Radiative energy loss in a two temperature argon plasma

Citation for published version (APA):
Wilbers, A. T. M., Beulens, J. J., & Schram, D. C. (1991). Radiative energy loss in a two temperature argon plasma. In U. Ehlemann, H. G. Lergon, & K. Wiesemann (Eds.), *ISPC 1991: international symposium on plasma chemistry, 10th, Bochum, August 4-9, 1991* (Vol. 1). Article 1.1-4 International Union of Pure and Applied Chemistry.

Document status and date:
Published: 01/01/1991

Document Version:
Publisher’s PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

• A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
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RADIATIVE ENERGY LOSS IN A TWO TEMPERATURE ARGON PLASMA

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ABSTRACT

We have calculated the total radiative loss in an argon plasma at wavelengths from 100 nm to 100 μm as a function of temperature for several pressures under LTE and non–LTE conditions. The investigated non–equilibrium aspects are deviations of the neutral ground state population with respect to the equilibrium population. A difference between heavy particle and electron temperature is included. Almost all influences of pressure and deviations from equilibrium are incorporated in the electron density. Absolute measurements in an inductively–coupled plasma can be simulated with realistic values of the b factor (Saha decrement).

INTRODUCTION

Thermal argon plasmas are used in a variety of applications. For example, inductively–coupled plasmas (ICP) are used for spectrochemical analysis of atoms injected into the ICP. Expanding thermal arcs are used for deposition of carbon– or silicon–based films. To understand these plasmas and predict their properties a great deal of effort has been expended on modeling of the ICPs[1-3] and recently also of expanding arcs.[4-6] In general, ICPs and expanding arcs are similar: the system consists of a flowing thermal plasma which is heated in a channel (either by RF or DC power) and followed by an afterglow. The plasmas have ionizing and recombining regions and deviations from local thermal equilibrium (LTE) are present. Most attempts to model the plasma have been based on the assumption of LTE.[1,2] With this assumption, errors in the temperature profile occur which disturb the energy balance. At least a two–temperature model must be used. The problem which arises now is the dependence of the various contributions to the energy balance on the deviations from LTE. One of these contributions is the total radiative loss, which can be an important energy–transfer mechanism. In this paper, the loss due to radiation is investigated with respect to the influence of deviations from LTE. The condition of partial local thermal equilibrium (PLTE) is assumed to be valid. This implies that the higher levels of the neutral argon system are in equilibrium with the ion ground–level. The condition of PLTE allows for a deviation of the neutral ground–level population from the population calculated according to the Saha equation at a specified electron density and electron temperature. Also, a deviation of the heavy–particle temperature from the electron temperature has been introduced. Radiation to the argon ground level is not taken into account because most of this radiation does not escape the plasma. The contribution of this radiation to the energy balance is taken into account in the ionization–recombination part of the source term in the mass balance of the PLTE model described in Ref. 6. In the present paper, the radiative energy loss due to continuum and line radiation in the wavelength range from 100 nm to 100 μm is calculated. In the literature, the temperature range is limited to values above 10000 K.[7,8] For these temperatures, comparisons between LTE and measurements shows adequate agreement. Miller et al[9] extended their temperature range down to 5000 K but they used a numerical fit of the expression from Ref. 8, which is only valid above 10000 K. Yakubov[10] calculated the radiative energy loss in the temperature range from 6000 to 17000 K with the assumption of LTE. Our calculations have been performed in the temperature range from 3000 to 15000 K. The basic equations are first described. Next, the results of the calculations are given. Finally, a comparison between measured radiative losses[11,12] and our calculations will be discussed.

THEORETICAL

The radiative energy loss is due to continuum emission and line radiation. The continuum emissivity consists of the free–bound and free–free emissivities. The continuum emissivity of argon can be expressed as follows:[13,14]

$$\epsilon = \epsilon_{\mathrm{fb}} + \epsilon^{\mathrm{ei}}_{\mathrm{ff}} + \epsilon^{\mathrm{ea}}_{\mathrm{ff}} \,, \tag{1}$$

with

$$\epsilon^{\mathrm{ei}}_{\mathrm{ff}} = \sum_{\mathrm{z}} \frac{C_1}{\lambda^2}\frac{n_{\mathrm{e}} n_{\mathrm{z}}}{\sqrt{T_{\mathrm{e}}}} z^2 exp\left[-\frac{hc}{\lambda k T_{\mathrm{e}}}\right] \xi_{\mathrm{ff}}(\lambda, T_{\mathrm{e}}, z) \,, \tag{2}$$

$$\epsilon^{\mathrm{ea}}_{\mathrm{ff}} = \frac{\mathrm{C}_2}{\lambda^2} n_{\mathrm{e}} n_{\mathrm{a}} \sqrt{T^3}\, Q(T_{\mathrm{e}}) \left[\left[1+\frac{hc}{\lambda k T_{\mathrm{e}}}\right]^2 + 1\right] exp\left[-\frac{hc}{\lambda k T_{\mathrm{e}}}\right] , \tag{3}$$

and

$$\epsilon_{\mathrm{fb}} = \sum_{\mathrm{z}} \frac{C_1}{\lambda^2}\frac{n_{\mathrm{e}} n_{\mathrm{z}}}{\sqrt{T_{\mathrm{e}}}} z^2 \left[1-exp\left[-\frac{hc}{\lambda k T_{\mathrm{e}}}\right]\right] \frac{g_{\mathrm{z},1}}{U_{\mathrm{z}}} \xi_{\mathrm{fb}}(\lambda, T_{\mathrm{e}}, z) \,. \tag{4}$$

Below 15000 K, the number of doubly–ionized argon atoms is negligible. The density of ionized atoms is then equal to the electron density. More information on the continuum emissivity of argon may be found in Refs. 13 and 14 and the references therein. In order to calculate the total radiative loss, the continuum contributions are integrated from 100 nm to 100 μm. Above 5 μm, the contribution to the total radiative loss is negligible. The line emissivity is given by the following expression:

$$\epsilon_{\mathrm{line}} = \sum_{\mathrm{p}} n_{\mathrm{p}} A_{\mathrm{pq}}\, h\nu \,/4\pi \,. \tag{5}$$

This expression gives the integrated line emission. Because of the negligible doubly– ionized ion density, only neutral argon lines are included. Using Ref. 15, all lines (411) of the neutral argon system have been taken into account. Radiative decay to the ground level has not been included (6 lines). The density of the excited levels is calculated with the Saha equation for a two–temperature plasma:[16]

$$n_\mathrm{p} = n_\mathrm{e} n_\mathrm{i} \frac{g_\mathrm{p}}{2g_\mathrm{i}} \left[\frac{h^2}{2\pi m_\mathrm{e} k T_\mathrm{e}}\right]^{3/2} exp\left[\frac{E_\mathrm{i} - E_\mathrm{p} - \Delta E}{k T_\mathrm{e}}\right] . \qquad (6)$$

In Eqs. (2) and (4), the Bibermann factors from Ref. 13 have been used. Below 6000 K, the Bibermann factors are not known. In the calculations, the Bibermann factors at lower temperatures are taken to be equal to the values at 6000 K. This approximation imposes no grea error because the contribution of the continuum emissivity at these temperatures is negligible compared to the line emissivity. The deviation of the population of the argon ground level from the LTE population is described by the b factor,[17,18] where

$$b = n_\mathrm{a} / n_\mathrm{a,S} \, . \qquad (7)$$

Here, $n_\mathrm{a,S}$ is the argon ground–level population according to the Saha equation at specified electron density and electron temperature. The difference between the heavy particle temperature and the electron temperature is introduced in Dalton's law

$$p = n_\mathrm{e} k T_\mathrm{e} + (n_\mathrm{a} + n_\mathrm{i}) k T_\mathrm{h} . \qquad (8)$$

With Dalton's law, the Saha equation and the external parameters (pressure and electron temperature), the densities are calculated by using a numerical procedure.
Using the b factor and the ratio of $T_\mathrm{h}/T_\mathrm{e}$, deviations from LTE can be easily introduced in order to investigate the influence of these parameters on the total radiative energy loss.

RESULTS

To eliminate the influence of the electron density, the total radiative loss has been divide by the square of the electron density. In Fig. 1, the result is shown for several $T_\mathrm{h}/T_\mathrm{e}$ values. As can be seen from this figure, the differences between the curves decrease strongly and vanish nearly completely above 6000 K. This effect is also observed when a value $b \neq 1$ is introduced, as may be observed in Fig. 2. The b factor is taken to be smaller than 1 (0.1, 0.2, 0.4, 0.6, 0.8, 1), which corresponds to the case of a recombining plasma. When the b factor is larger than 1 (1, 5, 10, 50, 100, 500, 1000) the curves are similar to those of Fig. 1. The large values are most likely to occur in ionizing plasmas. Overpopulation may have large values, in contrast to underpopulation, which can be at most 0. It should be noted that although the calculations extend to 3000 K, an ionizing plasma at these low temperatures is unlikely to exist. The lower temperature limit for an ionizing plasma is about 10000 K. When a b factor smaller than 1 is chosen with an identical value of $T_\mathrm{h}/T_\mathrm{e}$, no significant difference is observed between the variou curves. The pressure range used in the calculations extends from 10 to 100.000 Pa. Again, the influence of pressure is mostly observed in the electron densities. Below 6000 K, the electron–neutral free–free continuum emissivity increases and the curves for the different pressures diverge. From Figs. 1,2 it is clear that the radiative loss divided by the square of the

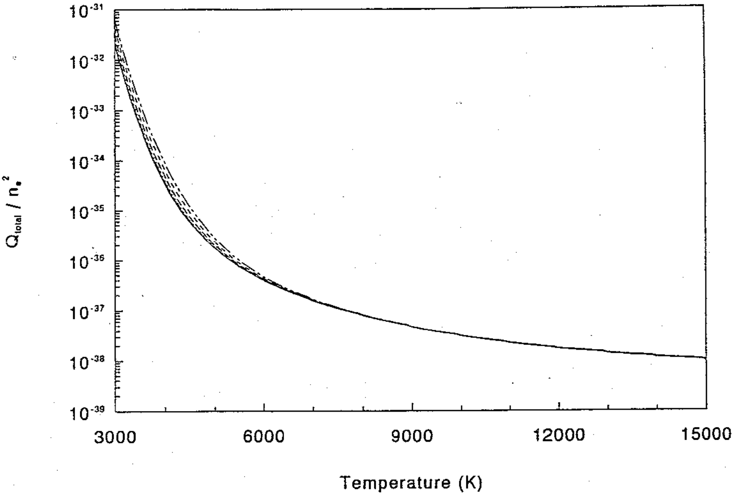

Fig.1. Total radiative loss divided by the square of the electron density as a function of temperature for discrete values of T_h / T_e: —·—, 0.1; · · ·, 0.2; ——— ———, 0.4; – – –, 0.6; —· ·—, 0.8; ——, 1.

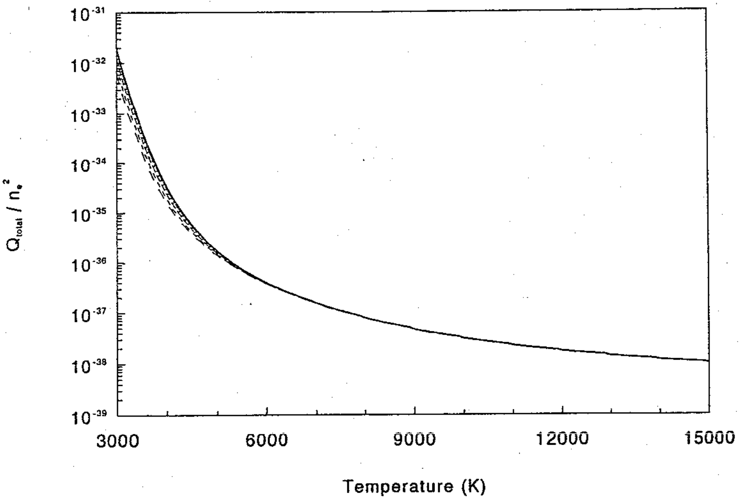

Fig.2. Total radiative loss divided by the square of the electron density as a function of temperature for discrete values of b: —·—, 0.1; · · ·, 0.2; ——— ———, 0.4; – – –, 0.6; —· ·—, 0.8; ——, 1.

electron density can be described in good approximation by one curve. Below 6000 K, differences between the various curves occur but the contribution of the radiative loss to the total energy balance is now not significant.

A numerical fit to the solid curve in Figs. 1,2 will represent a expression for calculation of the radiative loss as a function of temperature that is independent of small deviations from LTE. Finally, some comparison between measured radiative loss and our calculations must be made to verify if the influence of the *b* factor and the difference in temperatures can be represented satisfactorily.

In Fig. 3, measurements and calculations of the radiative loss are shown from various authors.

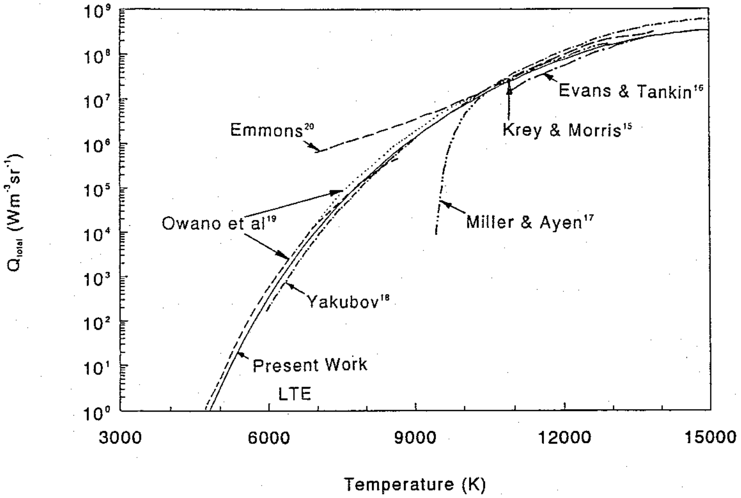

Fig.3. Absolute measurements and calculations. The work of various authors is indicated in the figure. The result with LTE assumption is indicated as a solid line.

Only Yakubov,[15] Owano et al[19] and Emmons[20] extend their values to lower temperatures. The values of Yakubov are calculations with the assumption of LTE. If we use in our calculations *b* equal to 1 and T_h/T_e also equal to 1, the calculated values are higher than those of Yakubov. This result is most likely due to the fact that our spectral range is larger than the spectral range of Yakubov. Owano et al have measured the radiative loss downstream in an ICP, which is called the radiative source strength in their paper. To simulate their values, e.g. at 6000 K, a value for *b* and T_h/T_e of 0.8 has to be assumed. In their paper, Owano et al state that no significant difference in temperatures occurs. In this case, a value for *b* of 0.6 is sufficient to obtain their measured values. This is a realistic value of *b*, as has been shown in Ref. 19. Owano et al made measurements from 250 nm to 2500 nm. The continuum emissivity between 100 nm and 250 nm and above 2500 nm is not taken into account. Their measurements show deviations between the measured values and the upper limits which increase with increasing temperature. This result is probably due to the increasing influence of the continuum emissivity in the omitte wavelength regions, which is absorbed in the path between plasma and the detector. The measurements of Emmons cannot be simulated with realistic values of either the electron densit or electron temperature, irrespective of the degree of non–equilibrium.

CONCLUSIONS

We have calculated the total radiative losses for wavelengths from 100 nm to 100 μm as functions of temperature and pressure and introduced deviations from LTE. The absorption has been taken to be zero in this wavelength region. The introduction of deviations of LTE has been achieved with a *b* factor which accounts for deviations from the neutral ground–state population with respect to LTE densities (PLTE) and a difference between heavy–particle and electron temperatures. If this total radiative loss is divided by the square of the electron density, a curve is obtained which gives the total radiative loss as a function of temperature. The influence of pressure and deviations from LTE on this curve are negligible. A numerical fit is provided which has yielded a simple expression to calculate the total radiative loss. All influences of deviations from LTE are incorporated in the electron density. Absolute measurements in an inductively–coupled plasma can be simulated with realistic values of *b*.

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