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Transient behaviors for Hg, Tl and Dy emission spectral lines in a halide lamp

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Abstract

Spectral measurements have been performed in order to see the changes in time dependence of the intensity for Hg, Tl and Dy principal emission lines in high intensity discharge plasma of a metal halide lamp. This kind of experiments put in evidence the starting moments of principal spectral events in the positive column of a discharge lamp, that can be correlated with the melting and boiling points of the salts inserted in the lamp enceinte. The time dependence of electron temperature deduced from spectroscopic measurements is also discussed.

Keywords HID lamps, emission spectrum

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1 Introduction

In metal halide (MH) lamps light is generated in a high-pressure mercury discharge to which other light-emitting species are added to improve the spectrum of the lamp. Hg (in small quantities) is used as a buffer gas due to their easy vaporization (boiling point of 356.73^{o}C), low ionization and excitation potential and to a small reactivity with silica [1]. The gaps in the visible spectrum of a mercury discharge are filled up with radiation from other metals, coming from corresponding metal halides. Pure metals cannot be used as additive because most metals have a low-vapor pressure and/or attack the material in which the discharge is contained. The metal iodide or bromide have vapor pressures of the order of Torr's and higher at bulb wall temperatures. As the metal halide diffuses from the wall into the area of the arc discharge the molecules will dissociate, the resulting being free metal atoms and free iodine or bromide. The big difference between the halogen and metal atoms excitation and ionization potentials determines that only excited metal atoms will participate in the operation of the electric discharge [1], yielding a high metal vapor pressure. In the arc discharge the atoms will be excited to higher energy levels and radiation can be emitted. By ionization will be a participation of the corresponding ions to the current through the tube. The metal atoms and iodine (bromide) atoms do not stay exclusively in the higher temperature zones of the lamp, they are coming out of the discharge region and will recombine again to metal halide compounds in the cooler part of the discharge vessel. This cyclic process results in lamps which are more efficient in generating visible light and which have better color rendering properties than mercury

lamps. The basic requirements for chosen metals used as halides in the lamp should be the following: 1) the metal halide must be stable at the operating temperature of the bulb [2]; 2) the vapor pressure of the metal halide needs to be high; 3) the metal atom must have excitation levels lower than for mercury; 4) a significant percentage of its emitted radiation should be in the visible spectral region. With respect to their contribution to the spectrum we have additives that give one or a few dominant lines in the spectrum (sodium - yellow), thallium - green, and indium - blue); additives emitting many lines (rare-earth metals) better covering the visible spectral region; molecular additives with wide emission bands (rare-earth halides).

Although there are some controversial opinions concerning the establishment of a local thermal equilibrium (LTE) in HID lamp we used the LTE hypothesis in our analysis. As known, LTE implies the availability of the Boltzmann law for the temperature dependence of the population of an excited energy level [3]:

If the plasma is in steady state, we have equilibrium between the number of particles leaving an excited energy level and those arriving on this level. Transition between energy levels in a plasma could be determined by a series of phenomena like: atom collisions, leading to the excitation of one species to a higher energy level; collisions of an excited species with another particle, leading to radiationless relaxation; excitation by collision with electrons; de-excitation with the transfer of energy to an electron; excitation of atoms or ions by the absorption of radiation; de-excitation of atoms or ions by spontaneous or stimulated emission [3].

The rate of formation and of disappearance of charged particles and neutrals are equal, and at a given temperature, T, we can state that the number of charged N^{ion} and uncharged N_0 species remains constant through excitation and de-excitation by collisions with neutrals, ions and electrons.

If the self-absorption could be neglected, the intensity of one emission spectral line is depending on concentration of atoms being in the excited state, on Einstein coefficients, A_{iq}, for spontaneous emission, and on the energy of the emitted photon, $h\nu_{iq}$ [3].

The aim of this work is to illustrate that by spectral measurements we can establish the real moments when each of the characteristic emission lines for different atoms (as Hg, Tl, Dy, and others) begins to appear in the positive column of a metal halide lamp. The time dependence of temperature calculated from intensities of Hg lines is also discussed.

2 Experimental results and discussion

A metal halide lamp of 400W (with transparent wall and no outer bulb, distance between electrodes being of 6 mm) has been investigated in our experiment. The time evolution of emission spectra has been recorded with the Ocean Optics HR4000 High-Resolution Spectrometer by using a detector Toshiba TCD 1304 AP linear CCD array in the wavelength range from 390 nm to 900 nm having the optical resolution of 0.02 nm and also with the Ocean Optics Spectrometer S-2000 UV-VIS by using a detector CCD with an Interface ADC500 in the wavelength range from 186.2 nm to 877.47 nm. To calibrate the wavelength of our spectrometer a HG-1 Mercury Argon Calibration Source and a 50 micron fiber have been used.

The spectral power distribution of the mercury discharge depends to a very great extent on the pressure at which it is operating. At low pressures the output is predominantly in the ultra-violet, but as pressure increases, self-absorption here results in the visible green, yellow and violet lines becoming relatively stronger. Above a pressure of

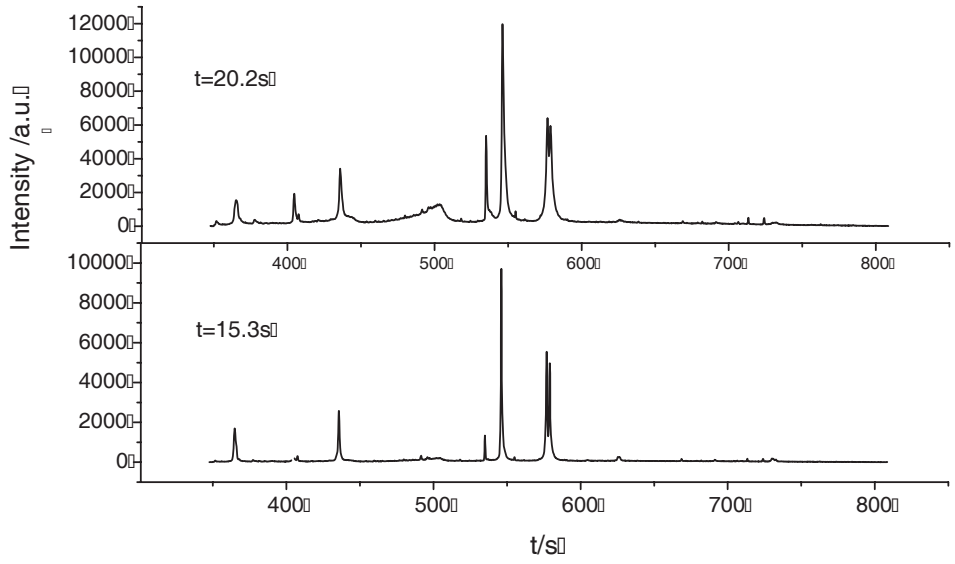

Figure 1: Two emission spectra recorded for a metal halide lamp at various time moments

about one atmosphere, a very small amount of continuum radiation begins to enter the spectrum and this progressively increases in strength as pressure continues to rise.

By analyzing the emission spectrum for the non-irradiated lamp, recorded at different moments of time after ignition, it is observed that from 3.3 s until t=11.6 s only the following strong characteristic emission lines for neutral mercury atoms appear in the case of non-irradiated lamp: 312. 56 nm, 313.13 nm, 313.15 nm, 365.02 nm, 404.65 nm, 435.83 nm, 546.14 nm, 576.96 nm, 578.97 nm. The mercury resonance line of 253.7 nm (from the transition $6^3P_1 \rightarrow 6^1S_0$ [4]) does not appear in the spectrum because it is strongly self-absorbed in the discharge plasma. The spectral lines of Hg ions are absent.

Two emission spectra corresponding to 15.3 s, and 20.2 s (the origin of time being the moment of ignition), are represented in Figure 1 (relative intensities are taken). At t=15.3 s only Hg and Tl emission lines are present in the spectrum. The other spectrum (for t = 20.2 s) also contain more visible emission lines obtained by ionization and excitation process of Dy, Eu, and Na lines.

In figure 2 the time dependences of the 365.12 nm (the 6 ^{3}D$_3$ $\longrightarrow$ 6 ^{3}P$_2$ transition [4]) 404.77 nm (the 7 ^{3}S$_1$ $\longrightarrow$6 ^{3}P$_0$ transition [4]) and 407.9 nm (the 7 ^{1}S$_0$ $\longrightarrow$6 ^{3}P$_1$ transition [4]) mercury emission lines ([5]) recorded in the case of the investigated halide lamp are given.

Another spectral strong line is the green 535.05 nm that appears in the spectrum of non-irradiated lamp after 11.6 s. This is the strongest line of Tl and its presence in the positive plasma column show that the temperature near the wall of the lamp is in the vicinity of the vaporization temperature of TlI (824^{o}C). After vaporization, the molecules of TlI are migrating towards warmer plasma regions and decompose into corresponding Tl metal and I halogen atoms. The other iodides are suffering similar processes. The resulting metal atoms are then available for ionization and excitation process, enriching the output spectrum of the lamp and modifying the electrical characteristics. Then the metal atoms diffuse to the tube wall and recombine with halogen atoms closing the cycle.

It should be mentioned that this spectral band is emitted as the result of the processes taking place mainly in the outer mantle of the arc when the temperature is high enough to excite the molecular states but not too high to cause dissociation. Due to vibrational and rotational effects, lines are broadened to such an extent that a quasi-continuous spectrum is generated.

The time evolution of principal intensities represented in Figures 2 and 3 allow us to determine the moments at which different processes occur in the positive column of the discharge plasma.

In Figure 3 the most prominent intensities versus time for Hg (546.13 nm), Tl (535.09 nm) and Dy (625.94 nm) atoms present in the positive column of discharge plasma are presented. Firstly we observe an increase followed by a drop in all Hg and Tl lines intensities and then by a plateau as the equilibrium in the lamp is reached. This behavior is expected because immediately after ignition, the number of neutral Hg, Tl and Dy and other atoms rises by vaporization. Consequently, time changes in plasma pressure and temperature contribute to a bigger number of atoms being in an excited state. Therefore, this evolution of the spectrum could be related to the rise in the number of electron-mercury collisions. Depending on the melting and vaporization temperature the presence of different atoms in discharge plasma is not observed at the same moment. Firstly, after 3.33 s from ignition the Hg lines become to be present in the emission spectrum as the Hg vaporization temperature is 356.73 $^{\circ}C$. Then Tl atoms (after about 11.6 s from ignition) begin entering in the plasma column (the melting and boiling points for TlI being 442 $^{\circ}C$ and 824 $^{\circ}C$, respectively) leading to a collisional transfer of energy between Hg atoms and Tl atoms. Thus the 535.05 nm Tl line appears in the spectrum while the Hg lines are dimmer. Later after 22.14 s from ignition by collision between Hg or Tl atoms and Dy atoms the red Dy 626.9 nm line enters in the spectrum (the melting and boiling points for for DyI_3 is 978 $^{\circ}C$ and 1320 $^{\circ}C$ respectively) causing a fall of Tl and Hg line intensities.

This lamp is generally used for video projection purpose. In this lamp the iodides introduce also the Na and Eu atoms in the column of discharge plasma. The yellow sodium doublet (589, 589.6 nm) and the sodium red lines 615.4, 616.1 and 619.5 nm, the red line of dysprosium atoms together with many visible lines of Eu atoms make the index color rendering to increase.

Over time, different methods have been developed to determine plasma temperatures inside discharge lamps: atomic emission spectroscopy, X-ray absorption (XRA), Thomson scattering (TS). An overview over this methods is given in ref. [6] and [7]. Comparing these methods Nimalasuriya concludes that LTE hypothesis is satisfactory for the center of the discharge in a HID lamp, while in the outer regions we have departures from equilibrium [7]. In our experiment we have picked our spectra aiming our spectrometer to the center of the lamp where the LTE hypothesis is holding. Considering a Boltzmann distribution, based on the theory of atomic structure and spectra we made rough estimates of the plasma temperature (in the case of LTE approximation) from the relative intensities of emission spectral lines using the formula given in ref. [8].

The time dependences of the electron temperature exhibits Gaussian dependences ($y = y_0 + A(w\sqrt{\pi/2}\exp[-2(x-x_c)^2/w^2]$), that is represented as a solid line in Figure 4.

As expected, the electron temperature in a plasma in both cases is decreasing during the evolution of the discharge until the lamp reaches the steady state (when near LTE is obtained).

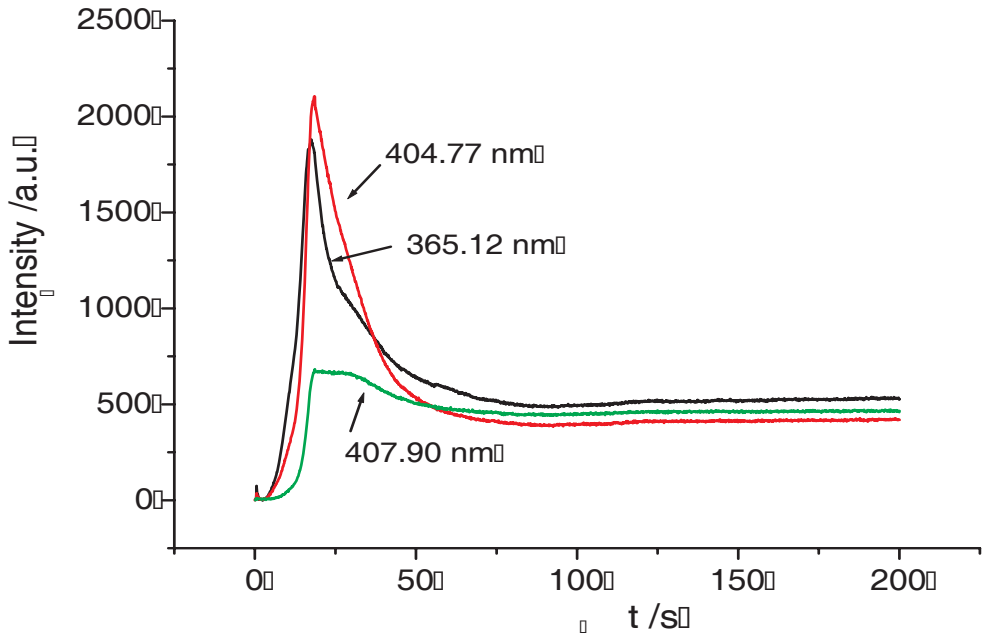

Figure 2: The time evolution of some of Hg lines.

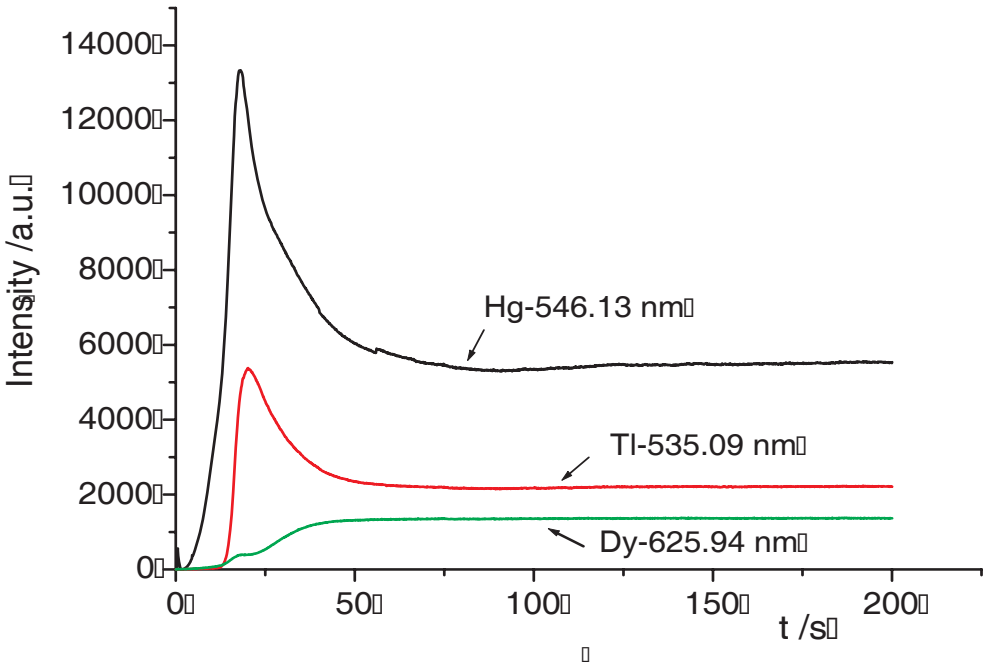

Figure 3: Time evolution for the most intense spectral lines characteristic to Hg, Tl and Dy atoms.

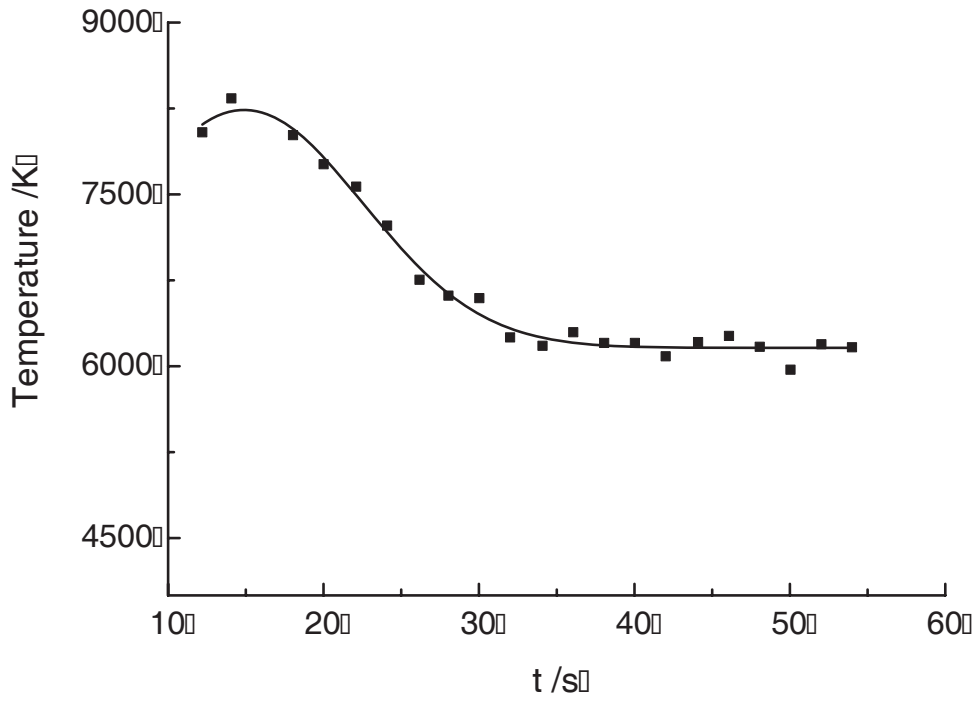

Figure 4: Electron temperature versus time.

3 Conclusions

This paper investigates the time evolution of recorded emission line intensities for a metal halide lamp having Hg as buffer gas and TlI and DyI_3 and other iodides as additive salts. The time evolution of the intensities from emission spectrum allow us to estimate the real moments when the metals from the additive salts enters in the discharge plasma. Those moments can be correlated with the melting and boiling points of the salts inserted into the lamp enceinte. We see also that the electron temperatures are high immediately after ignition and then they are dropping probably as a cause of greater pressure in the lamp which determines a loss of energy for the electrons through collisions.

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