed an intense pink-white color.

EXAMPLE VIII

A 400 watt S.P. envelope is filled with 8.1 mg NbI_5, 3.7 mg HgTe, 5.1 mg HgI_2, 2.4 mg HgO, 46 mg Hg, and 23 torr argon. These components correspond to the chemical adduct formula $NbI_5 \cdot TeOI_2$. When operated at 412 watts and 1.55 amps, the lamp had a strike potential of 3,200 volts, a peak potential of 4,000 volts, and an efficiency of 78 lm/w. After an initial warm-up period of a few seconds, the discharge assumed an intense pink-white color.

In the following Examples IX and X, the adducts are of NbI_5 and a non-iodated oxide of sulphur.

EXAMPLE IX

A 400 watt S.P. envelope is filled with a 8.1 mg NbI_5, 0.36 mg S, 4.8 mg HgO, 48 mg Hg and 23 torr argon. These components correspond to the adduct having the formula $NbI_5 \cdot SO_2$. When operated at 388 watts and 1.55 amps, the lamp had a strike potential of 1,900 volts, a peak potential of 3,400 volts, and an efficiency of 64 lm/w. After an initial warm-up period of a few seconds, the discharge assumed an intense pink-white color.

EXAMPLE X

A 400 watt S.P. envelope is filled with 8.1 mg NbI_5, 0.36 mg S, 7.2 mg HgO, 46 mg Hg, and 23 torr argon. These components correspond to the adduct having the chemical formula $NbI_5 \cdot SO_3$. When operated at 400 watts and 1.68 amps, the lamp had a strike potential of 2,600 volts, a peak potential of 4,000 volts, and an efficiency of 58 lm/w. After an initial warm-up period of a few seconds, the discharge assumed an intense pink-white color.

EXAMPLE XI

In this Example light-emission is promoted by the microwave activation of the components of the fill within the sealed light-transmissive envelope. The envelope is made of quartz, is about 2.5 inches long, and about 0.75 inch in diameter. The ends are flat with one end having a side arm closely adjacent thereto. The end without the adjacent side arm is inserted into a waveguide. Approximately 1.3 inches of the length of the envelope is within the waveguide. Temperature adjustment and thus control of the amount of the active component in the fill, is by heating or cooling that portion of the envelope which extends out of the waveguide. Excess adduct is present, but definitive weight or concentration measurements of the fill are not taken.

The envelope is filled with NbI_5, HgO, PI_3, 50 mg Hg and 23 torr argon. When operated at a temperature above about 240°C and input power about 400 watts at a frequency of about 2.45 GHz, the discharge is pink-white.

EXAMPLES XII – XIV

Example XI is repeated substituting BiI_3, SeI_2 and TeI_2, respectively, for the PI_3. A similar pink-white discharge is attained.

The present invention is considered distinct and separate from the method described by Branden et al., Act. Chem. Scand., 17, 353 (1963) which describes the formation of a 1:1 complex between $NbCl_5$ and $POCl_3$. Although the authors of the aforesaid article teach that a volatile complex can be formed, they do not relate their activities to electric discharge devices, do not suggest the use of iodide complexes as herein proposed for use in electric discharge devices and, most importantly, because such materials were not used in electric discharge devices, they do not show light emission from the molecular species NbO.

While the present invention has been described with reference to specific embodiments thereof, it will be understood by those skilled in this art that various changes may be made without departing from the true spirit and scope of the invention. In addition, many modifications may be made to adapt a particular situation, material, apparatus, process, or then present objective to the spirit of this invention without departing from its essential teachings.

What is claimed is:

1. An electric discharge device comprising a sealed light-transmissive envelope; a fill within said sealed envelope, said fill including, as the primary active component, an adduct of NbI_5 with an inorganic oxo-compound, said adduct dissociating during operation of said electric discharge device to provide light-

emission from the molecular species Nb-O; and energizing means for producing an electric discharge within said envelope.

2. The discharge device of claim **1** wherein said oxo-compound is selected from the group consisting of sulphur dioxide and sulphur trioxide.

3. The discharge device of claim **1** wherein said oxo-compound is an oxytriiodide of a Group VA element.

4. The discharge device of claim **1** wherein said oxo-compound is selected from the group consisting of POI_3, $AsOI_3$, $SbOI_3$ and $BiOI_3$.

5. The discharge device of claim **1** wherein said oxo-compound is POI_3.

6. The electric discharge device of claim **1** wherein said oxo-compound is $AsOI_3$.

7. The electric discharge device of claim **1** wherein said oxo-compound is $SbOI_3$.

8. The electric discharge device of claim **1** wherein said oxo-compound is $BiOI_3$.

9. The electric discharge device of claim **1** wherein said oxo-compound is an oxydiiodide of a Group VIA element.

10. The electric discharge device of claim **1** wherein said oxo-compound is selected from the group consisting of SOI_2, SO_2I_2, $SeOI_2$, and $TeOI_2$.

11. The electric discharge device of claim **1** wherein said oxo-compound is SOI_2.

12. The electric discharge device of claim **1** wherein said oxo-compound is SO_2I_2.

13. The electric discharge device of claim **1** wherein said oxo-compound is $SeOI_2$.

14. The electric discharge device of claim **1** wherein said oxo-compound is $TeOI_2$.

15. The electric discharge device of claim **1** wherein said adduct is generated in-situ.

16. The electric discharge device of claim **1** wherein said adduct is generated in-situ by providing as a part of said fill, NbI_5, an oxygen donor and a triiodide of a Group VA element or a diiodide of a Group VIA element.

17. The electric discharge device of claim **1** wherein said adduct is present in a partial pressure from about **2** torr to about 100 torr.

18. The electric discharge device of claim **1** wherein said fill further includes a small quantity of a noble gas.

19. The electric discharge device of claim **1** wherein said fill further includes a quantity of mercury.

20. The electric discharge device of claim **19** wherein said mercury is in a quantity sufficient to generate a pressure during operation of about one-half portion about 10 atmospheres.

21. The electric discharge device of claim **1** further including a pair of electrodes spaced within said envelope and capable of being connected, at those portion thereof extending outside of said envelope, to means for applying an electrical potential thereto.

22. The electric discharge device of claim **1** further including microwave activation means for causing Nb-O light-emission from said fill.

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UNITED STATES PATENT OFFICE
CERTIFICATE OF CORRECTION

Patent No. 3,748,520 Dated July 24, 1973

Inventor(s) H. Graham Silver

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

Column 3, line 55, change "volatized" to --volatilized--.
Column 8, Claim 20, line 3, after "one-half" delete "portion" and insert --to--.

Signed and sealed this 26th day of February 1974.

(SEAL)
Attest:

EDWARD M.FLETCHER,JR.
Attesting Officer

C. MARSHALL DANN
Commissioner of Patents

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