THE VUV EMISSIVITY OF A HIGH-PRESSURE CASCADE ARGON ARC FROM 125 TO 200 nm

A. T. M. WILBERS and D. C. SCHRAM
Physics Department, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands

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Abstract—We have investigated the vacuum ultraviolet (VUV) emissivity of a cascade arc in argon from 125 to 200 nm. The temperature and pressure dependences of this emissivity are the subject of this paper. Enhancement of the emissivity has been obtained by adding nitrogen to the argon. In some wavelength ranges, the increase of the emissivity due to broadened lines is more than a factor of 10 in a spectral range of several nanometers. These lines reach the blackbody limit, corresponding to the electron temperature of the plasma inside the arc. The electron temperature has been varied between 12,200 and 14,500 K. The relative temperature dependence of the free-bound Biberman factor as a function of wavelength in this temperature range has been determined from continuum measurements and compared to the theoretical temperature dependence of the free-bound Biberman factors according to Hofsaess. The agreement is good.

INTRODUCTION
Vacuum ultraviolet (VUV) radiation from 50 to 250 nm has been the subject for fundamental research and has also been used in several applications. Studies on radiation standards have been performed. If the radiation source is enclosed, the lower wavelength limit is usually determined by the LiF transmission limit at 115 nm. The maximum photon energy is then limited to 10 eV (124 nm). The principal interest in this type of radiation derives from the fact that the photon energy above the LiF window-transmission limit is sufficient to photo-ionize and photo-dissociate molecules into fragments. Also, catalytic effects may occur. The selection of a spectral range is also a selection of the kind of particles which can be created: ions, fragments of molecules or radicals. In this way, the VUV radiation can be used in order to increase our understanding of the processes which take place in deposition and etching experiments.

However, it is difficult to achieve a continuous source of radiation in this spectral range. There are a number of traditional sources available, which all have both advantages and disadvantages. Discharge lamps such as the deuterium lamp or xenon lamp are easy to handle and small. The intensity increases towards a broad maximum at 190 nm. However, it is difficult to calibrate the lamp since the discharge always involves different spots on the anode and cathode. Also, the aging characteristics are not well known and repeated calibrations of the absolute intensity are necessary. If high-grade quartz envelopes are used instead of MgF₂ with these lamps, radiation below 160 nm is blocked. A much stronger radiator of VUV emissivity is the hydrogen cascade arc. The increase in brightness is at least a factor of 100 over the entire range. A disadvantage of the hydrogen arc is the high power which is needed to run the arc. Another candidate is the same arc but run with argon gas. The advantage of using argon as the discharge gas is the much lower power consumption of the arc and also the longer lifetime. A disadvantage is the somewhat lower emissivity. Comparisons of these sources in terms of radiation standards have been done extensively elsewhere.

In this paper, we will focus on the temperature and pressure dependences of the VUV emissivity of an argon cascade arc and the effect of additional gases on the VUV emissivity. The emissivity of the arc in the near VUV and visible wavelength range has been investigated in another paper. One conclusion stated in this paper is that the emissivity of the arc can be predicted well by using the emissivity theory of noble gases (for details see the references cited in Ref. 9). From this theory,
two parameters may be extracted which may be used to increase the emitted radiation in the VUV part of the spectrum. An increase of current enhances the emissivity by raising the temperature and density. The temperature rise is, however, limited by cooling of the arc. If the temperature of the arc is raised too much, its lifetime is drastically reduced. The maximum temperature which can be achieved in this arc design is 16,500 K at atmospheric pressure. We varied the temperature in the arc from 12,200 to 14,300 K. The second procedure for increasing the emitted intensity is raising the pressure inside the arc. If the temperature remains constant, a linear increase of the pressure results in a quadratic increase of the emitted intensity. The pressure inside the arc is between 2 and $6 \times 10^4$ Pa.

Another possibility for increasing the emissivity in the VUV is the introduction of impurities into the argon gas. We have added nitrogen at two different concentrations (1 and 10%).

In the following sections, we present a short review of the emissivity theory of noble gases. Next, the experimental setup used by us to measure the VUV emissivity is described and this presentation is followed by the results of the measurements. Finally, some conclusions will be drawn about the use of a cascade arc as a VUV radiation source.

EMISSIVITY THEORY

In this section, we give a short review of the most important expressions which are used to represent the emissivity of a cascade argon arc. Further details may be found in Wilbers et al. and in the references therein.

The continuum emissivity $\epsilon_\lambda$ of the discharge is governed by the following expressions:

$$\epsilon_\lambda = \epsilon_n + \epsilon_\beta,$$

(1)

where

$$\epsilon_\beta = C_1 \cdot \frac{n^2_2}{\lambda^2/\sqrt{T_e}} \exp \left[ -\frac{hc}{\lambda k T_e} \right] \xi_\beta(\lambda, T_e, 1),$$

(2)

and

$$\epsilon_n = C_1 \cdot \frac{n^2_2}{\lambda^3/\sqrt{T_e}} \left[ 1 - \exp \left( -\frac{hc}{\lambda k T_e} \right) \right] \frac{g_{n,1}}{U_1} \xi_n(\lambda, T_e, 1).$$

(3)

The symbols in these expressions are identified in the Nomenclature. The Biberman factor for the free–free emissivity $\xi_\beta$ is constant in good approximation and may be taken to be equal to 1.23.

Accounting for the effect of absorption, the intensity $I_\lambda$ emitted by the cascade arc is described by

$$I_\lambda = S_\lambda [1 - \exp(-\kappa l)] = S_\lambda [1 - \exp(-\epsilon_\lambda l/S_\lambda)],$$

(4)

where the Planck formula yields

$$S_\lambda = \frac{2hc^2}{\lambda^3} \left[ \exp \left( \frac{hc}{\lambda k T_e} \right) - 1 \right]^{-1}.$$

(5)

It is clear from Eqs. (2) and (3) that a linear increase of pressure (increase of the electron density) results in a quadratic increase of the emissivity $\epsilon_\lambda$ provided the temperature remains constant and the limit of blackbody radiation has not been reached. The dependence on the length is linear. Roughly, the emissivity below 200 nm depends on the electron temperature as in the exponent of Eq. (3).

The emissivity is mainly determined by the free–bound contribution in Eq. (3). The wavelength dependence in this expression is strongly influenced by the Biberman factor. In Fig. 1, this Biberman factor is shown according to Hofsaess in the wavelengths and temperature ranges used in the experiments. The strong temperature dependence of this Biberman factor in the specified wavelength range should be noted. In the visible wavelength range, the temperature dependence is small.
VUV emissivity of a high-pressure cascade Ar arc

![Graph showing emission factors vs. wavelength.](image)

**Fig. 1.** The Biberian factor according to Hofsaess in the wavelength range from 100 to 200 nm. The curves represent the following: (---) 12,200 K; (---) 13,500 K; (---) 14,500 K.

**EXPERIMENTAL STUDIES**

The light source has been described elsewhere. We will therefore not provide details concerning the construction of the arc. In Fig. 2, a schematic drawing is given of the experimental arrangement. A 0.5 m Seya–Namioka vacuum monochromator is kept at a pressure of $1 \times 10^{-4}$ Pa with a turbo molecular pump. A grating of 1200 lines/mm, blazed at 150 nm, allows us to use a spectral range from 50 to 350 nm. Slit widths of 50–100 μm give a spectral resolution from 0.08 to 0.16 nm. A solar blind photomultiplier (Hamamatsu R972), with a spectral range from 115 to 200 nm, was used. The use of this photomultiplier limits the influence of stray light. The lower usable wavelength limit is also defined by the presence of several LiF lenses, a LiF window and an MgF$_2$ window in the photomultiplier. The optical system between the arc and the monochromator converts the opening angle of the emitted beam such that the grating of the monochromator is completely filled. All components are made of LiF and the optical system is kept at the same low pressure as the monochromator itself. The window between the arc and the optical system is also made of LiF, allowing radiation with wavelengths down to 115 nm to pass. Between the end of the plasma (anode) and the exit window is a volume of cold argon gas at the same pressure as in the arc itself. The path length from the anode to the exit window is about 60 mm. The cold argon gas is flushed continuously at a rate of 2 sec/sec to eliminate light-absorbing species produced at the boundary of the arc plasma. Also, traces of water can absorb heavily in the VUV. From 140 to 185 nm, a broad absorption band of water is present. Near the broad maximum of this absorption band, the absorption is 100%. Below 140 nm, the absorption decreases to 20%. Above 185 nm, the absorption has nearly vanished. In Fig. 3, the ratios of the counts obtained with and without flushing are shown for a 60 A and 6 × 10$^3$ Pa arc. This condition has been chosen for comparison because light absorption by water increases with increasing pressure and arc temperature. Although the concentration of water is very low (the hydrogen Lyman-α line could be barely detected), the absorption is strong. With the protecting cold argon flow, that causes flushing from the LiF window to the anode of the arc, the absorption can be eliminated. The cold argon gas serves another important purpose. Radiation at photon energies greater than 11 eV is strongly absorbed by LiF. Severe degradation occurs after several hours of exposure to this radiation. However, the flushed cold argon gas absorbs this radiation, which is emitted by the hot argon, thereby protecting the window. Over several days, the degradation of the window transmission was only a few percent.

To ensure a large intensity range, we used single-photon counting. The count rate was monitored by using a PC for further analysis of the data.
RESULTS

Calibration of an optical system in the VUV is not straightforward. The intensity of the normally-used radiation standards is far too low to calibrate the optical system below 200 nm. Calibration can be done by using hydrogen or argon arcs and line-intensity ratios. This latter technique involves the use of the intensities of lines emitted from the same upper level to different lower levels, one in the visible and the other in the wavelength range to be calibrated.

To calibrate our optical system for the spectral range from 100 to 200 nm, we used the following procedure. In previous measurements, the same optical system was calibrated from 250 to 320 nm using a tungsten ribbon lamp at a temperature of 2667 K. Below 250 nm, the intensity of this source was too low. From the measurements above 250 nm, the electron density and the electron temperature were calculated. In the present set-up, the same arc conditions were used as in the measurements of Ref. 9. The spectral range covered during the measurements extended from 100 to 250 nm, thus providing a link between the calibrated upper wavelength range in the measurements of Ref. 9 and the lower wavelength range in the present measurements. Behringer and Thoma have shown that the free-bound Biberman factor in the wavelength range covered is in agreement with the calculated free-bound Biberman factor of Hofsaess. The new calculations improved the agreement at 16,900 K. At higher temperatures, the agreement is less satisfactory due to the increasing influence of ArII.

The measurements at 250 nm were repeated with and without the arc being flushed. These measurements showed that with increasing current and decreasing pressure, the pressure at the anode region is raised up to 40% (60 A, 2 \times 10^2 Pa) of the pressure at the cathode region. This result

Fig. 2. The experimental set-up. With a system of three lenses (LiF) and two pinholes, the arc radiation is directed into a Seya–Namlioka vacuum monochromator. The detected photons are counted and the counts are monitored and analyzed by using a PC. At the anode, the arc is flushed from the window towards the anode with a flow rate of 2 scs/sec argon. Through the arc channel, a flow rate of 0.2 scs/sec is maintained. Just before the anode, the gas exits from the system.
Fig. 3. The ratio of counts measured in a 60 A/6 \times 10^5 \text{ Pa} set-up with and without flushing.

can be explained by the increasing viscosity of the arc at higher temperatures and by the direction of the flow. The higher rate of flushing towards the anode from the exit window induces a pressure rise at the anode region. Due to the increasing viscosity of the plasma with increasing temperature, the pressure equalization between anode and cathode is inhibited. Only when the current was as low as 20 A, was the pressure at the anode region the same as at the cathodes. With increasing pressure (from 2 to 6 \times 10^5 \text{ Pa}) inside the arc, no discrepancy occurred between theory and measurements. At 2 \times 10^5 \text{ Pa}, the ratio of the intensity according to theory and measurements was 0.99. At 4 and 6 \times 10^5 \text{ Pa}, this ratio was, respectively, 0.98 and 0.99 (this ratio is obtained by dividing the measured intensity by the calculated intensity at several wavelengths).

We therefore used the 20 A, 6 \times 10^5 \text{ Pa} condition to obtain the transmission curve of the vacuum system with a flushed arc. The measured continuum counts at an arc condition of 20 A and 6 \times 10^5 \text{ Pa} are compared with the theoretical continuum intensity of an argon plasma at 12,200 K and 6 \times 10^5 \text{ Pa}. In the calculation of the theoretical continuum intensity with Eqs. (1)-(5), the Biberman factors according to Hofsaess are used. Where lines are present, the continuum is interpolated. In Fig. 4, we show the transmission curve obtained from use of the specified procedure. Below 120 nm, the number of continuum counts after correction for the contribution of stray light was not sufficient to calculate the transmission curve. Stray light from the zeroth order of the grating causes a background which decreases slowly towards higher wavelengths.

The drop in transmission to lower wavelengths is a combined effect of window and lens materials (LiF and MgF_2) and the grating which is blazed at 150 nm. The decrease towards higher wavelengths is a combined effect of the sensitivity of the photocathode material, which decreases exponentially, and the blaze of the grating.

We varied the current through the arc from 20 to 60 A, thereby creating a temperature between 12,200 and 14,500 K. A typical measurement is shown in Fig. 5. The arc current is 40 A and the pressure at the cathode side is 4 \times 10^5 \text{ Pa}. In the spectrum, various atomic lines can be seen. These lines are due to small amounts of nitrogen, oxygen, carbon, and water which is present in the argon gas. Although we used high-grade argon gas (Messer Griesheim 5.0), these lines could not be avoided. Also, some weak argon-ion lines are present. In Table 1, the most important spectral lines are given together with the corresponding atom. Towards 125 nm, the number of counts decreases rapidly. It is clear from Fig. 5 that below this wavelength, the sensitivity of the optical system is not sufficient to yield accurate data on the continuum radiation. The measurements have been extended to 100 nm in order to allow us to subtract the contribution of stray light due to zeroth order of the grating.
The change of pressure results in a strong increase of the emissivity. In Fig. 6, we show measurements at a temperature of 13,500 K for 2 and $6 \times 10^5$ Pa. The blackbody limit curve at 13,500 K is also shown in Fig. 6.

In Fig. 7, the effect of the change of current is shown. The temperature has the values of 12,200 and 14,500 K. The pressure is $4 \times 10^5$ Pa.

In Fig. 6, the impurity lines do not significantly increase in strength when the pressure is changed from 2 to $6 \times 10^5$ Pa. The line intensity depends linearly on the electron density as long as the blackbody ceiling has not been reached. The continuum intensity is proportional to the square of the electron density, as can be seen in Fig. 6.

In Fig. 7, we observe that both the lines and the continuum increase due to the increase of temperature and electron density.
VUV emissivity of a high-pressure cascade Ar arc

Table 1. Atomic lines—the spectral range from 125 to 200 nm.

<table>
<thead>
<tr>
<th>Wavelength (nm)</th>
<th>Line</th>
<th>Intensity (A)</th>
</tr>
</thead>
<tbody>
<tr>
<td>127.75</td>
<td>C I</td>
<td>149.47</td>
</tr>
<tr>
<td>130.22</td>
<td>O I</td>
<td>156.1</td>
</tr>
<tr>
<td>130.55</td>
<td>O I</td>
<td>157.5</td>
</tr>
<tr>
<td>131.07</td>
<td>N I</td>
<td>160.04</td>
</tr>
<tr>
<td>131.95</td>
<td>N I</td>
<td>160.35</td>
</tr>
<tr>
<td>132.93</td>
<td>C I</td>
<td>160.65</td>
</tr>
<tr>
<td>133.53</td>
<td>C II</td>
<td>165.72</td>
</tr>
<tr>
<td>141.19</td>
<td>N I</td>
<td>174.27</td>
</tr>
<tr>
<td>145.91</td>
<td>C I</td>
<td>174.53</td>
</tr>
<tr>
<td>146.33</td>
<td>C I</td>
<td>175.19</td>
</tr>
<tr>
<td>148.18</td>
<td>C I</td>
<td>193.09</td>
</tr>
<tr>
<td>149.28</td>
<td>N I</td>
<td></td>
</tr>
</tbody>
</table>

From the continuum measurements, the free–bound Bieberman factor can be extracted. After correction for the pressure rise due to flushing, the measured values can be compared with the theoretical free–bound Bieberman factors. This is done in the following manner. The measured continuum intensities have been calculated using the transmission of the optical system. This transmission is obtained from measurements at the arc condition of 20 A and $6 \times 10^5$ Pa. In this procedure, the free–bound Bieberman factor of Hofsaess at 12,200 K has been used in the calculation of the theoretical continuum emissivity. Therefore, the transmission function of the optical system contains a quotient of the experimental and theoretical free–bound Bieberman factors. This quotient is now also present in the calculations of the intensities at the other arc conditions. From the measured continuum intensities calculated according to our procedure, the free–bound Bieberman factor has been extracted.

An absolute value cannot be extracted because the ratio of the experimental to the theoretical Bieberman factor in the transmission function is still present at 12,200 K. In Fig. 8, we show the measured and theoretical free–bound Bieberman factors at 12,200 K ($2 \times 10^5$ Pa), 13,500 K ($6 \times 10^5$ Pa) and 14,500 ($4 \times 10^5$ Pa). The agreement between the theoretical and measured

![Intensity plot](image.png)

Fig. 6. The intensities at 2 and $6 \times 10^5$ Pa for the arc at a temperature of 13,500 K. The blackbody limit is also shown.
free-bound Biberman factors is good. For the values at 14,500 K a small water absorption is still visible. Below 135 nm, the total number of counts in the measurements does not allow us to draw any conclusions about the absolute dependence of the Biberman factor on the wavelength.

If a part of the VUV emissivity needs stronger enhancement than can be obtained by raising the pressure and current, one can introduce impurity gases. The change of intensity when 10% nitrogen is added to the argon gas can be seen in Fig. 9. As reference the pure argon measurement and the blackbody ceiling are also shown. The lines at 149.33 and 174.36 nm are both due to resonance to two ground levels of the neutral nitrogen atom. The transition probabilities are large and the lines are strongly broadened. The line wings cause an increase of the emissivity over a large range of wavelengths as can be seen from Fig. 9. The continuum intensity decreases slightly. In

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**Fig. 7.** The VUV intensities at $4 \times 10^9$ Pa and 12,200 and 14,500 K.

**Fig. 8.** The theoretical and measured free-bound Biberman factors. The curves and symbols represent the following temperatures: $(--, \triangle) 12,200$ K; $(--\cdots, \circ) 13,500$ K; $(---, +) 14,500$ K. The curves are the values of Hofsass, whereas the symbols refer to our measurements.
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Fig. 9. The intensities of the cascade arc at 13,500 K and 4 × 10^9 Pa with and without 10% molecular nitrogen. The blackbody ceiling at 13,500 K is also shown.

combination with the increase due to the line wings, the net effect is a decrease of continuum intensity only above 160 nm. Below 150 nm, the rise in emissivity is greater than a factor of two.

The influence of the pressure on the broadening of the lines is shown in Fig. 10. The group of lines at 149.33 nm is more strongly broadened than the lines at 174.36 nm because the transition probability is larger, i.e., 5.5 × 10^8 and 2.0 × 10^8 sec^−1.15

As expected, the broadening of the lines increases with increasing pressure thereby raising the total intensity further compared to the increase of intensity when no additional nitrogen is present.

CONCLUSIONS

Measurements of the VUV emissivity of the cascade argon arc have shown that the arc is a strong VUV radiator, the intensity of which can be calculated in good approximation. An increase of pressure and/or current enhances the emissivity considerably, in agreement with the emissivity

Fig. 10. The influence of the pressure on the broadening of the nitrogen lines. The pressures are 2 and 4 × 10^9 Pa. The blackbody limit at 13,500 K is shown as a reference.
theory. Further increases of the intensities at selected wavelength regions can be achieved by adding impurity gases. With increasing concentration and pressure, the lines become stronger and are broadened. The addition of nitrogen increases the intensities at several wavelength sites considerably (by a factor of 10) as a result of the contributions by wings of the broadened lines. Below 150 nm, the intensity was raised by more than a factor two.

The relative temperature dependences of the free–bound Bierbman factors have been determined as functions of the wavelength in the spectral range from 135 to 200 nm. Below 135 nm, the number of photon counts in the measurements was not sufficient to yield reliable results. The agreement between the measurements and the values of Hofsaess is good.

REFERENCES


NOMENCLATURE

\[ \varepsilon_t = \text{Total emissivity} \]
\[ \varepsilon_f = \text{Free–free emissivity (electron–ion)} \]
\[ \varepsilon_b = \text{Free–bound emissivity} \]
\[ \lambda = \text{Wavelength of the radiation} \]
\[ C_i = \text{Electron–ion continuum constant (1.63 \times 10^{-43} \text{Wm}^4 \text{K}^{-5} \text{sr}^{-1})} \]
\[ n_e = \text{Electron density} \]
\[ T_e = \text{Electron temperature} \]
\[ \xi_f = \text{Bierbman factor for free–free emissivity} \]
\[ \xi_b = \text{Bierbman factor for free–bound emissivity} \]
\[ U_i = \text{Partition function of the ion (1)} \]
\[ \xi_{1,1} = \text{Statistical weight of the ion ground level} \]
\[ k = \text{Absorption coefficient} \]
\[ S_i = \text{Source function} \]
\[ c = \text{Velocity of light} \]
\[ k = \text{Boltzmann's constant} \]
\[ h = \text{Planck's constant} \]
\[ l = \text{Length of the discharge} \]