Paper

Basic Performance of VUV Exposure Systems Using Head-on Type Ar₂⁺ and Kr₂⁺ DBD Excimer Lamps

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ABSTRACT

As an application of dielectric barrier discharge(DBD) lamps, vacuum-ultraviolet(VUV) exposure systems of both Ar₂⁺ and Kr₂⁺ excimers have been developed and their basic performance has been evaluated. Each system comprises a 20-W DBD excimer lamp, a sinusoidal wave high-voltage power supply, and a cylindrical lamp holder. A high-voltage transformer and a lamp are designed to be set as close together as possible in the holder. The inside of the holder is purged and filled with nitrogen gas when it is in use. The excimer lamp has an MgF₂ window at the lamp end and is a "head-on type" lamp. The head-on type Ar₂⁺ excimer VUV system produced monochromatic light at 127 nm (with a 9.8-nm FWHM). Its irradiance had an almost homogeneous distribution within a 15-mm diameter planar circle at 50 mm from the output window and was about 0.4 mW/cm². The head-on type Kr₂⁺ excimer VUV system produced monochromatic light at 147 nm (with a 13.2-nm FWHM). Its irradiance had an almost homogeneous distribution within a 15-mm diameter planar circle at 50 mm from the output window and was about 1.4 mW/cm². The lifetime of the lamps in these system is defined as the time point at which irradiance drops to 50% of its initial value. It was 500 and 1500 h, respectively, for the Ar₂⁺ and Kr₂⁺ excimer lamps.

KEYWORDS : Ar₂⁺ excimer, Kr₂⁺ excimer, excimer lamp, exposure system, dielectric barrier discharge, vacuum ultraviolet

1. Introduction

Lamps of vacuum-ultraviolet radiation with high photon energy have been required as the light sources for dry cleaning, surface reforming, photolysis, photosynthesis, photochemical vapor deposition, and so on that use photochemical reactions. It is known through experience that the light source to be used for the photochemical reactions in experiments is required to have an irradiance area with diameter exceeding approximately 10mm and an irradiance intensity exceeding 0.1W/cm²/10nm. Furthermore, it is desirable to have an irradiance of higher efficiency in a specific wavelength zone alone.

The deuterium discharge lamp, which has been used as a practical vacuum-ultraviolet light source with the feature of easy handling, has a continuous spectrum in the vacuum-ultraviolet zone. This lamp is excellent as a point source of light, but it is considered inadequate for obtaining a high irradiation intensity.

In recent years, excimer lamps have been studied and developed as efficiently radiating light in specific wavelength zones. It has been known for a long time that excimer radiations of rare gases are formed by dielectric barrier discharge ^{1) 2)}. Moreover, studies aiming at the practical use of rare gas and rare gas-halogen excimer lamps have been made in Europe since 1988 ^{3) 4) 5)}.

We previously developed a Xe₂⁺ excimer radiation exposure system excited by dielectric barrier discharge in order to obtain a vacuum-ultraviolet light source aimed mainly at photo-cleaning and surface reforming ^{6) 7)}. In this study we have developed head-on type Ar₂⁺ and Kr₂⁺ excimer radiation exposure systems. It is known that Ar₂⁺

and Kr₂⁺ excimer bring higher photon energy, and these exposure systems are suitable for practical use in a basic research on photochemical reactions or a measurement in which high photon energy is required, and we have clarified the performances of these exposure systems. Although a measurement of vacuum-ultraviolet radiation below 150nm is generally done in a vacuum condition, through a quantitative survey of the absorption of Ar₂⁺ and Kr₂⁺ excimer radiation in nitrogen gas, we have found that there is no practical influence in the gas when the distance between a window of the exposure system and a sample is less than 250mm. Furthermore, an operating method to force the decay time of the excimer radiation be constant was studied for evaluating decay properties of phosphors for plasma display panels.

2. Outline of System

The developed exposure system comprises a lamp, a lamp holder containing a transformer therein, and a operating power supply.

2.1 Structure of Lamp

The purpose of this system is to cause a photochemical reaction or to perform measurements by irradiating a material of about 10mm diameter at an irradiation intensity exceeding 0.1W/cm²/10nm. Since an excimer radiation to be indifferent from a resonance line of atoms has no self-absorption, the radiation from a deep inside of thick plasma can be radiated without loss. In view of these two factors, we have adopted a structure of a head-on-type

Fig.1 A schematic side view of the head-on type excimer lamp.

Fig.2 Spectral transmittance of a MgF_2 window of the excimer lamp is shown together with that of synthetic silica glass. ($t=1.5\text{mm}$)

lamp that brings high irradiation intensity in comparatively small area.

The schematic side view of the head-on type Ar_2^* and Kr_2^* excimer lamps is shown in Fig. 1. Two pieces of silica glass with different diameters were mounted and sealed coaxially, and a discharge gas was filled between them. Metallic electrodes of cylindrical sections were put on the inner surface of the inner tube and the outer surface of the outer tube, creating a structure in which the discharge is along the radii of the tubes. An output window was set at one end of the lamp, through which the excimer radiation was emitted.

The outer diameter of the lamp was $\phi 26\text{mm}$, the overall length was 220mm, the length of the discharging space was 150mm, and the input electric power for the lamp was about 20W. The discharging gap was about 4mm, the gas pressure of the Ar_2^* and the Kr_2^* excimer lamps was about 70kPa, and the discharge gap and the gas pressure were adjusted so that the efficiency was maximized with under 20W of lamp power input.

MgF_2 was used for the window material. The spectral transmittance of the MgF_2 used is shown in Fig. 2. For reference, the spectral transmittance of synthetic silica glass (thickness 1.5mm) is also shown. The transmittance of MgF_2 is greater than approximately 60% at 120nm, and

Ar_2^* and Kr_2^* excimer radiation are transmitted acceptably.

2.2 Lamp Holder and Source of Operating Power

Fig. 3 shows a cross sectional view of the cylindrical lamp holder. A water-jacket cooling structure was adopted for the lamp holder in order to suppress changes in temperature of the operating lamp as far as possible and to minimize the fluctuation of light output in the initial stages of operation.

The radiations from Ar_2^* and Kr_2^* excimer are in vacuum-ultraviolet zone and are absorbed by oxygen in the air. Therefore, it is necessary to have the inside of the luminaire evacuated or filled by non-absorbing rare gases. In the case of vacuum, however, it is necessary to maintain a high vacuum below $8 \times 10^{-7}\text{Pa}$ in order to avoid deposition of contaminants on the window of the light source. A vacuum pump is required to maintain the high vacuum in the lamp holder, which results in an enlarged system. Additionally, from a view point of outgassing from the surface of the lamp holder or lamp replacement, it is not practical to keep high vacuum condition in the lamp holder inside for long time. The case of filling the holder with the rare gases is similarly not practical. The method of rare gas purging also does not meet cost performance. Therefore, we decided for this exposure system to have a small quantity of nitrogen gas purging inside the lamp holder. Details of the influence of absorption by nitrogen will be explained in section 5.

An ICF70 flange was attached at the front end of the lamp holder, and it could be attached to a vacuum chamber and so on via the MgF_2 viewing port. There was a space of about 10mm between the light output window of the lamp and the front end of the lamp holder.

The voltage applied on the lamp had a frequency of about 20kHz and a peak-to-peak voltage of about 9kV. Its waveform was sinusoidal but not symmetrical (Refer to Fig. 5). A transformer for resonance and step-up was set in the rear of the lamp holder. The high-voltage generation section was built into the lamp holder for safety and to

Fig.3 A cross sectional view of the cylindrical lamp holder. The high-voltage transformer was affixed near the lamp. The lamp was purged with nitrogen gas and the lamp holder jacket was cooled by water when in use.

suppress the generation of electromagnetic noise. The operating power supply include a logic circuit to control on and off periods of lamp operation except power circuit modules.

3. Light Output Properties

3.1 Spectral Distribution

The vacuum-ultraviolet spectrometer (Acton Research VM502) was used in measuring spectral distribution. Vacuum-ultraviolet radiation coming out of the exit slit of the spectrometer was converted into visible light by sodium salicylate and detected by a photomultiplier. The lamp was set at a distance of about 200mm from the inlet window (MgF_2) of the spectrometer, and the spectral profile of the excimer radiation was measured after the space between the lamp and the inlet window was purged and filled with nitrogen. Absorption by nitrogen will be discussed in section 5. Also, a 30W deuterium discharge lamp with a MgF_2 window was set at the same distance and the spectral profile was measured in order to compare the intensities.

Fig4 Typical spectral profiles of head-on type Ar_2^* and Kr_2^* excimer lamps are shown. The spectrum of a deuterium discharge lamp is also indicated.

Fig5 Waveforms of VUV radiation and high-voltage are indicated. The repetition frequency is 20kHz.

The results of measurements at a 2.4nm measuring width is shown in Fig. 4⁸⁾. The results in Fig. 4 are without spectral sensitivity adjustments associated with the measuring system. The central wavelengths of the Ar_2^* and Kr_2^* excimer radiations were 127nm and 147nm, respectively, and their FWHM were 9.8nm and 13.2nm, respectively. Although there is a dip in the vicinity of the peak of the Kr_2^* excimer radiation, it is attributable to nitrogen absorption, which will be explained later. Comparison of these intensities versus that of a deuterium discharge lamp was made by integrating the range of $\pm 5\text{nm}$ of the central wavelengths of the Ar_2^* and Kr_2^* excimer radiations. The intensities of the Ar_2^* and Kr_2^* lamps were approximately 5 times and 8 times of the intensity of the deuterium discharge lamp, respectively.

3.2 Time-dependent waveforms of Excimer Radiations

Time-dependent waveforms of the head-on type Kr_2^* excimer radiation exposure system were by a sensor which consisted of an $\text{Y}_2\text{SiO}_5: \text{Ce}^{3+}$ phosphor with an 80ns 1/10 decay time and a photomultiplier. The results of measurements of radiation waveforms and the applied voltage on the lamp are shown in Fig. 5. Although the applied voltage was slightly distorted, it was close to a sinusoidal wave. Radiation was observed in the period when the voltage was increasing and decreasing, and it is noted that there was a break in the radiation between these two periods. Also, the radiation waveforms of increasing voltage and decreasing voltage were significantly different because of subtle differences in the voltage waveforms. This is a peculiar phenomenon attributable to dielectric barrier discharge. The peak level of the radiation waveform was about 8.4 times that of the temporal average of the radiation waveforms.

3.3 Distance from Output Window and Irradiance

The spatial distribution of the irradiation by the head-on type Kr_2^* excimer radiation exposure system was measured. A sensor having an aperture of 1mm diameter, sodium salicylate, and a silicon photo-diode was used for measuring. Fig. 6 shows the irradiance distributions of the orthogonal sections on the central axis of the lamp in the cases of the distance from the radiation outlet window being 0, 25, 50, 100, and 200mm. The space between the system and the sensor was filled with nitrogen gas. The irradiance was shown in relative intensity in which the maximum level in the case of 0mm of distance between the light outlet window and the sensor was taken as 100. Since the radiation along the axial line of a coaxial, hollow structure of the lamp was measured, the radial distribution was caldera-shaped when the distance from the light outlet window was small. While the distribution became even when the distance became larger. The diameters of the planar circles in which the unevenness of the irradiance became less than $\pm 5\%$ were 15mm and 24 mm at distances

from the light outlet window of 50mm and 200mm, respectively. The irradiance of the head-on type Ar₂⁺ excimer radiation exposure system was almost the same because of the same lamp shape.

The relationship between the distance from the lamps and the irradiance with the head-on type Ar₂⁺ and Kr₂⁺ excimer lamps is shown in Fig. 7. The abscissa shows the distance from the head of the discharge space z (the distance from the head of the lamp or the light output window can be obtained by $z-60\text{mm}$) and the ordinate shows the irradiance on the central axis of the lamp. The absolute value of the irradiance was obtained adopting the Xe₂⁺ lamp for which the absolute irradiance was known as a standard and assuming that the quantum efficiency of sodium salicylate is constant at 100 - 200nm⁹. The solid lines in Fig.7 show the calculated irradiance $L(z)$ that were obtained by the following two-dimensional equations. These equations were driven from an assumption that a perfect diffuse light radiates from the circular plain surface of the head of the discharge space ($z = 0, R = 9.6$) toward the direction of the z axis.

$$L(z) = 2 B \cos^2 \theta / (R^2 + z^2) \quad \dots\dots\dots (1)$$

$$\theta = \tan^{-1}(R/z),$$

Fig.6 Radial irradiance distributions of the head-on type Kr₂⁺ excimer lamp at different distances from the output window.

Fig.7 Axial direction irradiance for head-on type Ar₂⁺ and Kr₂⁺ excimer lamps as functions of distance from the discharge space.

Where B was the practical, effective light intensity, and the level of B was decided to fit the measured level of the irradiance matched $L(z)$ at $z = 260\text{mm}$. The square of $\cos \theta$ reflects a consideration on the complete scattering characteristics of the light source as well as its incident angle to the sensor. The measured level nearly matched the calculated level at the large distances, while the former was lower than the latter at the small distances. Thus, it was clarified that head-on type excimer lamps radiate directional light. There was a difference in the light decrement properties between the Ar₂⁺ excimer lamp and the Kr₂⁺ excimer lamp, but the causes for it have not yet been clarified.

The irradiance at the distance of 50mm was about 0.4mW/cm² in the Ar₂⁺ excimer radiation exposure system and 1.4mW/cm² in the Kr₂⁺ excimer radiation exposure system.

3.4 Temporal Changes of Light Output

The temporal stability of the light output of the head-on type Kr₂⁺ excimer radiation exposure system was measured. The light output reached 95% of that of the stable state within one second after the start of operation and it reached a stable state in about ten minutes. The light output after 10 minutes was recorded by a pen recorder with a response speed of 1.5 second resulting in drift below 1% in one hour and variation less than $\pm 1\%$.

4. Lifetime Property

The change in light output of the head-on type Ar₂⁺ and Kr₂⁺ excimer lamps by operating time is shown in Fig. 8. The time elapsed for a reduction of output to a half-level was about 500 hours in the Ar₂⁺ excimer lamp and about 1500 hours in the Kr₂⁺ excimer lamp. As a result of the analysis of the spectral transmittance, it is considered that the reduction in output was caused by deterioration of the MgF₂. The reduction due to the MgF₂ was generated in the whole below 200nm, and the profile of the spectral distribution remained almost unchanged.

Fig.8 Maintenance of irradiance for head-on type Ar₂⁺ and Kr₂⁺ excimer VUV exposure systems.

5. Influence of Absorption of Ar_2^* and Kr_2^* Excimer Radiation by Nitrogen¹⁰⁾

It has been known that there is an absorption band in nitrogen molecules called the Lyman-Birge-Hopfield (L-B-H in following) band between 100nm and 150nm. We have studied the level of influence of nitrogen gas on transmission the Ar_2^* and Kr_2^* excimer radiation.

The study condition was set so that the distance between a spectrometer and the light output window of a lamp was 250mm, and the spectral distribution in the cases of the light path filled with nitrogen gas or argon gas was measured. The equipment width of the spectroscopy was set at 0.42nm.

The measured spectral distributions are shown in Fig. 9, and the observed absorption peak wavelengths versus the L-H-B band are shown in Table 1. The absorption peak wavelengths in the case of nitrogen gas-filled light path matched each wavelength of the L-H-B band. An absorption at the wavelength of 124nm with the Ar_2^* excimer radiation was observed in both the argon gas and nitrogen gas cases. It is considered that this was attributable to impurities in the light path since the absorption did not appear when the light path was shortened, but the absorbing gases have not yet been determined.

The whole absorption quantity due to the L-H-B band in the case of the 250mm light path filled with nitrogen gas was calculated based on the spectral distribution of Fig. 9 to obtain about 9.4% in the Ar_2^* excimer radiation and about 5.8% in the Kr_2^* excimer radiation. Also, the absorption inside the lamp holder or the absorption in the distance of about 10mm from the light outlet window of the lamp to the outlet of the lamp holder was calculated based on this result to obtain 0.39% and 0.24% of absorption by nitrogen in the Ar_2^* excimer radiation and the Kr_2^* excimer radiation, respectively.

It was known from these results that the loss due to absorption is small even if the inside of the lamp holder is filled with nitrogen gas. Also, it was revealed that, in the case of a distance of less than 250mm between the exposure system and an irradiated object, filling the space with nitrogen gas is sufficient for practical use, except in cases using the light of a narrow band within the L-H-B band, since the ratio of absorption by nitrogen gas is below 10%.

Fig.9 Absorption characteristics of both Ar_2^* and Kr_2^* excimer radiations in nitrogen and argon gases.

Table 1 Assignment of each peak observed wavelength to Lyman-Birge-Hopfield band of nitrogen.

Ref.11,12		Absorption peak wavelength observed (nm)	
Shift of L-b-H band	Absorption wavelength (nm)	Ar_2^* excimer radiation	Kr_2^* excimer radiation
0,0	145.0	145.2	145.2
1,0	141.6	141.8	141.8
2,0	138.4	138.6	138.6
3,0	135.4	135.6	135.6
4,0	132.5	132.8	132.8
5,0	129.9	130.0	130.0
6,0	127.3	127.6	127.6
7,0	124.9	125.2	-
8,0	122.7	122.8	-
9,0	120.5	120.8	-
10,0	118.5	118.6	-
11,0	116.6	116.8	-

6. Burst Operation of Excimer Lamp¹³⁾

There are many plasma display panels (PDP) that use xenon gas and excite the phosphor with the 147nm resonance line of xenon atoms. Since the central wavelength of the Kr_2^* excimer radiation is 147nm, which is consistent with the resonance line of xenon atom, it has started to be used for phosphor development and as a light source for evaluation. One of the important properties for the PDP phosphor is the decay time. A burst operation of the excimer lamp was developed for the purpose of measuring decay properties.

A block diagram of the operating power circuit is shown in Fig. 10. The burst operation was made by setting the frequency for the on and off timings and the duty cycle ratio at oscillator number 2 and transferring the operation stop signal to the gate and damper. This stop signal is synchronized with oscillator number 1 by the flip-flop. The extinction of the burst operation is made by simultaneous stops of the inverter and the lamp voltage oscillations by the rapid discharge of the electric charge of the resonance capacitor by the damper. The operation stop signal also outputted outside the circuit. Fig. 11 shows the operation stop signal, applied lamp voltage, and radiation waveform under burst operation. Measuring the radiation waveforms was made by the same method as described in section 3.2. The difference of periodic characteristics between the radiation and applied voltage waveforms are due to the sampling beat of the digital oscilloscope. The shapes of the waveforms of applied lamp voltage and radiation under the on-operation are the same as was shown in Fig. 5.

The enlarged waveforms of radiation at the time of extinction are shown in Fig. 12. Simultaneously with the operation stop signal, the lamp applied voltage got closer to zero and the radiation of the lamp disappeared about 15 μs after the operation stop signal. The last waveform at the time of extinction remained unchanged after repeated measuring. Also, the 1/10 decrement time of the last radiation in this measuring system was about 5 μs .

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