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DEVICE AND METHOD FOR DOSING MERCURY INTO A LUMINESCENT LAMP

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Abstract- The work examines the environmental problems of luminescent lamps (LL) associated with mercury and the thermodynamic processes of evaporation and condensation of mercury in the gas-discharge gap. Theoretical studies of a multicomponent mixture of mercury in the gas-discharge gap of a low-pressure LL were carried out. Formulas were obtained for calculating the rate of evaporation and condensation of mercury vapor and the radius of mercury droplets in the lamp. A technical device has been developed for dosing mercury into a LL, in which mercury is dosed in a vapor state. The device contains a heater for evaporating mercury and a controlled electromagnetic valve located along the axis of the reservoir with mercury. By reducing the channels for transporting mercury, the proposed device ensures the accuracy of the dosage of the required amount of mercury and improves the ecological state of the environment. At the end of the article, conclusions and recommendations are made regarding the device and method of vapor dosing of mercury into the lamp, as well as the resulting calculations of evaporation and condensation of mercury droplets of various sizes.

Keywords: Luminescent Lamps, Mercury, Mercury Dosing Device, Mercury Evaporation, Mercury, Condensation.

1. INTRODUCTION

LL production occupies a leading position in the lighting industry. These lamps are widely used in lighting industrial premises, administrative buildings, offices, educational institutions, in technological processes, in medicine, they are used in everyday life, etc. Their distribution is associated with high energy efficiency and lighting quality, determined by the spectral composition of the radiation. That is why in the coming years there are no plans to noticeably replace LL with other light sources. Although intensive work is underway to develop the parameters of competitive LED light sources [1-3], LL also continue to be improved, both in the direction of further increasing luminous efficiency and in the possibility of expanding applications [4]. An integral element of LL is metallic mercury, which in the vapor state serves as a source of ultraviolet radiation, converted by the phosphor into the visible part of the spectrum [5].

There is currently no functionally equivalent replacement for mercury. At the same time, from an environmental point of view, the use of mercury creates a number of problems in the production process and in the disposal of LL that is not suitable for further use [6]. Mercury is a toxic metal, has high biochemical activity, high mobility in natural conditions and resistance to natural neutralizing factors. These characteristics of mercury are dangerous for the human body, where it can enter in the form of vapor, with water, with plant and animal food [7, 8].

In all countries of the world, solving environmental problems not only in LL, but also in other areas of the energy system is of very important strategic importance [7, 9-11]. A radical way to eliminate the environmental problem associated with mercury is to abandon the use of mercury in LL. To a certain extent, the solution to the problem is to expand the production of environmentally friendly light-emitting diode light sources. However, in terms of the complexity of the technology and a number of characteristics, light-emitting diode lamps have not yet reached a noticeable level and therefore cannot serve as a complete alternative to them. The second, less radical way to improve the environmental situation is the well-organized democratization of LL that is not suitable for further use. At the same time, high efficiency democratization requires significant capital investments with relatively low returns as a result of recycling lamp elements. Analysis shows that the overwhelming majority of failed lamps end up in landfills without democratization [12]. Individual lamp democratization installations only to a small extent solve the problem of mercury pollution in landfills, from which mercury is uncontrollably distributed into the environment, soil and water sources [6, 13-15]. The appropriate legislation is needed on mechanisms for the collection and disposal of unusable mercury lamps, which are currently insufficient and whose effectiveness is questionable.

Finally, the third way to significantly reduce environmental pollution from metallic mercury is to minimize amount of mercury introduced into the lamp. The amount of mercury in a LL required for its normal functioning throughout its entire service life is about 5-10 mg [16]. However, a few milligrams are enough to burn a lamp with a standardized light output.

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Mercury is gradually bound by the lamp elements located inside the flask, mainly by the phosphor, which forces the amount of mercury to increase to the given values. Technological difficulties encountered in industrial practice sometimes force us to increase the amount of mercury introduced into the lamp by tens of times compared to the amount that ensures the required service life of the lamp. This is clearly confirmed by the data that in Germany, more than 2 tons of mercury are consumed for the production of 40 million pieces of lamps per year, and in the former USSR, when producing 178 million pieces of mercury lamps, the amount of mercury is about 24 tons [6]. As a result, lamps that are not suitable for further use contain large amounts of free mercury, which pollutes the environment to a much greater extent than mercury bound to the lamp elements.

The second aspect of the issue of limiting the amount of mercury introduced into the lamp is its saving, since mercury is an expensive and scarce metal. European legislation on requirements for sustainable design and construction of lamps has adopted a number of regulations aimed at reducing the mercury content in discharge lamps. The regulations formulate requirements to increase the luminous efficiency of LLs and reduce the mercury content in them, based on the gradual abandonment of lamps with halo phosphate phosphors [17]. An analysis of technical regulations based on the directives of the European Parliament regarding the requirements to reduce the mercury content in LL shows that the manufacturer must provide consumers with the following information: the amount of mercury in the lamp; how to dispose of the lamp after its failure; how to deal with lamp waste if it is accidentally destroyed [18]. Conducted in scientific and technical developments and projects, the issue of accurate dosing and mercury content in LL is also very acute [16]. The limit values for the amount of mercury by types of LL are shown in Table 1.

Table 1. Amount of mercury in LL [16]

Type of fluorescent lamp	Characteristics of lamps	The amount of mercury in the lamp, mg
Linear fluorescent lamp T12 with a diameter of 38 mm	Poor energy efficiency and high mercury content	5-90
Linear fluorescent lamp T8 with a diameter of 26 mm	The most commonly used fluorescent lamp	<10
Linear fluorescent lamp T5 with a diameter of 16 mm	Alternative to T8 fluorescent lamps	<5
Compact fluorescent lamp	Alternative to incandescent lamps	<5

Currently, a significant part of artificial lighting is generated by mercury lamps and it is almost impossible to provide a hygienically sound level of lighting without the use of mercury lamps. The growing trend in light energy consumption indicates that in the near future the volume of use of mercury lamps will remain quite large. Therefore, it is advisable to limit the amount of mercury dosed into the lamp so that during the service life all the mercury is consumed.

For many decades, the most widely used devices dosing mercury in the liquid state [19-21]. With liquid dosing, it is necessary to take into account the large length of the mercury transportation path from the dispenser to the exhaust tube. When transporting mercury into the lamp, a drop of mercury can be deposited on the walls of the dispenser, which leads to an underestimation of the amount of mercury in the lamp, and if a drop of mercury is trapped on the walls, it can lead to an overestimation of its amount. Dosing liquid mercury is difficult to achieve accuracy, which leads to excessive use of mercury, which ultimately affects the environment.

In an effort to improve the accuracy of mercury dosing, technical solutions based on the rejection of liquid dosing prevail. One such solution is the dosing of mercury in the solid state [22]. The melting point of mercury is -38.9 °C, so its use in the solid state requires the use of a special cryogenic installation. Although such a method can be implemented in principle, its application significantly complicates the lamp manufacturing technology. In recent years, dosing of mercury in LL in the form of amalgams has become widespread [23, 24]. Amalgam is a solid or powdered combination of mercury with other metals. The amalgam is welded to the electrode assembly of the lamp and, due to heating with a high-frequency current, the amalgam is thermally decomposed and free mercury is released into the lamp. At the same time, it should be noted that the accuracy of mercury dosing using amalgam dispensers' places high demands on their manufacture and the release of vaporous mercury inside the lamp. Installation of amalgam and its processing inside the lamp also complicates the LL manufacturing technology.

A number of technical developments provide for the dosing of mercury in a liquid state enclosed in an ampoule or capsule [21, 23]. The required amount of mercury is placed in ampoules or capsules made of fusible material. An additional electrode is soldered into the stem. The capsule or ampoule is fixed on the stem using a metal plate and wire, which are welded to the holders of the electrode assembly. When the current source is turned on, the wire heats up and the capsule or ampoule is depressurized and mercury enters the lamp. With this technology, high dosing accuracy can be achieved with a very low mercury content (less than 3 mg) [23]. However, the placement of a strictly dosed amount of mercury in hermetically sealed ampoules or capsules with their installation inside the lamp, further destruction of the shell after the sealing of the lamp bulb also significantly complicates the technology and design of the lamp.

An analysis of existing dosing devices and methods for dosing mercury in LL shows that each direction has positive and negative sides. Dosing mercury in liquid state is difficult to achieve accurate dosage, which leads to excessive consumption of mercury, ultimately negatively affecting the environment. The introduction of mercury in the solid state reduces the excess consumption of mercury, however, this significantly complicates the technological process.

Therefore, the accuracy of mercury dosing increases the durability of lamps, leads to mercury savings, and improves the ecological state of industrial premises and the environment. To improve the environmental situation, solving the problems of accurate dosing of mercury in LL is very important. The aim of the work is to develop effective methods for the dosing of mercury, the design of devices suitable for use in the production of LL.

2. FEATURES OF EVAPORATION PROCESSES AND CONDENSATION OF MERCURY IN LL

Depending on the state of aggregation of mercury in gas-discharge lamps, it is of great interest to analyze theoretical phenomena, methods for calculating the physical processes of evaporation and condensation of mercury in various temperature conditions. A theoretical analysis of the kinetics of evaporation of a two-component mixture (mercury + alkali metal) is considered in [25]. The mechanism of electrical breakdown in gas-discharge lamps involving a mixture of cesium, mercury, xenon, as well as the method of lamp ignition by heating and evaporating the amalgam, is given in [26]. The study of the generation of ultraviolet radiation with a wavelength of 185 nm by a gas discharge in mercury vapor and a Ne-Ar mixture in a low-pressure amalgam lamp has determined the dependence of the intensity and efficiency on the pressure of mercury vapor, buffer gas, and discharge current [27].

There is a clear trend in the work carried out by scientists using a multicomponent mixture in the gas-discharge gap of amalgam mercury lamps. However, there are no results on studies conducted with one-component mercury, and it becomes necessary to analyze thermodynamic processes for small amounts of mercury in LL.

2.1. Calculation of Saturation Vapor Pressure of Mercury Depending on Temperature

The gas discharge in LL occurs in mercury vapor [5]. Numerous experimental results are known for the temperature dependence of saturated mercury vapor pressure [28-30]. The data are obtained using different methods and they are basically the same. However, there are isolated cases where different results are obtained. The greatest discrepancy is observed in the region of relatively low temperatures (≤273 K), in the vicinity of the solidification temperature of mercury and below, reaching a tenfold value. The discrepancies in the region of relatively high temperatures (beginning from room temperature and above) are small and are within 10%. In [28], an empirical formula is given that describes the dependence of the pressure of saturated mercury vapor on temperature in the range of 40-240 °C.

$$\log P = 10.3735 - \frac{3808}{T} - 0.8\log T \tag{1}$$

In this and subsequent formulas, the pressure of saturated mercury vapor P is expressed in mm Hg, temperature T-in Kelvin. In the temperature range of 250-360 °C, there is another expression [29].

$$\log P = 10.59901 - \frac{3335.027}{T} - 0.865327\log T \tag{2}$$

For further analysis, it is desirable to have one formula that describes the dependence of pressure on temperature in a wider range. In [30], the results of measurements of the heat capacity of mercury and calculations of thermodynamic functions are presented. Based on the values of thermodynamic functions and vapor pressure, an equation is given for the dependence of vapor pressure on temperature for liquid mercury in the range of -38.88-500 °C.

$$\begin{gathered}\log P = 11.257555 - \frac{3339.202}{T} - 1.153092\log T + \\ +2.95697\times10^{-4}T - 7.4588\times10^{-8}T^2 - 1.5605\times \\ \times10^{-11}T^3 + 3.6\times e^{-\frac{5360}{T}}\end{gathered} \tag{3}$$

The vapor pressure of liquid mercury, calculated on the basis of Equation (3), is given in Table 2. These data can be used to calculate the rates of evaporation and condensation of mercury droplets of various sizes.

Table 2. Dependence of saturated mercury vapor pressure on temperature [30]

Temperature °C	Mercury vapor pressure, mm Hg	Temperature °C	Mercury vapor pressure, mm Hg
-30	4.78×10^{-6}	100	0.3
0	2×10^{-4}	130	1.19
30	2.7×10^{-3}	150	2.8
50	10^{-2}	170	6.12
80	9×10^{-2}	190	12.8

2.2. Calculation of Evaporation and Condensation Parameters of Mercury in LL

To analyze the processes of evaporation and condensation of mercury in LL, it is necessary to compare the number of molecules that have left the liquid and the number of molecules that condense in the presence of saturated vapor. Since the saturation vapor pressure is known, it is possible to count the number of vapor molecules hitting a unit surface area per unit time. According to the kinetic theory of gases, the indicated number of molecules is determined by Equation (4) [31].

$$N = \frac{1}{4}n\bar{v} \tag{4}$$

where, n is the concentration of vapor molecules; $\bar{v}$ is the arithmetic mean velocity of molecules. The mass of the above number of molecules can be determined by Equations (5) and (6).

$$M_k = N \times m_0 \tag{5}$$

or

$$M_k = \frac{1}{4}n\bar{v}\times m_0 \tag{6}$$

where, m_0 is the mass of one vapor molecule. From the basic equation of the molecular kinetic theory, one can write [31]:

$$n = \frac{P}{kT} \tag{7}$$

where, P is the vapor pressure, T is the temperature, k is the Boltzmann constant equal to 1.38×10^{-23} J/K.

The arithmetic mean velocity of molecules is determined from Equation (8) [32]:

$$\bar{v} = \sqrt{\frac{8kT}{\pi m_0}} \tag{8}$$

that,

$$M_k = \frac{1}{4}\times\frac{P}{kT}\sqrt{\frac{8kT}{\pi m_0}}\times m_0 \tag{9}$$

or

$$M_k = P\sqrt{\frac{m_0}{2\pi kT}} \tag{10}$$

These formulas are valid for an ideal gas. In the considered temperature range, the mercury vapor density is low, and the vapor can be considered as an ideal gas. If each molecule that hits the surface of the liquid was deposited on it, then Equation (10) would describe the rate of condensation. However, a certain part of the molecules is reflected from the surface of the liquid, and to take into account the degree of reflection, the evaporation coefficient α is introduced, called the Langmuir coefficient. Thus, the specific rate of vapor condensation is:

$$M_k = \alpha P\sqrt{\frac{m_0}{2\pi kT}} \tag{11}$$

The value of the Langmuir coefficient for pure mercury in the liquid phase is approximately equal to unity (α=1) [33]. When there is saturated vapor above the liquid phase, from the equality of the rates of condensation and evaporation, for the specific evaporation rate, one can write (for α=1):

$$M_e = P_1\sqrt{\frac{m_0}{2\pi kT}} \tag{12}$$

where, P_1 is saturation vapor pressure of mercury. Equation (12) can be used to calculate the evaporation time for mercury droplets of various sizes. If the interface between the liquid and vapor phases is not flat, then the curvature of the surface affects the rate of evaporation and condensation processes. This effect can be determined by the change in pressure above the liquid. Saturated vapor pressure at the phase boundary depending on the curvature of the surface is described by the Kelvin equation [33]:

$$\frac{P_r}{P_0} = \exp\left(\frac{2\sigma}{r_0}\times\frac{V_1}{RT}\right) \tag{13}$$

where, P_0, P_r is saturated vapor pressure over flat and curved surfaces, σ is surface tension of the liquid; r_0 is the radius of curvature of the surface; V_1 is the molar volume of the condensed phase; and R is universal gas constant.

The curvature of the liquid surface leads to a change in the equilibrium vapor pressure P_r above it compared to the saturated vapor pressure P_0 above a flat surface at the same temperature. The decrease or increase in vapor pressure depends on the sign of the surface curvature: in the case of convex surfaces (r_0>0) P_r>P_0, and over concave surfaces (r_0 <0) P_r<P_0. It follows from Equation (13) that, near small droplets, the mercury vapor pressure increases; Evaporation from a convex surface is faster than from a flat one. Let there be a hemispherical drop of mercury with mass M, its radius can be determined by formula:

$$r_1 = \sqrt[3]{\frac{3M}{2\pi\rho}} \tag{14}$$

and the evaporation surface can be defined as:

$$S = 2\pi\times r_1^2 = (2\pi)^{\frac{1}{3}}\times\left(\frac{3M}{\rho}\right)^{\frac{2}{3}} \tag{15}$$

Considering that approximately 5 mg of mercury is introduced into the LL, calculations using Equation (14) show that the radius of the mercury drop in the lamp is 0.56 mm, smaller drops can also be present in the lamp, as follows from Equation (13), their evaporation rate is higher than the evaporation rate of large drops. Calculations using Equation (13) show that the increase in pressure and, consequently, the evaporation rate for small drops is noticeable only when their sizes are less than 10^{-4} mm, for large drops, the increase in the evaporation rate due to the curvature of the phase interface is almost imperceptible.

3. RESEARCH RESULTS

3.1. Design of Device

The developed device consists of a reservoir partially filled with liquid mercury for its vaporous dosing into the LL (Figure 1). The device contains an electromagnetic valve, which is placed vertically and coaxially with the release hole, made in the lower part of the reservoir. A mercury heater and a vertical separating branch pipe are introduced into the device, the lower end of which is connected with the release hole. The valve cage is made at the upper end of the branch pipe. The coil of electromagnetic valve wraps around the top of the reservoir. Thermocouple, heater and coil of electromagnetic valve are connected to the control block.

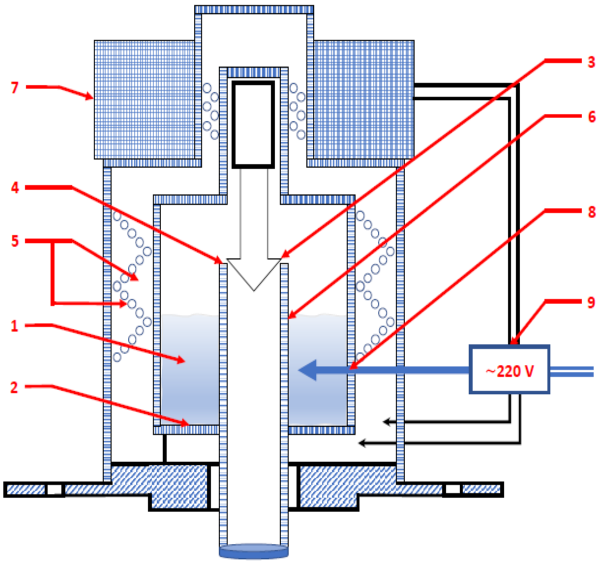

Figure 1. Device for dosing mercury into a lamp, 1. Reservoir for mercury, 2. Release hole, 3. Valve, 4. Cage, 5. Heater, 6. Branch pipe, 7. Electromagnetic coil, 8. Thermocouple, 9. Control block

3.2. The Principle of Operation of the Device

To dose mercury into the lamp by using the heater, the mercury is heated to a temperature set by the control block, for example, up to 250 °C. As a result of heating, mercury evaporates into the reservoir cavity, and the saturated vapor pressure in it increases.

Then voltage is applied to the coil of electromagnetic valve. At the same time, the valve rises out of the cage and the mercury vapor under the influence of the pressure difference (vacuum in the lamp) enters the lamp exhaust tube through the branch pipe. When passing inside the branch pipe, mercury vapor is accelerated by the pressure difference and exits as a beam, which enters the lamp through the exhaust tube. The mass of mercury entering the lamp is controlled by changing the temperature controlled by a thermocouple, as well as the duration of the voltage pulse applied to the coil of electromagnetic valve. If it is necessary to seal the valve, a spring can be additionally introduced into the device.

The reservoir can be filled with mercury by sucking it through the branch pipe while cooling the reservoir with the valve open. The processing of structural elements and operating modes of a device for dosing mercury in a vapor state requires taking into account a number of factors. So, it is desirable that the volume of the reservoir cavity above liquid mercury satisfies the condition:

$$v > \frac{m}{\rho} \tag{16}$$

where, m is the mass of mercury supplied to the lamp; and ρ is the density of mercury. Compliance with this condition provides a relatively short duration of the valve in the open state. It should also be taken into account that with a rapid decrease in the pressure of mercury vapor in the reservoir, they are cooled, which also indicates the need to increase the volume v.

4. CONCLUSION AND RECOMMENDATIONS

Calculations have established that when dosing mercury into the LL in amounts of 5 mg and 50 mg, the radius of mercury droplets is 0.56 mm and 1.2 mm, respectively. For this case, the hemispherical surface area of mercury drops is calculated to be 2 mm^2 and 9 mm^2. When mercury droplets are smaller, their rate of evaporation and condensation is greater than the rate of evaporation and condensation of larger droplets. During the operating mode, after the pressure of saturated mercury vapor has been reached, the lamp operates stably and as the mercury vapor is consumed, further evaporation of liquid droplets occurs in the gas-discharge gap. It follows those small amounts of dosed mercury in the LL simultaneously ensures optimal operation of the lamps.

Based on the work performed, the following conclusions can be drawn:

1) In contrast to devices for the liquid introduction of mercury, when dosing in a vapor state, losses of mercury on the way to the lamp are practically eliminated, since the volume of the channels when administered in a vapor state is tens of times less than when dosing liquid;

2) The device and the mercury supply channels located in close proximity to it are at a higher temperature than the lamp, so condensation of mercury in the lamp will occur more intensely and this promotes the diffusion of mercury vapor from the hotter parts into the colder cavity of the lamp;

3) Calculations of the processes of evaporation and condensation of mercury in low-pressure LL can be used to evaluate and justify methods for measuring the amount of mercury in a lamp, both in laboratory and industrial conditions.

To substantiate the efficiency of the device and the method of vapor dosing of mercury in the LL under special production conditions on a manufactured model, it is necessary to conduct laboratory studies of the modes of supplying mercury to the lamp. Using the data obtained, it is possible to calculate the dependencies of the parameters of pressure, temperature, mass of mercury, and current pulse duration to create a computer program for an automatic control unit of the dosing device.

NOMENCLATURES

1. Acronyms

LL	Luminescent Lamp

2. Symbols / Parameters

N: Number of molecules
n: Concentration
$\bar{v}$: Arithmetic mean velocity of molecules
M_k: Mass of molecules
m_0: Mass of one vapor molecule
P: Vapor pressure
T: Temperature
k: Boltzmann constant
α: Langmuir coefficient
M_e: Specific evaporation rate
P_1: Saturation vapor pressure of mercury
P_0, P_r: Saturated vapor pressures over flat and curved surfaces
σ: Surface tension of the liquid
V_1: Molar volume of the condensed phase
r_0: Radius of curvature of the surface
R: Universal gas constant
r_1: Mercury drop radius
v: Volume of the reservoir cavity above liquid mercury
m: Mass of mercury
ρ: Density of mercury

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