TRANSIENT EMISSION SPECTRA OF A HIGH PRESSURE HG LAMP EXPOSED TO X-RAY

Novac Adrian HARABOR[1], Ana HARABOR[2], Ion M. POPESCU[3]

În acest articol este prezentat un studiu privind modificările spectrelor optice de emisie pentru Hg şi alte componente ale plasmei, pentru o lampă de descărcare cu mercur la presiune înaltă, ce apar prin expunerea la raze X, pentru mai multe puteri de operare ale lămpii. Forma curbei intensității liniei rezonante având lungimea de undă 253.73 nm a Hg în funcție de timp este complet diferită de cele ale altor linii Hg. În regiunea cvasi-stabilă de funcționare a lămpii intensitățile liniilor sunt puternic dependente de puterea de operare, fiind în toate cazuri de iradiere cu raze X (pentru diferiti parametri de funcționare stabiliți pentru sursa de raze X) mai mari decât acelea care corespund lămpii neiradiate. Dependența temperaturii electronice de puterea de operare este descrisă de o funcție de tip Boltzmann. De asemenea, este discutat şi faptul că nu pare să existe nici un efect semnificativ al radiației X asupra temperaturii electronice din plasmă. Este dată şi o explicație calitativă a rezultatelor.

A study on optical emission spectra modifications for Hg and other plasma components in a high-pressure mercury discharge lamp is made at several operating powers before and during X-ray exposure. The shape of 253.73 nm resonant Hg line intensity versus time curve is completely different from those of the other Hg lines. In the quasi-stable region of the lamp its intensity is strongly dependent on the operating lamp power, being in all cases of X-ray irradiation (for various functioning parameters set for X-ray source) above those corresponding to the non-irradiated. The dependence of electron temperature on the operating power is described by a Boltzmann function. The fact that there is no significant X-ray effect on the plasma electron temperature will also be discussed. A qualitative explanation of the results is given.

Keywords: HID lamps; X-ray irradiation; spectrum analysis.

1. Introduction

It is known that different internal and external parameters can influence the emission spectrum of a high intensity discharge (HID) lamp: plasma composition, electrode material and shape, the lamp current, the envelope, internal pressure, external temperature and other factors. For determining these parameters

[1]Asist., Depart. of Physics, University POLITEHNICA of Bucharest, Romania, e-mail: adrian.harabor@yahoo.com
[2] Reader, Fac. of Physics, University of Craiova, Romania
[3] Prof., Depart. of Physics, University "Politehnica" of Bucharest, Romania

several investigation methods have been developed. Relatively new, the XRA is a good diagnostic tool to determine the gas temperature in high pressure discharge lamps and several authors have used this method in their studies (see [1-3]). In this paper we want to see if the X-ray exposure has any influence on the characteristics of the lamps.

In most studies the researchers are assuming that the plasmas in high-pressure lamps (pressures over 10^5 Pa) are in local thermodynamic equilibrium (LTE). In general, this assumption gives satisfactory results in predicting the plasma behavior when the lamp has reached the steady state operation, but poorer results for the transitional state between ignition and full operation.

One of the aims of this work is to illustrate that by transient spectral measurements we can establish with precision the moments of appearance of different Hg emission lines and of principal characteristic emission lines for the emissive mixture components, deposited in the space between tungsten electrode and the wolfram coils. We will discuss the changes that appear in the intensity of some emission lines and in the electron temperature if the lamp is operated at different powers and is under X-ray exposure.

2. Theoretical considerations

Although the local thermal equilibrium (LTE) is not actually attained in HID lamps we can safely use 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 [4]:

$$\frac{N_j}{N} = \frac{g_j e^{-E_j / k_B T}}{Q(T)} \tag{1}$$

where N_j is particle number (concentration) in the excited state, j; N is the total number (concentration) of one species; g_j is the statistical weight for the excitation energy, E_j, of j state; k_B is the Boltzmann constant; T is the temperature; $Q(T) = g_i e^{-E_i / k_B T}$ is the partition function (a parameter that gives the total number of particles that are available for the distribution in different energy states). If the plasma is in steady state, there is 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 radiation-less 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 [4].

The rate of formation and of disappearance of charged particles and neutrals are equal, and at a given temperature, T, then 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:

$$\frac{N^{ion}}{N_0} = \frac{g_i e^{-E_{ion}/k_B T}}{g_0} \tag{2}$$

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, hv_{iq} [4]:

$$I_{iq} = A_{iq} N_{iq} h v_{iq} \tag{3}$$

In Eq. (3) N_{iq} is the number density of atoms in the excited state i and should be substituted using Boltzmann equation, giving the expression:

$$I_{iq} = A_{iq} h v_{iq} N_q \frac{g_i}{Q_q} e^{-E_i/k_B T} \tag{4}$$

where N_q is the total number density of one atom species being in the excited state q, v_{iq} is the frequency of the emitted photon given by measurements; the other parameters like the Einstein coefficients A_{iq}, the statistical weights g_i and the excitation energy E_i are tabulated in literature [5] .

When the intensities I_q and I_p of two emission lines q and p, respectively, generated by the corresponding transitions, are known, the excitation temperature T can be calculated from the expression

$$\frac{5040\left(E_q - E_p\right)}{\left(\ln\left(\frac{g_q A_q}{q_p A_p}\right) - \ln\left(\frac{\lambda_q}{\lambda_p}\right) - \ln\left(\frac{I_q}{I_p}\right)\right)} = T$$

given in ref. [4].

Two phenomena are expected to happen by X-ray irradiation: 1) a real absorption that determines the conversion of the X-ray energy in kinetic energy of the electrons that are created by expulsion from the atoms; 2) a diffusion process characterized by an energy transfer from the incident X-ray incident radiation to the diffused X-ray radiation by the material atoms [6].

For studying the X-ray effect we chose a mercury HID lamp due to the following advantages: a) the high vapor pressure of Hg and the high resistance of mercury plasma giving the possibility to operate the lamp at low currents and high voltages (desirable with respect to lamp efficiency) [2] b) the presence in the steady-state regime of a single dominant plasma component (Hg), which simplifies data interpretation.

3. Experimental results and discussion

In the experiment we used a mercury high pressure discharge lamp with 15 mg Hg having the inner volume of about 2.94 cm^3. After ignition, the atoms in the arc are partially ionized, making them electrically conductive, and a ”plasma” is created. For avoiding the influence of the outer bulb (having also a ”phosphor” coating) over the spectrum we have used the lamp without the outer bulb.

If the plasma is in steady state, there is equilibrium between the number of particles leaving an excited energy level and those arriving on this level. In the case of HID lamps the plasma should be weakly ionized in order to have light emitted by the neutral atoms through processes of de-excitation [7].

The time evolution of emission spectra has been recorded with an Ocean Optics Spectrometer S-2000 UV-VIS by using a detector CCD with an Interface ADC500 in the 186.2 nm to 877.47 nm range. For the wavelength calibration of our spectrometer a HG-1 Mercury Argon Calibration Source that produces lines from 253-922 nm and a 50 μm diameter 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 range, but as pressure increases, self-absorption here results in the visible green range, yellow and violet lines becoming relatively stronger [8].

As seen from the spectrum given in Figure 1, recorded immediately after ignition, for the lamp (without outer bulb) having as nominal power 125 W but operated at 10.23 W, only the following characteristic emission lines for neutral Hg atoms have been observed: 253.73 nm, 365.12 nm, 404.77 nm, 435.96 nm, 546.23 nm, 577.12 nm and 579.23 nm. The highest intensity is that of ultraviolet 253.73 nm emission line, followed by green 546.23 nm and by blue 435.96 nm.

As expected, in the spectrum represented in Fig. 1, the spectral lines of the Ar atoms (belonging to the buffer gas) are also observed at the beginning. An interesting phenomenon is the presence in the spectrum (in the first about 4 seconds) of the characteristic strongest Ba^+ visible emission lines (455.53 nm, 493.54 nm, 614.3 nm and 649.8 nm) surpassing in intensity some emission lines of Hg (312.66 nm, 365.12 nm, 577.12 nm and 579.23 nm).

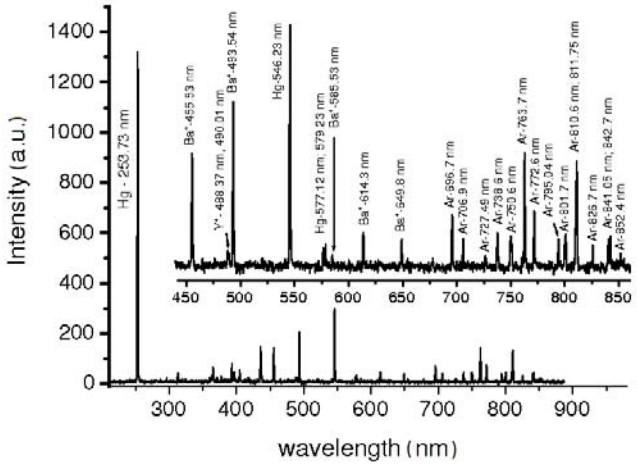

Fig. 1. Spectrum for the non-irradiated HID lamp recorded immediately after the ignition. A more detailed profile for the range from 430 nm to 860 nm is seen in the inset

Three emission spectra corresponding to 0.54 s, 16.44 s and 500.44 s (the origin of time being the moment of ignition), recorded for the non-irradiated lamp operating at 10.23 W are represented in Figure 2. At t=0.54 s the presence of Ba^+ ions, Hg atoms and Ar atom emission lines are observed in the spectrum. As seen, in the spectrum recorded at t=16.44 s the Ba+ ion emission lines are already absent. At t=500.44 s only Hg atoms emission lines remain in the spectrum.

The Ba+ ions present in the plasma arc are coming from the emissive mixture (having as constituents BaO, CaO and Y_2O_3) deposited in the space between tungsten electrode and the wolfram coils around the main electrode. We also detected in the same spectrum the principal lines of Ca^+ (370.6 nm, 393.37 nm and 396.85 nm) and of Y^+ (371.03 nm, 377.43 nm, 378.87 nm and 488.36 nm) but with intensities that are small compared with that of Ba^+ ions, and Ar atoms [9].

The presence of Ba^+, Ca^+ and Y^+ ions is noticed for a very short period of time at the start of discharge as seen from Figure 3, where the relative intensity of Ba^+ ion line having λ=493.54 nm is plotted versus time. This fact is a result of chemical processes taking place at the electrode level as related in ref [9] and [10] one of the effects being the migration of the excess of Ba atoms towards the cathode surface (resulting an extraction work of 2.61 eV, much lower than pure W extraction work - 4.5 eV) and from there into the discharge plasma as ions. Then, as pressure is rising into the discharge vessel, those ions are not getting into the discharge anymore being rapidly reabsorbed by the electrodes.

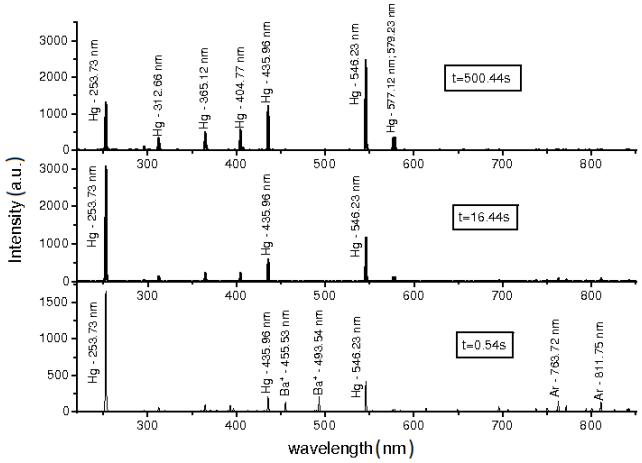

Fig. 2. Emission spectra for the lamp operating at 10.23 W at three moments: 0.54 s, 16.44 sand 500.44 s.

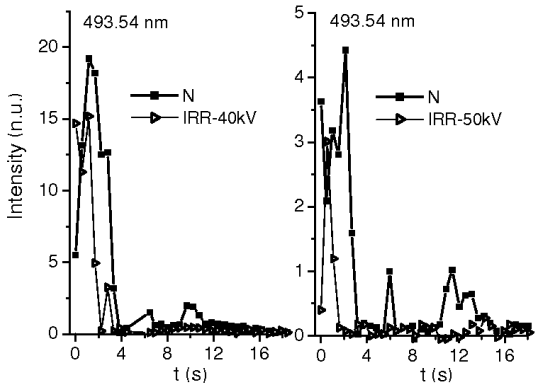

Fig. 3. The time evolution for the intensity of 493.54 nm Ba+ line for normal and irradiated lamp, operating at 10.23 W, when the X-ray source was set at 20 mA and at two acceleration potentials: 40 kV (left) and 50 kV (right).

A Cu-K_α X-ray source (A40-Cu type, maximum input 2 kW) set at a current of 20 mA and at different acceleration potentials (20 kV, 30 kV, 40 kV and 50 kV), has been used to irradiate for 9 min four mercury HID lamps of the same type. Each lamp was placed at around 18 cm from the X-ray source. The X-ray irradiation angle of the XRD tube shield was 12° (no divergence slit was used). The lamp was positioned horizontally and carefully fixed to irradiate the whole discharge tube.

A limited spectrometer time resolution together with processes happening in rapid succession prevented us from finding a clear interpretation of the differences between the transient Ba^+ ions emission line intensities for non-irradiated and irradiated lamp, respectively as seen in Figure 3. A different behavior is observed in Figure 4 where the rate of descend of intensity versus time for Ar 763.7 nm emission line is found to be lower in the case of the irradiated lamp (X-ray source set at 30 kV and 20 mA) than for the non-irradiated one. This means that until about 300 s, during X-ray irradiation, we still have in the discharge plasma a very small percent of Ar atoms being in excited state, justified if we look to the low value of argon K edge energy of 3.207 keV (see Table 1).
The resulting spectral power distribution of high-pressure mercury discharge lamps consists therefore not only of resonance lines but also of spectral lines due to transitions between excited states. In fact, the resonance lines are even missing in high-pressure discharge lamps of sufficiently high pressure, because they are most probably reabsorbed in the outer regions of the plasma in the discharge tube [11]. This is caused by a high density of metal atoms in high-pressure discharge lamps, which is even higher in the colder plasma regions. Since these atoms are mostly in the ground-state it is very likely that the absorption of resonance radiation takes place [2].

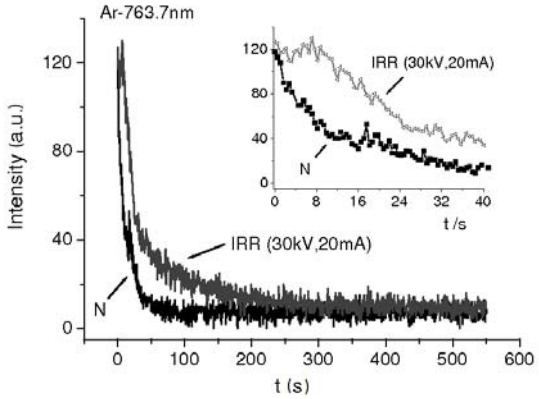

Fig. 4. Time evolution for the intensity of 763.7 nm Ar line in the case of normal and irradiated lamp operating at 10.23 W (X-ray source set at 20 mA and at an acceleration potential of 30 kV).

Tabelul 1

X-ray absorbtion edges for lamp plasma components

Atoms	K edge (keV)	L_1 edge (keV)	L_2 edge (keV)	L_3 edge (keV)
Hg	83.111	14.85	14.22	12.29
Ba	37.45	6	5.62	5.26
Ca	4.04	0.44	0.36	0.36
Y	17.05	2.38	2.15	2.08
Ar	3.21	0.33	0.252	0.25
W	69.53	12.1	11.54	10.2

The above mentioned ideas are confirmed by our experiment as seen in Figure 5 a) (n.u. - denotes the "normal units" obtained considering as maximum the peak corresponding to 546.23 nm). For the lamp operating at lower powers (10.23 W, 22 W) the mercury UV 253.73 nm emission line (resulting from $6\ ^3P_1 \rightarrow 6\ ^1S_0$ transition) is not entirely self-absorbed, but its intensity falls dramatically at higher operating powers (57.64 W, 126.73 W). Looking at the graphs of 253.73 nm Hg resonance line intensity versus time for operating powers of 10.23 W and 22 W (Figure 5 a)) we observe, at first, a sharp peak followed by a plateau when the steady state is attained in the lamp.

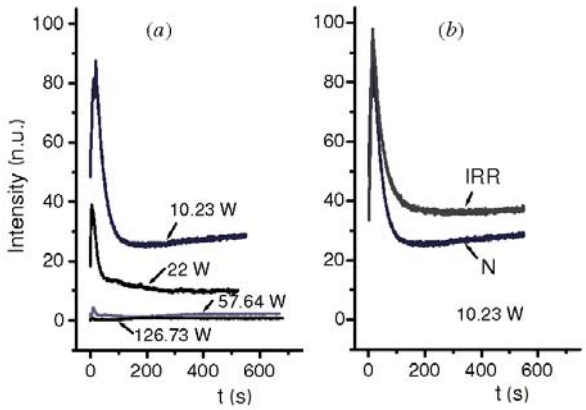

Fig. 5. a) Transient curves for 253.73 nm line intensities recorded at different lamp operating powers; b) Time evolution for the intensity of 253.73 nm Hg resonance line for normal and irradiated lamp operating at 10.23 W when the X-ray source is set at 20 mA and 30 kV.

This behavior is explained by the Hg vaporization immediately after ignition (rising the number of electron-mercury collisions), followed by self-absorption due to the high pressure. For the irradiated lamp (30 kV) Figure 5 b) the curve I(t) is above that corresponding to the non-irradiated one probably due to a higher population for the excited $6\ ^3P_1$ level [9].

All the other Hg lines have almost the same behavior, but different from that of 253.73 nm. As an example, in Figure 6 the yellow 579.23 nm emission line is represented versus time for three values of acceleration potential chosen for the X-ray source: 20 kV, 40 kV and 50 kV.

In Table 1 the energy values for the absorption K, L_1, L_2 and L_3, edges [12] for the elements present in the plasma are given in order to understand the possible effect of the X-ray irradiation using a A40-Cu X-ray source.

Because the absorption K edge for Hg is 83.11 keV, it is practically impossible to expel electrons from K- shell with the photons Cu-$K_{\alpha 1}$ (of 8.048 keV), Cu-$K_{\alpha 2}$ (of 8.028 keV), Cu-K_{β} (of 8.9 keV) characteristic X-ray radiations emitted by X-ray source used in the experiment, nor with the photons more

energetic (20 keV, 30 keV, 40 keV and 50 keV) from the continuum spectrum obtained in the cases of the chosen acceleration potentials (20kV, 30kV, 40kV and 50kV). Nevertheless these more energetic X-ray photons from the continuum X-ray spectrum could expel electrons from L_1, L_2, L_3 shells of Hg atoms as well from the other higher Hg levels.

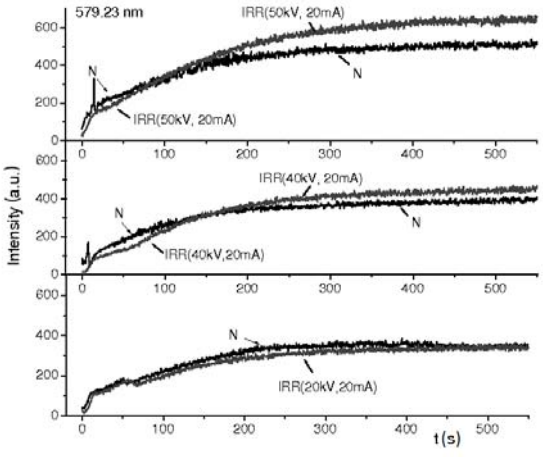

Fig. 6. Time dependence of the 579.23 nm Hg line intensity for the lamp operating at 10.23 W in the case of normal and irradiated lamp, when the X-ray source is set at 20 mA and at different acceleration potentials: 20 kV, 40 kV and 50 kV.

A greater number of the electrons would imply a greater number of the atoms being in excited states and also higher intensities for the emission lines. However in the Figure 6 we can see that there is a region were the intensity of irradiated lamp is under that corresponding to the non-irradiated lamp denoting a smaller population of the excited 6 $^3D_1$. The time interval corresponding to that region is shorter as the acceleration potential is getting bigger. We are assuming that because the lamp functions in a non-equilibrium regime we have fewer neutral Hg atoms in the irradiated than in non-irradiated lamp due to ionization produced directly or indirectly by X-rays [9]. As the acceleration potential is increasing we will have more energetic photons and then more electrons with the energies capable to excite more Hg atoms.

By irradiation with Cu-$K_{\alpha 1}$, Cu-$K_{\alpha 2}$, Cu-K_{β} radiations the electrons seems to be expelled more easily from the absorption K-edge in the case of Ca atoms and from L levels of Ba and Y. When the acceleration potential is 20 kV, the X-ray photons with the energy of 20 keV from the continuum spectrum could expel some electrons from K level of Y atoms and from higher levels of Y and of other elements.

The electrons from K level of Ba atoms could be expelled only by the energetic photons having more than 37.45 keV which are obtained when acceleration potential for the X-ray source was set at 40 kV or 50 kV. The W atoms are in similar situation as Hg atoms. As we suggested in ref. [9] the Auger electrons coming from lamp wall could also enter into the plasma giving their own contribution to the plasma collisions.

Considering a Boltzmann distribution, based on the theory of atomic structure and spectra we estimated the plasma electron temperature (in the case of LTE approximation) from the relative intensities of emission spectral lines using the formula given in ref. [9] and [13].

For a given spectrum, a plot of logarithmic term, $Y = ln(I\lambda^3/gf)$ versus $X = -E_{upper}/k_B$ yields a straight line whose slope is equal to 1/T where I, λ and *f* are the relative intensity, wavelength and oscillator strength respectively; E_{upper} is the energy of the upper level and g is the statistic weight of the lower level; T is the temperature in Kelvin and k_B is the Boltzmann constant. All constants used were taken from ref. [5] and are also given in ref. [9].

Temperature calculations were made for a wide time interval and for several operating powers and we observe that we have small differences between temperatures for non-irradiated and irradiated lamps. Some of the results were published elsewhere [9].

First of all we should notice that the electron temperature exhibits a strong dependence on the lamp operating power (as seen in Figure 7), having a temperature variation from 4135 K to 6124 K (calculated in the steady state regime) for an operating power distributed in the 10.23-126.73 W range. As observed for fitting the data a sigmoidal (boltzmanian) function was used. It seems that the electron temperature has a tendency towards saturation as the power given to the lamp is increased.

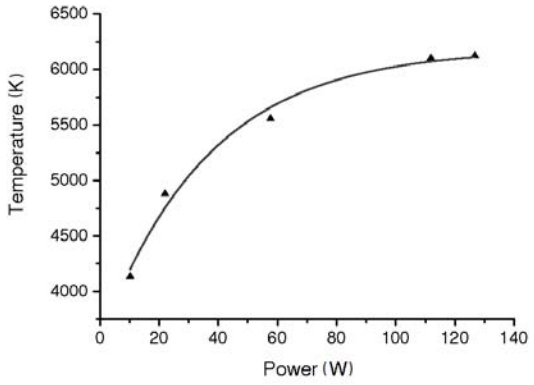

Fig. 7. Electron temperature as function of lamp operating power in steady state regime.

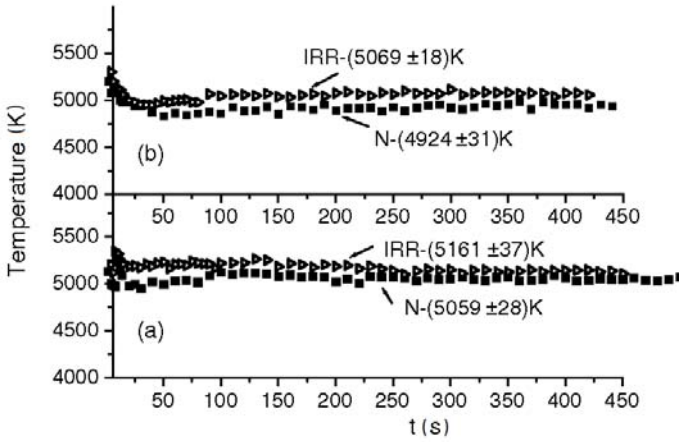

Fig. 8. Electron temperature versus time for normal and irradiated lamp (operated at 22 W) in two cases: (a)-X-ray source set at 40 kV and 20 mA; (b)-X-ray source set at 50 kV and 20 mA.

In this work we analyze the effects of X-ray on the calculated electron temperature over time for the lamp operating at a power of 22 W. The temperatures mentioned in Fig. 8 are those corresponding to 100-450 s range, where thermal equilibrium seems to be realized. Temperature differences between irradiated and non-irradiated lamps are: (a) ΔT = 102 K when the X-ray source was set at 40 kV and 20 mA; (b) ΔT = 145 K when the X-ray source was set at 50 kV and 20 mA. However these differences are smaller than the errors introduced by the calculating method. If the acceleration potential for X-ray sources is set at lower values (30 kV and 20 kV), the differences between temperatures (normal and irradiated) are almost negligible. This shows that the XRA method for determining the plasma temperatures is a reliable one and if we want a better precision it is justified to make a temperature calibration as given in ref. [1].

4. Conclusions

This paper investigates the time variation of recorded emission line intensities for Hg, Ar atoms as well as for Ba^+, Ca^+ and Y^+ ions in a high pressure Hg lamp in the case of non-irradiated and irradiated lamps. Differences in shape and values between recorded Hg line intensities versus time have been observed. These differences are depending on lamp operating power and on acceleration potential set for the X-ray source. A special time evolution is remarked in the case of 253.73 nm Hg resonance line due to the self-absorption whose intensity falls down as the operating lamp power is increased from 22 W to 57.64 W and to higher values. But for the same operating lamp power the intensity of UV mercury resonance line have bigger values for the irradiated lamp than the non-irradiated

one, depending on acceleration potentials chosen for the X-ray source. The effect of X-ray irradiation on the plasma electron temperature evolution over time is also discussed in the case of an operating power of 22W. The differences obtained (for the steady state) are small when the X-ray source is set at 40 kV and 50 kV and are negligible for lower acceleration potentials. A qualitative explanation has been given elsewhere, in the framework of Beer-Lambert, law taking into account the X-ray absorption energy edges characteristic for the atoms present in the plasma [9].

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