Paper

Radiant Efficiency of Coaxial KrBr* Excilamps with High Power Density

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Received May 2, 2013, Accepted February 24, 2014

ABSTRACT

The characteristics of the emission spectra of coaxial KrBr* excilamps are investigated with high electric power density ranging from 557 to 803 W/m driven by sinusoidal electronic control gear (ECG) as well as 367 to 546 W/m by pulsed ECG, based on the dielectric barrier discharge. The radiant power of intense narrow band ultraviolet radiation at 207 nm is derived from the irradiance according to Keitz formula while the input power is measured and compared by Lissajous figure and integration of voltage and current. It is shown that the measurement power of integration of voltage and current achieves more accuracy. And the radiant efficiency of 207 nm varies with the gas parameters, demonstrating an optimum 4.1% of energy converted to 207 nm band radiation at pressure 300 mbar and bromine percentage 0.25%.

KEYWORDS: dielectric barrier discharge; KrBr* excilamps; 207 nm; Keitz formula; Lissajous figure

1. Introduction

In recent decades more attention is paid to excilamps which are characterized by a relatively narrow-band radiation of ultraviolet (UV) and vacuum ultraviolet (VUV). The excimer molecules can consist of rare gas (Rg*)1–7), halogen (X2*)2–4)8), rare gas halide (RgX*)2–3)9–17) or mercury halide (HgX*)11)18). Excilamps have advantages over the commonly used mercury lamps whose radiant efficiency at 254 nm exceeds 40% while that at 185 nm reaches about 9%. Thanks to the absence of resonance radiation trapping by ground state, no self-absorption occurs when high intensity narrow-band radiation is emitted from the discharge, which means that the radiant efficiency will not be limited by increasing power density. Therefore, it can be applied in industrial applications that demands large amounts of UV radiations. Besides, excilamps are more environmental friendly compared to mercury lamps as the mercury is harmful to human bodies and hard to be treated. And the geometry and the wavelength of the excilamps can be designed according to various applications. As for KrBr* excimer which provides a narrow band UV radiation at 207 nm, the photon emitted has high energy and its absorption rate by DNA molecules is 60% higher than that of 254 nm19). Therefore, it has great potential in certain UV applications, especially in those that are sensitive to UV wavelengths.

There are a few reports have shown the research on KrBr* excilamps. Feng et al. indicated that the optimum total pressure should be 400 mbar when Kr:Br2=30:1 or bromine pressure constant at 20 mbar for a planar lamp with discharge gap 3 mm, but no data about the radiant efficiency at 207 nm was given12). The discharge was studied by Erofeev et al. in a chamber with distance between anode and cathode varying from 5 to 15 mm, attaining the maximum efficiency of 4% at total pressure of 1000 mbar with Kr:Br2=100:114). A high voltage pulse with amplitude 150 kV was used. Shuabov et al. studied on the KrBr* excilamps based on low-pressure glow discharge with a 100-mm distance between electrodes, obtained the total VUV–UV efficiency of 8–10% at optimum krypton pressure 4–6 mbar and bromine pressure 0.5–1.5 mbar20). A relatively more uniform coverage of the short wavelength radiation (160–300 nm) was achieved instead of maximum radiant power at 207 nm. Avdeev et al. reported the optimum gas parameters under the condition of dielectric barrier discharge (DBD) at pressure of 308 mbar with Kr:Br2=400:1, which leads to the efficiency of only 2.4%21). But the data reported was not referred to the input electric power density. Sosnin et al. researched on the radiative and thermodynamic parameters of a DBD based KrBr* excilamp with gap 11.5 mm, presented the maximum UV power and efficiency at 1.6 W and 3%, respectively17). According to the figure given in the article, the input power in unit length ranges from 218 to 480 W/m. There is obviously big discrepancy in these results,
which may be caused by various geometries and discharge power density. And few reports have been published to study the high power density discharges which meet the industrial applications.

In this paper, the electrical and radiant parameters are studied for coaxial KrBr* excilamps based on DBD, driven by sinusoidal and pulsed electronic control gears (ECG). The measured power density driven by sinusoidal ECG varies from 557 to 803 W/m while 367 to 546 W/m by pulsed ECG. The 207 nm radiant power is calculated through the measured irradiance according to the Keitz formula. The dependences of 207 nm radiant efficiency on the total pressure and the bromine percentage are illustrated. As for the input power, Lissajous figure method and integration method of voltage and current are adopted and compared.

2. Experiments

2.1 Lamps preparation

The sample lamp is illustrated in Figure 1 with a coaxial geometry. Each lamp is made of two coaxial quartz tubes sealed with each other, which have outer diameter (OD) 38 mm and 30 mm respectively, both with wall thickness of 1.5 mm. The discharge cavity has a total length of 150 mm. And the transmittance of the quartz tubes is about 54% at 207 nm.

Two pieces of stainless steel foils, attached firmly to the inner surface of the inner tube, are used as a high voltage electrode. And a ring metal mesh is mounted on the outer tube as a grounded electrode. The annular discharge cavity with gap of 6.5 mm is filled with bromine and krypton.

The lamps are respectively filled with krypton and bromine gas mixture at various pressure as well as bromine percentage. The gas was firstly mixed into a glass bottle and then filled in the discharge tubes for different total pressure, which ensures the constant partial pressure ratio of bromine. The saturated vapour pressure of bromine is very high, i.e. 246 mbar at temperature 295 K (22°C)\(^2\)). And considering its small amounts (no more than 4 mbar for total pressure 400 mbar and bromine percentage 1%), bromine is unsaturated and kept in gas state whether the lamp is on or off. The bottle and excilamps bulbs were kept in room temperature (25°C) in the processes of mixing and filling gases, which confirmed accurate vapour pressure of bromine.

2.2 Radiant power measurement

Figure 2 shows the experimental setup for radiant power measurement, which is similar to that adopted by Zhuang et al. for KrCl* excilamps\(^15\). The lamps are fixed horizontally with two Teflon rods, driven by a sinusoidal ECG with frequency 35–50 kHz and a pulsed ECG with pulsed width about 0.75 μs and repeating frequency 90 kHz. The 207 nm band irradiance normal to the KrBr* excilamp is detected by a combination of UV sensor (Hamamatsu H8025-222, modified to 207 nm) of the UV power meter (Hamamatsu C8026) which is calibrated at this wavelength, as well as a spectrometer. The spectrum of the KrBr* excimers is obtained by a 8-channels spectrometer (Avantes AvaSpec-2048FT-8-RM, resolution 0.05 nm) in range 200–300 nm, which have been calibrated by a deuterium tungsten halogen calibration standard lamp (Mikropack DH2000-CAL, spectral irradiance standard). All the surfaces in the lab are covered with light-absorbing black velvet cloth so as to avoid the radiation reflection from the walls, table and instruments.

Due to the wide spectral response of the UV sensor from 160 to 350 nm, the reading of the UV power meter includes the contributions of all the bands shown in Figure 5 instead of 207 nm band only. Thus the UV spectrum of the excilamp is used to calibrate and separate the irradiance of each band. And “207 nm band irradiance” refers to the integral intensity of the first whole band in Figure 5 whose peak at 207 nm. Consequently, “207 nm radiant power” and “207 nm radiant efficiency” etc. in the text represent the band integrals as well.

Then the radiant power \( P_{rad} \) of 207 nm can be calculated from the band irradiance according to Keitz formula\(^23\), in respect that the excilamp can be considered as a linear light source, i.e. the diameter is much
less than the length:

\[ P_{\text{rad}} = \frac{2\alpha^2 DLE}{2\alpha + \sin 2\alpha/\tau} \]  

(1)

where \( E \) is the band irradiance (W \cdot m^{-2}), \( D \) represents the distance (m) from lamp center to the sensor, \( L \) is the discharge length (m) of the lamp covered with metal mesh, and \( \alpha \) refers to the half angle (radians) subtended by the lamp at the sensor position\(^{19}\). For this calculation, \( D \) should be more than 2.5 times of \( L \). And \( \tau \) means the transmittance of the quartz tube which is 54\% for 207 nm in this paper.

### 2.3 Electric power measurement

The input power of the KrBr\(^*\) excilamps are measured by two methods respectively: integration method and Lissajous figure method. For integration method, the average input power \( P_{\text{in}} \) can be derived from the direct integration in period \( T \) of transient power which is the product of lamp voltage \( u \) and current \( i \), as see in Eq. (2).

\[
P_{\text{in}} = \frac{1}{T} \int_0^T u i dt
\]  

(2)

A high voltage probe (Tektronix P6015A, 1000 : 1, rise time 5 ns) and a current probe (Pearson P4100, 1 V/A, rise time 10 ns) are used to detect the voltage and current of the excilamp and connected to a digital storage oscilloscope (Nicolet Sigma 60, bandwidth 200 MHz, sample rate 200 MS/s, working at storage length 8 bit) to store the waveforms, as presented in Figure 2. In order to improve the measurement accuracy, the waveforms are recorded at least in four cycles.

As regards Lissajous figure method, the input power can be derived from the capacitance of a mica capacitor in series and the area of Lissajous figure\(^{25}\). The charge of the capacitor can be presented in Eq. (3):

\[
dQ = idt = C_m u d\mu_m
\]  

(3)

where \( C_m, u_m \) are respectively the capacitance and the voltage of the mica capacitor. So the input power of the excilamps can be derived by

\[
P_{\text{in}} = \frac{1}{T} \int_0^T i u d\mu = \lambda\mu_m C_m u d\mu_m = C_m f A
\]  

(4)

where \( f \) is the frequency of the ECG and \( A \) represents the area of the \( u-u_m \) Lissajous figure. The lamp voltage and capacitor voltage are measured and then plotted by Origin software, which are integrated to obtain the area of the figure and the consequent electric power.

### 3. Results and discussion

#### 3.1 Input lamp power

The waveforms of lamp voltage, current and power driven by the sinusoidal ECG are displayed in Figure 3. It is clear that the lamp voltage has a perfect sinusoidal wave while the current is distorted with several pulses. The waveform of \( i \) is not symmetrical in the positive and negative cycle, which cause the relating curve of \( ui \). The asymmetry may be caused by the different structures of inner and outer electrodes which lead to the different pulses in the waveform of current. Depending on the total gas pressure (100–400 mbar) and the bromine percentage (0.1–1.0\%), the peak-to-peak voltage ranges from 10.87 to 12.27 kV, and the root mean square (RMS) value of current keeps at about 0.12 A. The lamp power in unit length is calculated from 557 to 803 W/m for various pressure and ECG power settings.

The voltage waveform of the lamp driven by the pulsed ECG is plotted in Figure 4, showing a very short
negative pulse width (0.75 μs) at repeating frequency of 90 kHz. The amplitude of the voltage pulse varies from 3.87 to 6.93 kV at a relatively constant RMS of current around 0.29 A under the same gaseous parameters to those driven by the sinusoidal ECG. The corresponding power density ranges from 367 to 546 W/m. There are two peaks observed in the current waveform with the width of 0.3 and 0.1 μs respectively, consistent with the case of pulsed-driven DBD. Due to the unipolar-pulse of the ECG, the secondary peak has the opposite polarity to that of voltage, which agrees with the results of Liu et al.26 Differed from the case of bipolar driven such as sinusoidal wave, all the electric parameters return approximately to zero after each pulse. Therefore, the secondary discharge is energized by the memory charges and corresponding energy that is stored from voltage pulse and released at or shortly after the pulse falling, leading to the opposite phase of the current peak.

The measured results of input lamp power by integration method and Lissajous figure are compared in Table 1. Mica capacitors $C_{m}$ with capacitance of 19.3 nF and 36.6 nF have been chosen. It is shown that the power of Lissajous method is 12.9% and 9.3% lower than integration method for capacitor of 19.3 nF and 36.6 nF. The underestimation of lamp power consumption through Lissajous method may be resulted from the partial voltage of $C_{m}$. Due to nearly constant output of ECG, the voltage drop on $C_{m}$ leads to reduction of lamp voltage and resulting lower lamp power. As the capacitance and the voltage drop of $C_{m}$ are negatively related, partial voltage of lamp connected with 19.3 nF capacitor ($V_{p} = 9.44$ kV) is further smaller than that in 36.6 nF case ($V_{p} = 9.80$ kV). Therefore, the Lissajous method (19.3 nF) shows the smallest power values.

Besides, the integrated lamp power in series with the mica capacitors has been listed in Table 1 as well. Compared to those integral values, the results of Lissajous figure are still smaller in the same situation, which may be caused by the uncertainty of the measurement on the capacitors, such as the underestimation of the capacitance and the accuracy of the probe used to test the partial voltage of $C_{m}$. For integration method, the measured results are more accurate since it is only influenced by the uncertainty of the measurement instruments.

The total uncertainty of the measured electric power $\delta_{\text{tot}}$ is the result of the combination of the system error $\delta_{\text{sys}}$, and the random error $\delta_{\text{ran}}$, while $\delta_{\text{tot}}$ is influenced by the accuracy of instruments and measurement method, such as the accuracy of oscilloscope $\delta_{\text{osc}}$ 1%, high voltage probe $\delta_{\text{vp}}$ 3%, current probe $\delta_{\text{cp}}$ 0.4%, error caused by phase difference of voltage and current probe $\delta_{\text{pd}}$ 0.6%. Therefore, the system error of lamp power measured by integration method is derived

$$\delta_{\text{sys}} = \sqrt{\delta_{\text{osc}}^2 + \delta_{\text{vp}}^2 + \delta_{\text{cp}}^2 + \delta_{\text{pd}}^2} = 3.2\%,$$

resulting in the total uncertainty $\delta_{\text{tot}} = \sqrt{\delta_{\text{sys}}^2 + \delta_{\text{ran}}^2} = 3.4\%$ due to the random error caused by period selection $\delta_{\text{ran}}$ is about 1.0%.

### 3.2 Spectrum of the KrBr* excilamps

The spectrum of KrBr* excilamp are shown in Figure 5. It is obvious that the peak strong band of KrBr* excimer locates at 207 nm, with full width at half maximum (FWHM) 1.53 nm, which is emitted by the transition $B \rightarrow X$. There are still several other spectral bands shown in Figure 5, which are relatively weak. The bands at 222 and 228 nm, respectively resulted from the transition $C \rightarrow A$ and $B \rightarrow A$, are combined here due to the high pressure of 300 mbar. Besides, the band radiation at 291 nm is emitted by Br$_2^+$ excimers.

At the constant bromine percentage of 0.25%, the dependence of the spectrum of KrBr* excilamp on the total pressure is illustrated in Figure 6, indicating the main effect on the transition $B \rightarrow X$. The FWHM of 207 nm band radiation increases from 1.45 nm to 1.53 nm when the total pressure is enhanced from 100 to 300 mbar. Different from that at 300 mbar, the bands emitted because of the transition $C \rightarrow A$ and $B \rightarrow A$ are separated at 100 mbar.

### 3.3 Radiant efficiency of 207 nm

The radiant efficiency of 207 nm band, obtained from the given UV radiant power $P_{\text{rad}}$ and the input lamp power $P_{\text{in}}$

$$\eta_{207} = \frac{P_{\text{rad}}}{P_{\text{in}}}$$

(5)

can be used to determine the optimum discharge parameters such as total pressure and bromine percent-

### Table 1  Comparison of the results by integration and Lissajous figures

| Methods                  | Test 1  | Test 2  | Test 3  | Average | Deviation |
|--------------------------|---------|---------|---------|---------|-----------|
| Integration (no $C_{m}$) | 96.57   | 96.44   | 96.54   | 96.52   | 0.06%     |
| Lissajous (19.3 nF)     | 84.31   | 83.92   | 84.06   | 84.10   | 0.19%     |
| Integration (19.3 nF)   | 88.27   | 87.86   | 88.18   | 88.10   | 0.20%     |
| Lissajous (36.6 nF)     | 88.18   | 87.10   | 87.50   | 87.59   | 0.51%     |
| Integration (36.6 nF)   | 92.96   | 91.74   | 92.09   | 92.26   | 0.55%     |
The total pressure dependence of 207 nm radiant power and efficiency are respectively displayed in Figure 7 and Figure 8. As shown in Figure 7, the 207 nm radiant power is generally increased with total mixture pressure, due to more excited and ionized atoms to produce KrBr* excimers. However, the maximum radiant efficiency is reached at the optimum pressure about 300 mbar, which is 4.1% for pulsed ECG, and 1.5% for sinusoidal ECG considering the transmittance 54% of quartz tubes at 207 nm. This dependence is associated with the kinetics of the formation and decay of the KrBr* excimers. The electrons first gain the energy from the electric field and initiate the reactions:

\[ e + \text{Kr} \rightarrow \text{Kr}^* + e \]  
\[ e + \text{Kr} \rightarrow \text{Kr}^+ + 2e \] (6)  

Then the KrBr* excimers are produced through three-body recombination and Harpooning reactions respectively:

\[ \text{Kr}^+ + \text{Br}^- + \text{M} \rightarrow \text{KrBr}^* + \text{M} \]  
\[ \text{Kr}^* + \text{Br}_2 \rightarrow \text{KrBr}^* + \text{Br} \] (9)  

At low pressure, the rate of three-body recombination is relatively small, so the major contribution to the formation of KrBr* excimers is implemented by Eq. (10). Meanwhile it is possible to form the Br₂* molecules by the collision of excited Kr* and Br₂ molecules, which results in comparatively strong radiation at 291 nm. When the total pressure is enhanced, the rate of three-body recombination increases with gas density, resulting in more KrBr* excimer radiation and less Br₂* radiation. However, the dissociation and quenching rate of
excimers was enhanced at higher pressure owing to more frequent collisions between excimers and other heavy particles, which leads to the reduced spontaneous emission of the excited molecules. Therefore, there is an optimized pressure which is 300 mbar here. A similar tendency for the case of KrCl* and XeCl* excimers has been reported respectively\(^{15,27}\) .

Figure 9 and Figure 10 respectively illustrate the dependence of 207 nm radiant power and efficiency on bromine percentage at the constant total pressure of 300 mbar. It is obvious that both of them reached maximum at the optimum bromine percentage about 0.25%. It is due to the competition of intense three-body recombination emitting 207 nm radiation and the negative characteristic of bromine ions which is harmful to lamp start-up and UV emission when the Br\(_2\) density increases. As shown in Eqs. (11) and (12), the collisions of bromine molecules with other particles may also produce Br\(_2^*\) whose formation opportunity is positive related to Br\(_2\) density. Superabundant bromine will lead to less KrBr* excimers and consequently UV reduction. Besides, Br\(_2\) will absorb electrons and form negative ions, increasing the ignition voltage of the lamp.

\[
\text{Kr}^* + \text{Br}_2 \rightarrow \text{Kr} + \text{Br}_2^* \quad (11)
\]

\[
e + \text{Br}_2 \rightarrow \text{Br}_2^* + e \quad (12)
\]

The uncertainty of 207 nm radiant power is mainly caused by the accuracy of UV power meter 10%. Besides, random errors must also be taken into account such as 0.5% of data acquisition, 0.25% of radiant length, 0.13% of distance between lamp and detector, as well as 1.7% of lamp positioning, with a total random error 1.8% from the sum of squares. Thus the uncertainty of 207 nm radiant power is 10.2% while that of radiant efficiency is 10.8%.

4. Conclusions

The electric and radiant characteristics of coaxial KrBr* excilamps have been studied based on DBD at high power density of 557–803 W/m for sinusoidal ECG and 367–546 W/m for pulsed one. The electric input power is measured by integration of voltage and current as well as Lissajous figure methods, indicating that both the methods can be adopted but the former is more accurate. The radiant power of 207 nm band radiation is calculated from the irradiance according to Keitz formula and therefore the radiant efficiency is derived, pointing out that the maximum efficiency is achieved at total pressure of 300 mbar with bromine percentage of 0.25%. The optimal radiant efficiency is 4.1% for pulsed ECG considering the transmittance 54% of quartz tubes at 207 nm.

Acknowledgements

This work is financial supported by National Nature Science Foundation of China (Grant No. 51108211 and 50407005), and Department of Science and Technology, Guangdong Province, China (Grant No. 2009B090300308).

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This paper is based on the authors’ presentation given at the 13th International Symposium on Science and Technology of Lighting (LS-13) held June 24–29, 2012, in Troy, New York, USA.