Simulation of the radiative spectra for Ar-H_2O-Hg mixtures in high pressure lamps: influence of the presence of H_2O

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Abstract: This work is focused on the influence of water radiation in Ar-H_2O-Hg mixtures used in high pressure lamps. The paper is divided into several parts describing how the radiations issued from atomic continuum and lines, molecular continuum and lines are obtained. Some radiative spectra are presented as a result in order to highlight the influence of water in the plasma, in the UV, visible and IR spectral ranges.

Keywords: Lamps, radiative transfer, spectra, simulation

1. Introduction

Plasma discharge lamps are used for several applications such as domestic or outdoor lighting, or UV radiation used in water sterilization. Some works have already been done on the optimization of the geometry, wall corrosion, electrodes damages, role of the radiation, localization of the heating volumes, stability of the process, lamp voltage, lamps life or efficacity. Usually, rare gases are used such as Ar, Ne, Kr or Xe mixed with metallic vapors or water. Previously, we studied the influence of metallic vapors in ceramic-halide high intensity discharge lamps containing additives in argon plasma, in order to minimize the presence of mercury and increase the visible radiation. Here, the study is focused on the radiation spectra of Ar-Hg mixtures and the influence of the presence of water H_2O on this radiation. In order to better understand the radiation escaping from these high-pressure lamps, it is necessary to solve the Radiative Transfer Equation (RTE) that describes the change of spectral intensity along a path. Several methods can be used such as the method of the Net Emission Coefficient (NEC), the P-1 model or the ray tracing method. In order to do it, the local absorption and/or emission coefficients must be known. Consequently, we developed a numerical code simulating the radiative spectra for a given pressure, several temperatures from 300K to 7000K, different Ar-H_2O-Hg mixtures and spectral interval from 30nm to 4500nm. We assumed the discharge to be at Local Thermodynamic Equilibrium (LTE), and emission/absorption coming from four phenomena: the atomic continuum, the molecular continuum, the spectral lines and the molecular band, all varying with the discharge parameters (size, temperature, pressure or composition). This study will provide an optimised percentage for each required radiation following the application (whether for UV, visible, or IR radiations).

2. Radiative mechanims

The total radiation is the sum of the radiation coming from the free-free, free-bound and bound-bound transitions. Here, we considered e^-, Ar, Ar^+, Ar^{2+}, Ar^{3+}, Hg, Hg_2, Hg^+, Hg^{2+}, Hg^{3+}, H_2, H_2^+, H_2^-, H, H^+, H^-, O_2, O_2, O_2^+, O, O^-, O^+, O^{2+}, O^{3+}, OH, OH_+, OH_-, H_2O, HO_2, and H_2O_2.

Atomic continuum: The atomic continuum is the sum of the radiation coming from the bremsstrahlung (electron-electron and electron-ion), the attachment, and the radiative recombination. The expressions to estimate the corresponding spectral absorption coefficient are given in [1-2]. Here, we just summarized each mechanism. To calculate the radiative recombination, we used the Biberman factors only found for argon species from Hofsaess [3]. For the other species, we applied the hydrogen-like approach [4] using the energy levels, the statistical weights and the quantum numbers given by NIST [5] and Kurucz et Peytremann databases [6-7]. For the attachment, we used the photodetachment cross-sections and the electron affinities of the atoms. As affinity is null for mercury,

we neglected its contribution in the calculation. For oxygen, we used cross-section given by Robinson and Geltman[8] for oxygen, and from McDaniel [9] for hydrogen. Finally, the bremsstrahlung electron-atom and electron-ion were calculated using the total elastic cross-sections and the Gaunt factors, issued from [8,10] or Fursa et al. [11] for e-Hg, specific works from Tanaka and Lowke [12] for e-Ar, [13] for e-O and [14] for e-H.

Molecular continuum: The molecular continuum can be important at low temperatures or when the absorption of the radiation in the surrounding or cold regions of the plasma must be considered. We considered the contribution of the molecules H_2, H_2O, O_2, O_3, OH, and H_2O according to the work of Billoux [15].

Atomic lines: The radiation issued from the atomic lines depends on the lines' shape. We used a line-by-line instead of the "escape factor" approach in order to better represent the variation of the spectra with the mixture. We considered the Doppler broadening and the pressure effects (resonance and Van der Waals broadenings) for the 18454 total atomic lines (11705 lines for Ar to Ar^{3+}, 94 for H, 6217 for O to O^{3+}, and 438 for Hg and Hg^{+}). The line shape of each atomic line was described according to a Voigt profile.

Molecular lines: All the diatomic systems have been treated with a/b Hund coupling. we simulated the molecular lines of H_2 (system Lyman $B^1\Pi \ - X^1\Sigma^+$, and the 3 systems Werner $C^1\Pi \ - X^1\Sigma^+$, $B'^1\Pi \ - X^1\Sigma^+$, $D^1\Pi \ - X^1\Sigma^+$); of O_2 (system Schumann-Runge $B^3\Sigma_u^- - X^3\Sigma_g^-$), of OH (system Meinel $X^2\Pi \ - X^2\Pi$ and Violet $A^2\Sigma^+ - X^2\Pi$).To estimate the radiation of each transition, we determined the emitting population number density from the composition, the line position corresponding to the energy difference between the two rotational levels of the transition, the molecular energies from the effective Hamiltonian formalism and the energy term values for each electronic states, the transition probabilities and the Hönl-London factors (for the molecules OH and H_2 containing hydrogen, the rotational dependence of the vibrational wave functions cannot be neglected and rovibrational transition probabilities have been used). As the atomic lines, the line shape of each line was calculated using a Voigt function taking into account the Doppler and the collisional broadenings (with linear pressure-dependence). For the polyatomic lines, the treatment is more complex so we used the HITEMP 2010 [16] and HITRAN 2012 databases to simulate the radiation of the molecules O_3, H_2O_2 and HO_2 up to 4000K. More details can be found in [15].

3. Influence of H_2O on the radiative spectra

From the last radiative mechanisms, we sum the different contributions to estimate the total absorption coefficient in the total spectral range. With a line by line method, the spectral step between each wavelength was automatically adapted to the temperature, the pressure and the spectral region in order to optimize the time calculation [15]. An example for pure argon plasma is given in figure 1, for 7000K and 6 bar. On this figure, we can see the strong influence of the atomic lines. In figure 2, we present the same calculation done for pure [90]Ar-[10]H_2O at 7000K and the same pressure. We can observe the strong influence of the molecular systems on the radiative spectra. Same results were observed for mixtures Ar-H_2O-Hg.

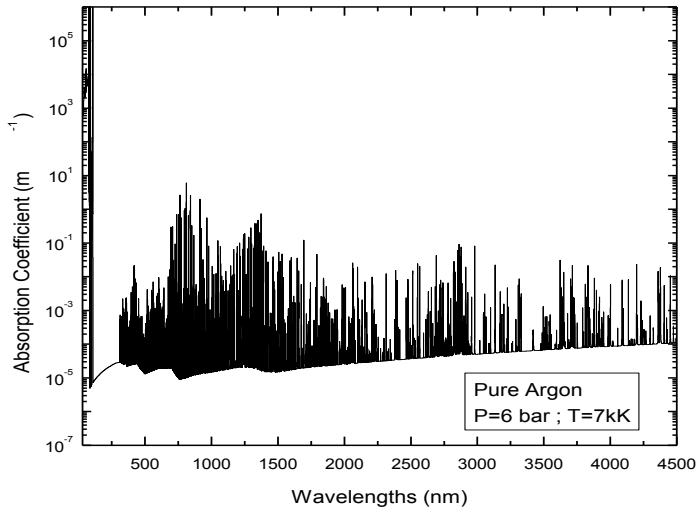

Fig. 1. Spectral absorption coefficient for pure argon at 7000K and 6bar.

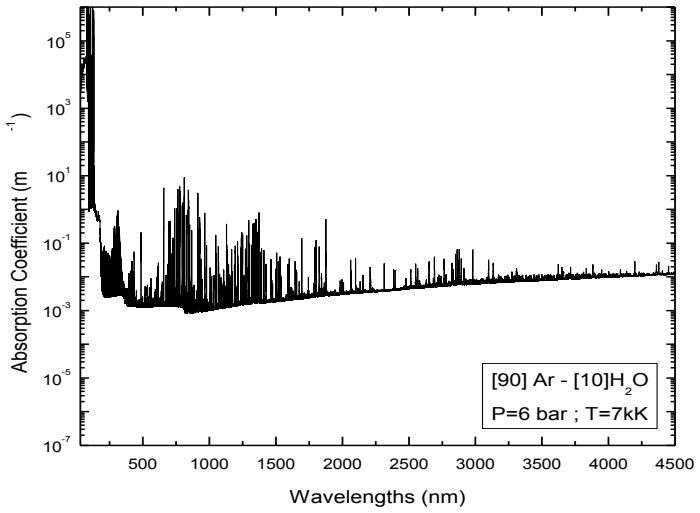

Fig. 2. Spectral absorption coefficient for [90]Ar-[10]H_2O at 7000K and 6bar.

4. Future works

From the total absorption coefficient, the next step is to estimate the divergence of the radiative flux through the method of the net emission coefficient, or to use it in a model for radiation transport calculation based on raytracing method. From a given temperature and pressure profiles, this model can deduce the luminous flux, luminous efficacy, photometric curves, color temperature and chromaticity coordinates.

5. References

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