Characteristic quantities of a cascade arc used as a light source for spectroscopic techniques

Citation for published version (APA):
Wilbers, A. T. M., Kroesen, G. M. W., Timmermans, C. J., & Schram, D. C. (1990). Characteristic quantities of a cascade arc used as a light source for spectroscopic techniques. *Measurement Science and Technology*, *1*(12), 1326-1332. https://doi.org/10.1088/0957-0233/1/12/012

DOI:
10.1088/0957-0233/1/12/012

Document status and date:
Published: 01/01/1990

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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Meas. Sci. Technol. 1 (1990) 1326–1332. Printed in the UK

Characteristic quantities of a cascade arc used as a light source for spectroscopic techniques

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Received 6 June 1990, in final form and accepted for publication 1 August 1990

Abstract. We have determined the characteristic quantities (the electron temperature and the electron density) of an argon plasma in a cascade arc (diameter of 2 and 4 mm) for a pressure range of 1×10^5–8×10^5 Pa and a current range of 20–70 A. The absolute continuum intensity was also determined in the wavelength ranges from 250–320 nm and 380–800 nm for pressures of 2, 4 and 6 bar and 20, 40 and 60 A in the case of a 2 mm arc. The plasma is close to local thermal equilibrium (LTE). Using the mentioned quantities, prediction of the absolute intensity as a function of the wavelength is possible from 140 nm to the infrared within 10%. The use of the arc as a light source in photon induced chemical vapour deposition, spectroscopic ellipsometry and infrared absorption spectroscopy is discussed.

1. Introduction

Continuum light sources that cover a large spectral range are applied in several analytical techniques. Depending on the nature of the technique the range of interest may cover the UV, visible or IR part of the spectrum. In the following we will focus on spectroscopic ellipsometry, IR absorption spectroscopy and photon induced chemical vapour deposition.

Spectroscopic ellipsometers that operate in the UV-visible range usually apply a xenon short arc as a light source. This kind of high-pressure enveloped arc has a number of features that inhibit its flexible use. The light that is emitted is slightly polarized by the curved quartz envelope. Therefore the dispersive element (usually a monochromator) has to be placed directly after the light source. The alignment is difficult and the complete system has to be operated in the dark. Furthermore, a complex optical system is needed to achieve an intense light beam with a small aperture, as is necessary for ellipsometry. In the near infrared wavelength (from 2–5 μm) region a Globar is used as the light source (Benferhat *et al* 1988). The wavelength limit of 5 μm is due to the relatively low intensity of the Globar in the infrared.

A cascade arc does not have these disadvantages. Since the light exits through flat windows no polarization occurs. Furthermore materials other than quartz (e.g. LiF, BaF_2, MgF_2 etc) can be used as a window in order to expand the spectral range. The aperture of the light beam is small (50 mrad) but the intensity within this small aperture is very large as will be shown below. Because of the large length of the plasma (40 to 60 mm) the sensitivity of the ellipsometer to defocusing effects (dispersion of the material of the several elements of the optical system) reduces substantially. Furthermore, the alignment of the ellipsometer can be done easily by directing a He–Ne laser beam through the plasma channel of the axially symmetric arc. We have implemented at our department the cascade arc in a spectroscopic ellipsometer covering the wavelength regions from 200 to 650 nm and 2.5 to 8.5 μm (Kroesen *et al* 1990a).

For IR absorption spectroscopy, using for instance a Fourier transform infrared spectrometer (FT IR), usually a Globar source is used as the infrared source. The radiating area of the Globar is rather large (several square cm) and its temperature is relatively low (1600 K). The FT IR present at our department used this Globar. With a FT IR it is necessary for the light source to have a small diameter to achieve high spectral resolution (large beam divergence and possible aberrations in the projection have their effect on the apparatus profile). With a cascade arc plasma of 13 500 K and 2.5×10^5 Pa, an increase in brightness by at least a factor of 20–200, depending on the wavelength, has been achieved. This implies an increase of the signal-to-noise ratio by the same factor. The small diameter of the plasma channel (2 mm) and the small aperture also enable a better spatial resolution to be achieved (Haverlag *et al* 1989).

The use of the cascade arc in photon induced CVD gives more understanding of the basic reactions which

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are responsible in the deposition process. Ultraviolet emission of the arc ionizes atoms or molecules. The visible and infrared radiation creates radicals. By blocking the UV radiation the ion concentration is diminished and the effect of the radicals on the deposition process can be investigated.

To optimize the emission of the arc plasma in a certain wavelength region one has to know the characteristics of the cascade arc (e.g. the relation between the external parameters such as current and pressure and electron density and electron temperature).

In this paper we will focus our attention on the properties of the arc. Also some remarks will be made concerning the use of this arc for the mentioned spectroscopic techniques. The determination of the characteristic quantities from the absolute continuum is described in detail in a separate paper concerning the continuum emission of this arc plasma (Wilbers *et al* 1990a). As is shown in that paper it is sufficient to determine the electron temperature and the electron density from the absolute continuum at 400 nm. These quantities can then be used to calculate the emission in the other wavelength regions. We have checked this procedure in the paper mentioned above. Therefore we calculated the electron density from the absolute continuum at three well documented wavelengths (two experimental values of the Biberman factor: measurements done with two-wavelength interferometry and the source method (Rosado *et al* 1979 at 696.5 nm, Timmermans 1990 and Timmermans *et al* 1982 at 468.8 nm and one theoretical, Hofsaess 1978). It showed that these values of the electron densities were the same within 5%.

In section 2 we will discuss briefly the emission theory of a thermal arc plasma. Section 3 treats the cascade arc in detail. In section 4 the results and conclusions will be given about the use of the cascade arc in general and particularly in the mentioned spectroscopic techniques.

2. The continuum emission of a thermal arc plasma

Thermal plasmas in the density and temperature range of our experiments are well described by theory. Quantities such as electron density, electron temperature and line or continuum emission can be calculated with high precision.

At high electron densities ($>10^{22}$ m^{-3}) and electron temperatures ($>10\,000$ K) the line emission which is mainly present at wavelengths above 700 nm has no significant contribution to the total emitted intensity. At higher pressures this contribution further decreases. Our cascade arc discharge has an electron temperature of approximately 13 500 K and electron densities of a few times 10^{23} m^{-3}. Therefore, we will concentrate on the continuum emission.

The density of doubly ionized atoms is negligible under the mentioned conditions. The contribution of electron–neutral interaction to the free–free emissivity is also very small (at high pressures the contribution of this interaction becomes increasingly important. At 30 bar and moderate temperatures the contribution can rise to 50%; the amount of this contribution should then be allowed for). So the continuum radiation originating from the interaction between singly ionized atoms and electrons is dominant in the observed temperature, wavelength and density ranges.

The continuum emission of the discharge in the mentioned ranges of temperature and density is governed by the following expressions (Cabannes and Chapelle 1971)

$$\varepsilon = \varepsilon_{\mathrm{fb}} + \varepsilon_{\mathrm{ff}}^{\mathrm{ei}} \tag{1}$$

where

$$\varepsilon_{\mathrm{ff}}^{\mathrm{ei}} = \frac{C_1}{\lambda^2}\frac{n_{\mathrm{e}}^2}{\sqrt{T_{\mathrm{e}}}}\exp\left(-\frac{hc}{\lambda k T_{\mathrm{e}}}\right)\xi_{\mathrm{ff}}(\lambda, T_{\mathrm{e}}, 1) \tag{2}$$

and

$$\varepsilon_{\mathrm{fb}} = \frac{C_1}{\lambda^2}\frac{n_{\mathrm{e}}^2}{\sqrt{T_{\mathrm{e}}}}\left[1-\exp\left(-\frac{hc}{\lambda k T_{\mathrm{e}}}\right)\right]\frac{g_{1,1}}{U_1}\xi_{\mathrm{fb}}(\lambda, T_{\mathrm{e}}, 1) \tag{3}$$

where $\varepsilon_{\mathrm{ff}}^{\mathrm{ei}}$ is the free–free radiation caused by electron–ion interaction, $\varepsilon_{\mathrm{fb}}$ the free–bound radiation, λ the wave length of the radiation, C_1 the *ei* continuum constant (1.63×10^{-43} W m^4 K$^{1/2}$ sr^{-1}), n_{e} the electron density, T_{e} the electron temperature, ξ_{ff} the Biberman factor for free–free radiation (Hofsaess 1982), ξ_{fb} the Biberman factor for free–bound radiation (Hofsaess 1978, 1982, Behringer and Thoma 1976, Wilbers *et al* 1990a), U_1 is the partition function (Drawin and Felenbok 1965) and c, k and h have their usual meanings. The Biberman factor for free–free radiation is fairly constant and can be taken to be 1.23. The Biberman factor for free–bound radiation can be taken from figure 1 (Wilbers *et al* 1990a) in the wavelength range from 250 nm to 800 nm. The curves in figure 1 are the theoretical values as calculated by Hofsaess. It is clear that below 350 nm his values or the measured ones can be used. Below 250 nm down to 140 nm experimental values as measured by Behringer and Thoma (1976) agree well with those calculated by Hofsaess.

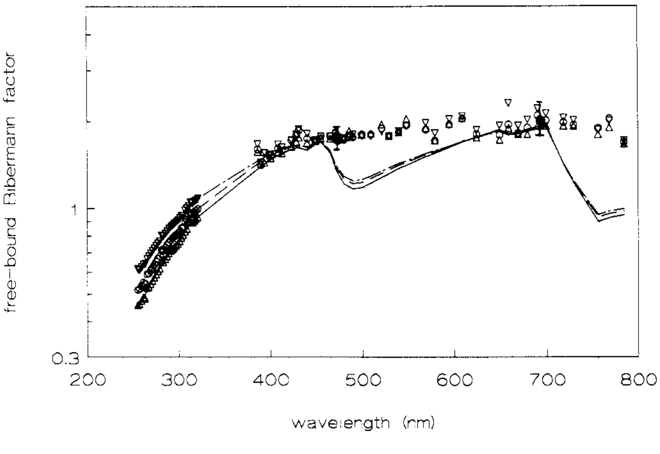

Figure 1. Free-bound Biberman factors: curves, Hofsaess; points, Wilbers. ——, △, 12 000 K; – – –, ○, 13 500 K; –·–·–, ▽, 14 500 K.

Allowing for the effect of absorption, the intensity emitted by the cascade arc is described by

$$I = S[1 - \exp(-\kappa l)] = S[1 - \exp(-\varepsilon l/S)] \qquad (4)$$

where S is given by Planck's formula

$$S_\lambda = \frac{2hc^2}{\lambda^5}\left[\exp\left(\frac{hc}{\lambda k T_e}\right) - 1\right]^{-1} \qquad (5)$$

where κ is the absorption and l the length of the discharge.

From the measurements of the absolute continuum we obtain an absolute value for I_{meas}. With relation (4) we can calculate the absolute total emissivity $\varepsilon_{\mathrm{meas}}$. If we assume LTE conditions we can express n_a, n_1 and n_2 in terms of n_e with the help of the Saha equation, quasineutrality and Dalton's law. The electron density and electron temperature can now be calculated from equation (1), the absolute emissivity and the absolute pressure. A small deviation of equilibrium (partial LTE) causes an overpopulation of the ground level. This will enhance the contribution of brehmsstrahlung due to interaction between neutrals and electrons. Under the present conditions however, this contribution is small and a deviation from equilibrium has no significant effect. The calculated electron density and electron temperature match the LTE values within 5%.

The continuum intensity depends on the square of the electron density and on the electron temperature by an exponential factor (and a square root). As for the electron density, its influence is the same over the entire wavelength region where the arc is not within the Planck limit. An increase in pressure results in an increase in intensity as long as the temperature is constant. The temperature dependence is large in the UV (the exponential factor sharply decreases with decreasing temperature). In the visible the exponential factor is almost equal to one and the temperature has no significant influence. Towards the infrared the intensity approaches the Planck limit. When this limit is reached the intensity is determined by the temperature. In the experimental environment, a compromise has to be chosen between a high pressure and a high current. With increasing pressure the temperature decreases (constant current).

2.1. The cascade arc

The cascade arc consists of three major sections: a cathode section, an anode section and a plate section in between. The plate section, which holds five copper plates stacked into a cascade, gives the cascade arc its name. The whole system has a cylindrical geometry with the plasma channel located on the symmetry axis. A schematic drawing of the arc is given in figure 2. All sections are water cooled. The sections and the plates are electrically insulated from one another by PVC spacers. Boron nitrite discs protect the vacuum sealings from damage caused by radiation emitted in the plasma channel. The cathode section contains three cathodes with sharp pointed, thoriated tungsten tips. These tips, which are the only components that show significant wear, can be easily replaced. They are screwed into the cathode shafts which can be taken out of the arc assembly and put back in just a few minutes. The tungsten thorium tips are made from commercially available welding tools. The lifetime of the arc is at least 1000 h burn time. The arc assembly without the cathode tips should last even longer.

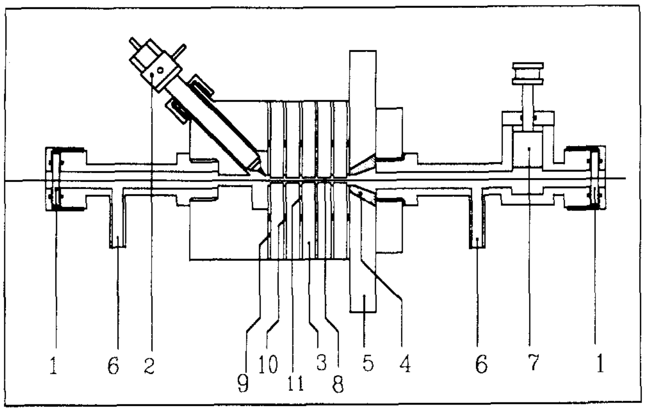

Figure 2. The cascade arc. 1, window (LiF); 2, cathode (3 ×); 3, cascade plates; 4, anode plate; 5, anode insert; 6, gas inlet and outlet; 7, shutter; 8, plasma/gas channel; 9, PVC spacer; 10, O ring; 11, boron nitrite disk.

To allow for optical alignment the cathode tips, as well as the anode spot, are not in the view line of the arc when observed end on. The anode section holds the conical anode insert. The arc is operated in argon. A small flow (refreshing the arc volume about twice per second) is used to prevent gradual contamination of the arc plasma by cathode and wall material. A schematic view of the gas system is presented in figure 3.

The arc is ignited at reduced pressure (10^4 Pa) using a high-voltage power supply (1200 V), designed and built at our institute. Within 1 s after ignition the arc is pressurized up to 1×10^5 Pa. During ignition and pressurizing the exit window has to be protected from damage caused by the hot gas which leaves the arc. This is done with a brass shutter. When the desired pressure has been reached the shutter can be withdrawn.

The windows have a diameter of 25 mm and are between 2 and 5 mm thick, depending on the material and the operating pressure. For instance, a LiF window needs to be thicker than a quartz window because it is more fragile.

The water cooling of the arc is done with normal water pressure (approximately 4 l min^{-1}). No high water pressure is needed. The electrical system and pump are put in one cabinet which is movable (or two cabinets with the heavy set-up of three 10 kW power supplies). The whole arc set-up is therefore mobile. This enables

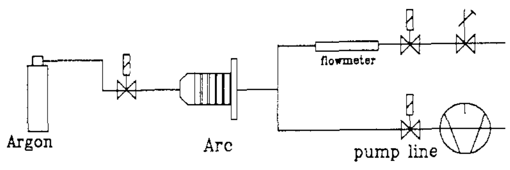

Figure 3. Schematic view of the gas system. The pumping line is only activated during start-up.

Table 1. Typical operational conditions and dimensions of the cascade arc.

Variables	
Arc current	20–70 A
Anode–cathode Voltage	85–120 V
Pressure	1–10 bar
Plate thickness	5 mm
Number of plates	5/6
Space between the plates	1 mm
Diameter anode insert hole	6 mm
Length of plasma channel	42 mm
Channel diameter	2 mm
Argon flow	0.5 scc s^{-1}

the flexibility of the arc to be used in several experiments.

The arc can be fed with either one power supply or three (one for each cathode). The total power is between 5 and 30 kW depending on the desired use. With one power supply care has to be taken that the current is divided equally among the three cathodes. This is done with three series resistors (in our case 3 Ω) put in each cathode power line. These resistors also pull the arc through its negative current–voltage behaviour during start-up. When an arc ignites the resistance is negative. A power supply cannot handle a negative load. Therefore a positive resistance has to be put in the power lines. In our case each of the cathodes is fed with a separate power supply (Electronic Measurements TCR250T40). More details can be found in a technical report on the construction of a cascade arc (Kroesen *et al* 1990b).

Small instabilities can occur at higher pressures or currents. Also, high-frequency noise (several kHz) can be generated between the plasma and the power supply (this depends on the kind of power supply and the length of the cables between the supply and the arc). These problems can be solved by including an inductance in the power lines between the power supply and the arc. The inductance is in the order of several mH. Also small variation of the flow (increase or decrease) can stabilize the arc. In table 1 the typical operating conditions and arc dimensions are given.

3. Optical system

The optical system used to measure the absolute continuum intensity is shown in figure 4. Two equivalent systems are used, one in the vacuum UV and one in the visible part of the spectrum. With lens 1 (quartz) and diaphragms 1 and 2 a small plasma volume (cylinder of 0.5 mm diameter) is selected. When mirror 1 is rotated a similar surface on a tungsten strip lamp can be selected. Lens 2 (quartz) increases the opening angle to the optimum required by the 1 m Jarrel Ash Monochromator. This set-up was used in the visible wavelength region from 380–800 nm. The lower wavelength region (115–320 nm) is measured in vacuum with LiF windows and lenses in combination with a Seya–Namioka 0.5 m McPherson monochromator. Because of the extreme intensity difference between the calibration lamp and the arc (2 to 5 orders of magnitude, depending on the wavelength region: 5 in the UV and 2 in the IR) a grey filter F is used when studying the cascaded arc in the visible wavelength region.

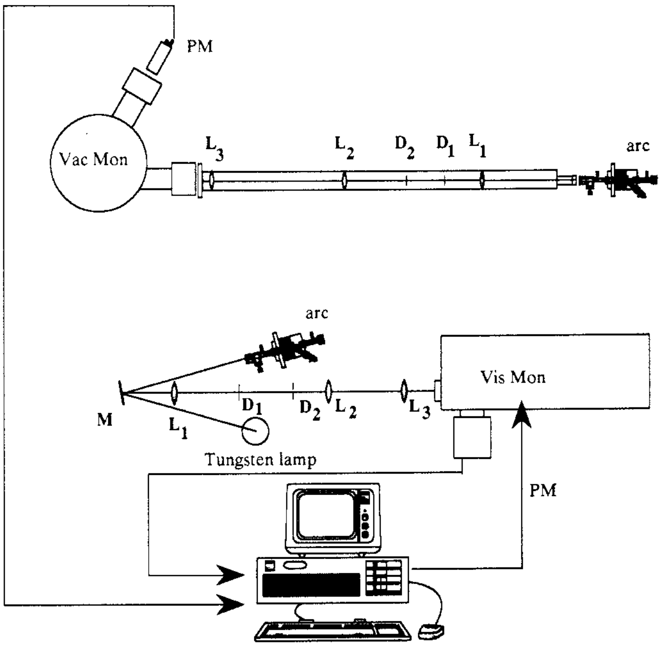

Figure 4. The diagnostic set-up. The wavelength region from 380 to 800 nm is handled by the visible monochromator (Vis Mon) system containing the monochromator, 3 quartz lenses and two diaphragms. From 250 to 320 nm the vacuum monochromator system is used, which is basically the same as the visible system. The calibration lamp (tungsten ribbon, W) is coupled into the optical system with mirror 1 (visible region) or put in place of the arc (vacuum region). The data are collected and processed with an IBM-compatible computer system.

The current pulses which the photons induce in the photomultipliers are counted after being amplified, discriminated to pulse height and converted to TTL pulses (photon counting). The signal treatment takes place as near as possible to the anode of the photomultiplier to eliminate noise. In this way we can cover three orders of magnitude in intensity without losing linearity (low intensity of the calibration lamp and high intensity of the cascaded arc). The TTL pulses are fed into an interface card mounted in a personal computer. The absolute calibration is done with a tungsten ribbon lamp operated at a true temperature of 2667 K.

4. Results and discussions

Properties of the cascade arc such as the electron density, electron temperature and the absolute intensity as a function of external parameters (pressure, current and flow) are considered.

In figure 5 the calculated intensity of the cascade arc is plotted for two different pressures. Also a 1000 W xenon short arc and a Globar of 1600 K are shown to compare the arc intensity with the commercially available light sources. The brightness of the arc is much higher than the other sources. It has to be noted however, that the total intensity achieved with a commercial xenon arc is of the same order due to the large opening angle (more than 1 srad) captured with a complex optical system (collimator, lenses). Our measurements presented here cover wavelength ranges of 250 to 320 nm and 380 to 800 nm. An extension down to 115 nm (LiF window limit) is one of our future options. In figure 6(*a*) and (*b*) the absolute intensity is shown for the arc conditions of 4×10^5 Pa and 40 A. As can be observed from figure 6(*b*) line radiation is present around 420 nm and above 700 nm. From the absolute continuum at 400 nm the electron density and the electron temperature are calculated.

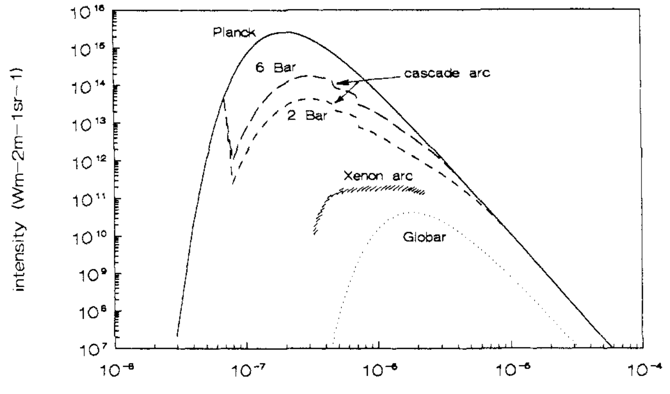

Figure 5. The calculated intensity of a cascade arc with pressures of 2×10^5 Pa and 6×10^5 Pa and a temperature of 14 500 K. Also the blackbody (Planck) radiation of 14 500 K, a 1000 W xenon short arc and a 1600 K Globar are given.

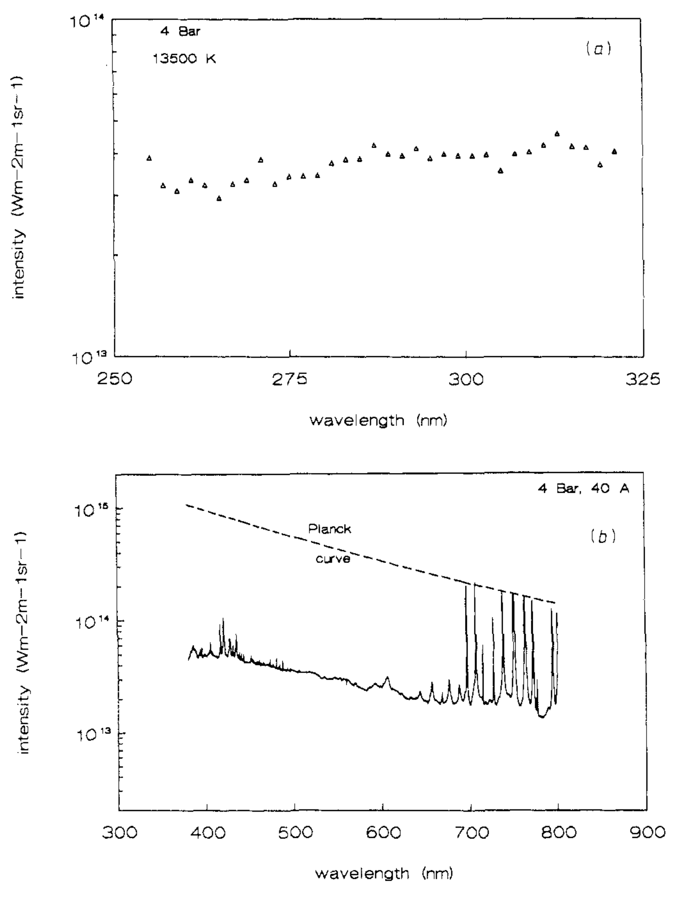

Figure 6. (*a*) The measured absolute intensity of a cascade arc between 250 and 320 nm for 4×10^5 Pa and 40 A. (*b*) The measured absolute intensity of a cascade arc between 380 and 800 nm for 4×10^5 Pa and 40 A. The electron temperature is 13 500 K.

In figure 7 the electron density and electron temperature are shown for the covered pressure and current ranges. As can be seen from the figure, the temperature increases as the pressure decreases. The desired n_e, T_e combination can be linked from this figure to the external parameters, pressure and current. To enhance UV radiation, for instance, electron temperature and electron density have to be as high as possible; on the other hand in the infrared n_e and T_e have to be chosen in such a way that the Planck limit is approached as closely as possible and at a temperature as high as possible. (Both are finally limited by the power supply and the cooling capabilities of the arc itself.)

The effect of the length of the arc on the intensity is wavelength dependent. As long as the plasma has not reached the Planck limit, as will occur in the visible and even more so in the UV, a longer arc emits more light. With increasing pressure the minimum wavelength at which the plasma has reached the Planck limit will decrease. For instance an increase of pressure from 1×10^5 to 5×10^5 Pa reduces this minimum wavelength

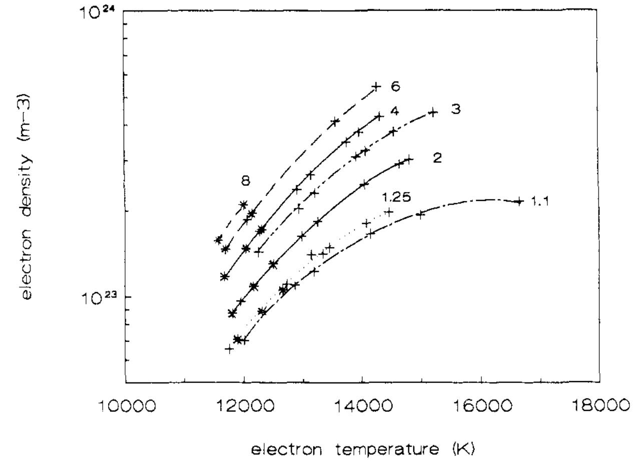

Figure 7. Electron densities as a function of electron temperature for different pressures and currents. The values of the pressure are indicated in 10^5 Pa on the figure. Two diameters of the arc are used: +, 2 mm; *, 4 mm. The lowest temperatures along the lines of equal pressure have been reached with a current of 20 A. The highest temperatures with a current of 60 A. The temperature of 16 500 K at 1.1×10^5 Pa has been achieved with a current of 70 A. The values at 8×10^5 Pa are measured in an arc of 4 mm diameter and currents of 20 and 40 A.

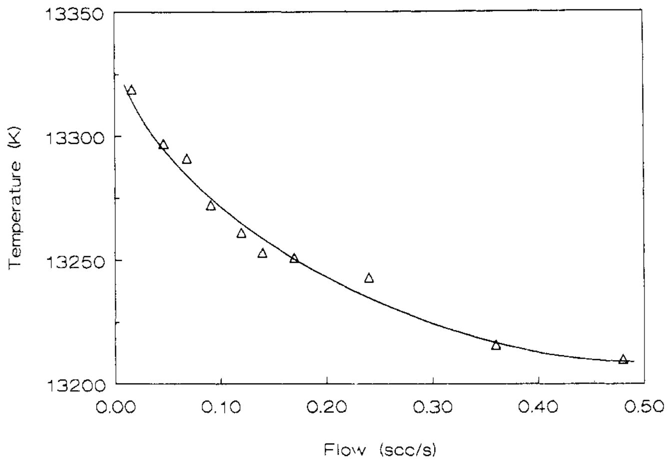

Figure 8. The electron temperature as a function of the gas flow through the arc.

limit from approximately 10 to 4.5 μm. Then the necessity for a longer arc vanishes and a power supply can be used with a lower voltage.

The influence of the gas flow on the electron temperature is shown in figure 8. As the flow increases the temperature decreases and therefore also does the intensity. The flow has to be chosen such as to refresh the arc volume once or twice a second and to keep the intensity decrease low.

5. Conclusions

If the arc is used as an absolute secondary reference (or else knowledge about absolute intensities is desired), the electron density and the electron temperature have to be known. When the arc is used merely as a light source an absolute calibration is not necessary. The electron density and electron temperature can be calculated from the absolute continuum at, for instance, 400 nm (Wilbers 1990a). With the so obtained electron density and electron temperature the emission of the arc can be calculated from 140 nm to the near infrared. If our arc set-up is used the electron density and electron temperature can be taken from figure 7.

For practical use as a light source the cascade arc has already proven to be a good choice. Klose *et al* (1987) used cascade arcs as secondary radiation standards in the VUV. In our spectroscopic ellipsometer an extension to 8.5 μm has been achieved and with a change of a few optical components the wavelength region from 200 nm to 8.5 μm can be covered. Measurements have been performed on thin films of amorphous carbon, amorphous silicon and carbon based polymers (Beulens *et al* 1989, 1990, Wilbers *et al* 1990b). In the IR interferometer used by Haverlag *et al* (1989) at the same institute, an improvement of a factor of 20–200 (wavelength dependent) in signal-to-noise ratio is obtained. A further improvement is in progress and an extension of the infrared wavelength limit to 12 μm is being prepared.

Compared with tunable lasers the cascade arc has several advantages. Tunable lasers need about ten different dyes to cover the wavelength range from 400 to 900 nm. With frequency doublers the ranges can be extended to 270 nm. Often different pump lasers are needed (Ar^+, Kr^+) to achieve maximum power at the peak wavelengths of the dyes. The arc has a larger wavelength range (140 nm to 12 μm). For infrared spectroscopy lasers cannot be used. Also, lasers are much more difficult to align and changing dyes is time consuming.

Acknowledgments

The authors would like to thank R van Haren, M Dirkx and R de Rooij for taking part in the measurements. We also thank M J F van de Sande who has done most of the construction work of the arc.

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