Development of 222 nm-UVC Lamps with Long-Term Stability for Disinfection in Occupied Spaces

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We developed 222 nm-UVC lamps for disinfection in occupied space. Since these lamps require an operation lifetime of several thousand hours, such critical characteristics as radiation spectra must be investigated after long-term operation. In this study, we investigated the output, spectral changes, and filtering characteristics after long-term operation of filtered 222 nm-UVC lamps and confirmed just minimal changes in the lamp spectrum even with continuous operation over 8000 h. We found no changes in the harmful ultraviolet ratio: 235 ~ 320 nm integrated intensity/200 ~ 230 nm integrated intensity. Lifetime tests continue, and a longer life expectancy is expected. We also explained the light distribution and dimming functions of next-generation lamps. This new source has a wider light distribution, and dimming will enable continuous operation. We anticipate that this will allow more effective infection control.

Key Words: Excimer lamp, 222 nm, Disinfection

1. Introduction

The first study of excimer emission in dielectric barrier discharge was reported by Tanaka in 1955.[1] In the 1980's, full lamps were developed by G. A. Volkoba[2] and U. Kogelschatz with double cylindrical excimer lamps.[3] In 1993, commercialized double cylindrical excimer lamps were developed with monochromatic light at 172 nm (Xe_2^*), 222 nm ($KrCl^*$), and 308 nm ($XeCl^*$) to optimize the electric field strength of discharges, enclosed gas species, gas pressure, and window materials.[4] Sosnin,[5] Matafonova,[6] and Clauss et al.[7] reported the bactericidal performance of 222 nm double cylindrical excimer lamps. For such applications as UV-ozone cleaning for liquid crystal display manufacturing, high-power, flat-type lamps were developed that can achieve power of 170 mW/cm^2 or more with Xe_2^* excimer lamps with a wavelength of 172 nm.[8]

Recently, filtered far-UVC excimer lamps (222 nm-UVC lamps) are being used for disinfection in occupied spaces. These highly safe lamps remove wavelengths longer than 235 nm, which are harmful to humans. Studies of 222 nm-UVC lamps have shown an excellent germicidal performance without causing acute or chronic harm to the skin and eyes after irradiation.[9-17] No erythema, an acute disease, appeared after a single irradiation of 500 mJ/cm^2 and 1000 mJ/cm^2 was applied to normal mouse skin. Nor did erythema occur when the normal skin of 20 human volunteers was irradiated at 50 ~ 500 mJ/cm^2.[11] In just one person, normal human skin was irradiated up to 18000 mJ/cm^2; however, there were no changes to the skin up to 1500 mJ/cm^2, and erythema did not occur until 18000 mJ/cm^2.[12] Skin cancer, which is a chronic disease, did not develop after multiple irradiation cycles of 500 mJ/cm^2.[13]

In terms of eye safety, we confirmed that the acute disease of photokeratitis did not occur in rats after they were subjected to radiation up to 1500 mJ/cm^2.[15,16] Furthermore, repeated irradiation of 500 mJ/cm^2 in mice did not cause such chronic diseases as cataracts, neoplasms, or pterygium.[12] Concerning bactericidal and virus-inactivating performance, it was reported that a 222 nm-UVC lamp had an approximately identical inactivation speed as a 254 nm-UVC lamp for vegetative bacteria, and the inactivation speed was over twice as fast for spore-forming bacteria and some viruses.[17]

We have developed a 222 nm-UVC lamp for this application. Important lamp specifications for disinfection in occupied space include high efficiency, compactness, and a simple structure for automated mass production. The lamp module consists of four compact single-tube KrCl excimer lamps with reflectors, and we developed a lamp housing and a band-pass filter (Care222) for those reasons.

Environmental infection control involves repeated daily irradiation for eight hours or more in occupied spaces. The most important performance criteria is the long-term stability of lamps and filters. So far no data about the lifetime performance of such 222 nm-UVC lamps have been published. We investigated the UV output, the spectrum changes, and the filtering characteristics of the Care222 lamp module after its long-term operation. Furthermore, we investigated the filter's harmful spectrum cut-off characteristics after long-term use as well as the potential variations in these characteristics that can arise from the production process.

2. Materials and Method

2.1 Single-tube KrCl excimer lamp

A quartz glass tube with high transmittance at 222 nm was used for the tube, resulting in a small, approximately 70 mm-long lamp and an outer diameter of approximately 6 mm for a Care222 module. Such KrCl lamps have higher efficiency and are more compact than double cylindrical and flat type lamps. In addition, an automated production line can be constructed for a simple shape, and we expect that even if demand explodes, the production capacity can easily be scaled to very high quantities. Kr and Cl_2 gas were enclosed inside the tube, and the KrCl excimer emission was managed through a dielectric barrier discharge system using high-frequency and high-voltage pulses. Aluminum was used for the electrodes based on its good conductivity and UV reflectivity.

2.2 Band-pass filter

We used a filter with a transmission-type dielectric multilayer film of SiO_2/HfO_2, which transmits light at 222 nm and removes the harmful wavelength range of approximately 235 ~ 340 nm. We used the filter to remove harmful UVC and UVB spectra with wavelengths over 235 nm. The KrCl spectrum with a peak of 258 nm, which continues to 270 nm, must be sufficiently removed because of the possible danger to skin and eyes. The spectrum with a peak at 222 nm should be transmitted as much as possible.

2.3 Care222 module

The structure and specifications of the Care222 lamp in this study are shown in Fig. 1 and Table 1. The lamp module consists of four single-tube KrCl excimer lamps with a main wavelength of 222 nm, electrodes, reflectors, UV-resistant plastic lamp housing, and a band-pass filter. The lamp module is operated by a Care222-specific inverter (PXZ120I2) with input of DC 24 V and input power of approximately 11 W.

2.4 Method

The Care222 modules described above were operated from 0 to 8000 h or more, and the changes in the optical characteristics were observed. We measured the optical characteristics for the lamp only (an unfiltered Care222 module with four lamps), the filter only, and the Care222 module. A multichannel spectrometer (QEPRO, Ocean Insight), which uses a cosine collector diffuser plate (CC-3-UV-S), was placed at 50 mm to measure the spectrum at a UV wavelength of 200 ~ 360 nm. The angular light distribution was measured at 1 meter in 5° intervals using a UV meter (UIT-2400, Ushio, Inc.) that was calibrated at 222 nm. The UV transmittance of the filter and the lamp tube at each operating time was measured using a spectrophotometer (V-7200, JASCO Corporation) in a wavelength range of 200 ~ 360 nm. For comparison, the UV transmittance was also measured for a filter with a different design from the Care222 filter.

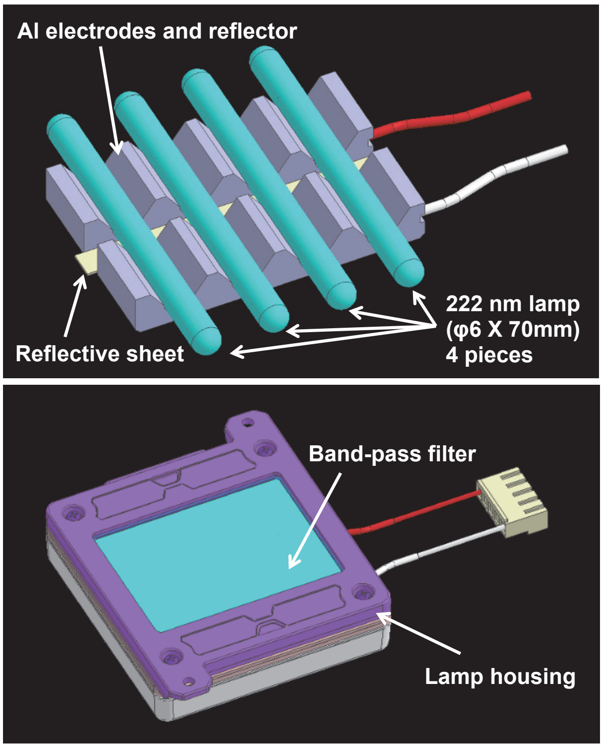


Fig. 1 Care222 lamp module structure.

Table 1 Care222 lamp module specifications.

Power consumption	Peak wavelength	UV intensity*	Lamp module size	Window size
11 W	222 nm	2.5 mW/cm^2	Approx. 80 × 75 × 25 mm	Approx. 60 × 45 mm

*50 mm from the window

3. Result and Discussion

3.1 Initial performance

Spectrum and irradiance distribution of lamp

The spectrum of the lamp without a filter (where the vertical axis is defined as 50% of the maximum 222 nm intensity in the linear range) is shown in Fig. 2. We confirmed a spectrum with a peak of 258 nm, which may be dangerous to the skin and eyes. Fig. 3 also shows the irradiance distribution of an unfiltered lamp at various output angles. Since it radiates UV at various angles, the spectrum needs to be investigated not only at an incident angle of 0° but also from 0° to 70°.

Transmission characteristic of filters

Figure 4 shows the transmission characteristic of a Care222 filter at various incident angles. It removes a wide range of wavelengths from 235 to over 320 nm at an incidence angle of 0°. It also removes the UVB region (280 ~ 320 nm) at an incident angle of 0° and even 270 nm at higher incident angles up to 70°. Ensuring high transmittance at 222 nm requires a filter characteristic that efficiently removes longer wavelengths at higher angles. Increasing the number of layers in dichroic filters can make the cut characteristics steeper, but this increases

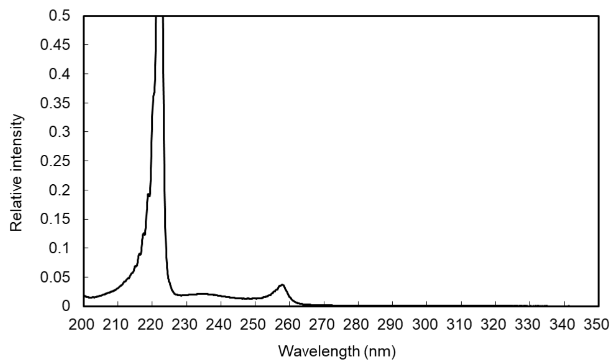


Fig. 2 Care222 lamp (no filter) spectral characteristics.

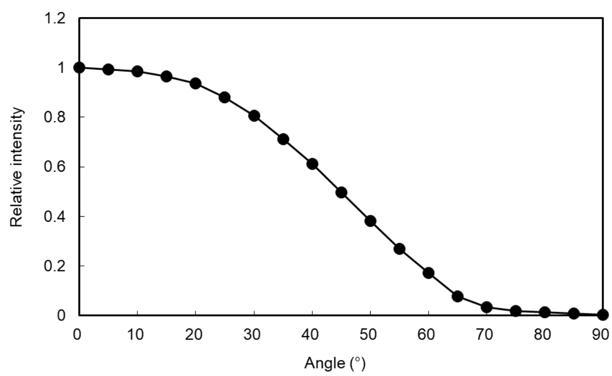


Fig. 3 Unfiltered Care222 lamp irradiance distribution.

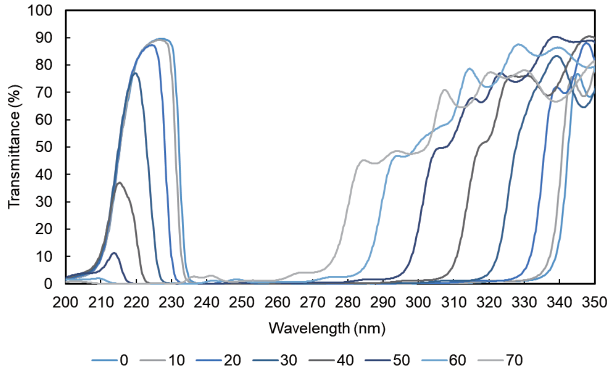


Fig. 4 Transmission spectrum of Care222 filter by incident angle.

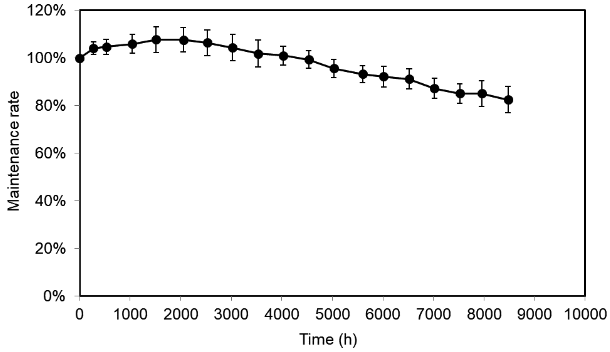


Fig. 6 Care222 irradiance degradation characteristics.

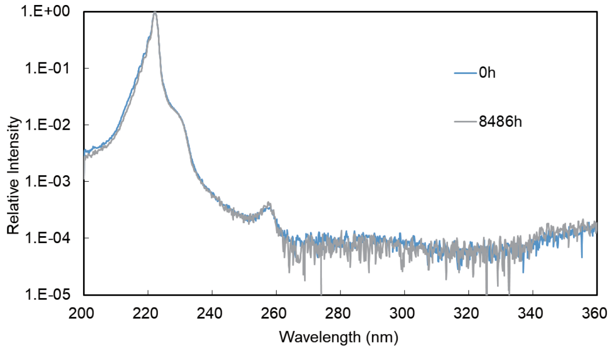


Fig. 7 Spectral distribution of lamp module at 0 h and 8486 h.

the film absorption and deteriorates the transmittance at 222 nm, so the optimal of the number of layers is extremely narrow.

Figure 5 shows the spectrum of a filter with narrower stop bands than the Care222 filter. Wavelengths up to 280 nm are removed at an incident angle of 0°, although the spectrum in the 258 nm range is not cut at incident angles over 50°. The cut characteristics are also not steep, so the transmittance of 222 nm is poor at a high incident angle. The harmful UV ratio (235 ~ 320 nm integrated intensity/200 ~ 230 nm integrated intensity) of the Care222 filter at 50 mm from the lamp was approximately 0.6%, whereas this filter was almost three times of that value. Furthermore the ratio will be 8% if a lamp module with a diffuser at high angles is used because it increases the risk of acute disease. This idea shows the need to carefully evaluate and report the characteristics of filtered 222 nm lamps.

3.2 Long-term stability

Care222 module (lamp and filter)

Figure 6 shows the irradiance degradation of the Care222 modules. The maintenance rate is defined as the ratio of the UV irradiance at each operation time to that at initial (0 h), and the data points represent the average of n = 20 samples ± standard deviation. The measurement results were obtained over 8000 h, and the maintenance rate was over 70%, which greatly exceeds current specifications. Currently, commercial products are operated with on and off duty cycles of less than 0.1 for eight hours a day. Based on the actual on-time, 8000 h equals 27 years of operation for applications at 70% output. However, since both the degradation of the irradiance and the ignition ability affect the lifetime, there is a need to separately confirm the ignition ability (non-operating).

Figure 7 shows an example of the spectral changes of the filtered Care222 lamp module. There were minimal spectral changes even after 8000 h, and the suppression of harmful wavelengths by the filter was maintained over its entire test time. Fig. 8 shows the ratio of the total intensity in the harmful UV wavelength range (235 ~ 320 nm) to that in the range of 200 ~ 230 nm over the lifetime. The harmful UV ratio was less than 0.6%, and a safe level was maintained. For example, the harmful UV output at a ratio of 0.6% was 0.13 mJ/cm^2 for an irradiation of 22 mJ/cm^2, 1.0 mJ/cm^2 for an irradiation of 160 mJ/cm^2, and 2.8 mJ/cm^2 for an irradiation of 479 mJ/cm^2. Even if the skin were irradiated with 479 mJ/cm^2, which is the revised biological exposure threshold for 2022,[20] the harmful UV level will not cause such an acute disease as erythema.

KrCl excimer lamp (lamp only)

We experimentally investigated the lamp factors that affect the deterioration of the irradiance of the Care222 module (lamp + filter). One is the slow escape of chlorine, which is an important part of the KrCl discharge. The second is the decrease in the transmittance of the discharge tube due to the UV irradiation. Third, we also confirmed the change in emission length. After that, we also conducted a transmittance deterioration ex-

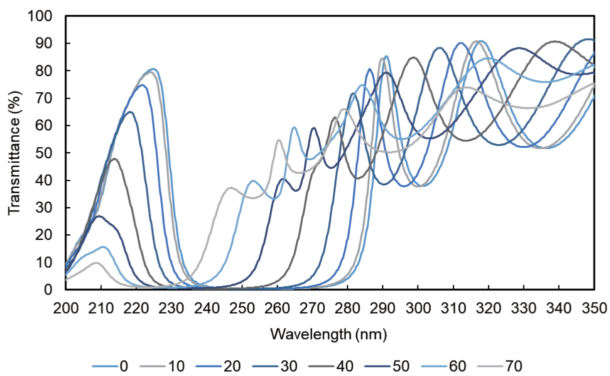


Fig. 5 Transmission spectrum of a filter with narrower stop bands than Care222 filter.

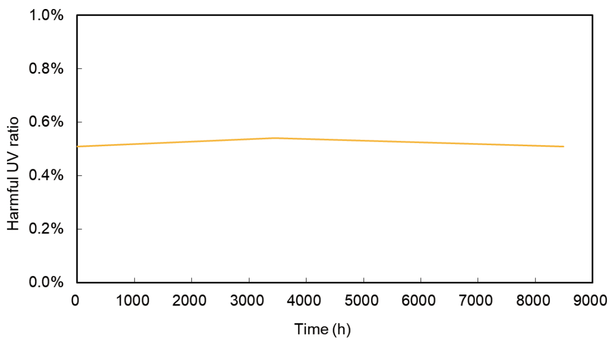


Fig. 8 Changes in harmful UV ratio during operating time (ratio of 235 ~ 320 nm integrated intensity/200 ~ 230 nm integrated intensity).

periment for the filter alone.

Figure 9 shows the spectrum of an unfiltered lamp in a wavelength range of 200 ~ 360 nm. Note that the vertical axis is shown on a logarithmic scale and the peak intensity at 222 nm was set to 1. The graph shows that the spectral intensity slightly increased with operating time mainly in the UVA region at 310 ~ 360 nm. However, this emission is an extremely small change in the intensity on the order of 10^{-4}. Because Kr_2Cl excimer emission occurs near 325 nm,[18] we assumed that the changes in the range of 310 ~ 360 nm between 0 and 8486 h are due to a change in the filling gas pressure. In particular, chlorine gas is filled with a small amount of pressure, and since it has high reactivity, if the energy is increased by the discharge, it will probably decrease due to a reaction with the SiO_2 tube or being injected into a SiO_2 tube.

To confirm the chlorine disappearance, we installed a KrCl excimer lamp with intentionally reduced chlorine pressure on the Care222 module and operated it until the UV maintenance rate was decreased to about 0%. This lamp's tube was observed in the depth direction by X-ray photoelectron spectroscopy analysis using argon ion sputtering, and chlorine atoms were injected up to about 100 μm inside the tube. On the other hand, the Care222 mass-produced lamp had a good maintenance rate even at 8000 h or more, and the intensity of the Cl_2^* emission spectrum (258 nm) was almost unchanged. This indicates that Cl_2 remains in the discharge space. Therefore, it has sufficient potential for even longer life.

Figure 10 shows the relationship between the gas pressure (proportional to the amount of chlorine) and the irradiance for the KrCl excimer lamp. Low gas pressure indicates a small number of Kr and Cl atoms, and the excimer radiation decreases. In contrast, an excessively high gas pressure reduces the collision energy, and as a result, the optimal energy for excimer generation cannot be reached. Therefore the graph has a peak irradiance at a certain gas pressure. Typically a gas pressure for achieving the highest output power would be selected, but to

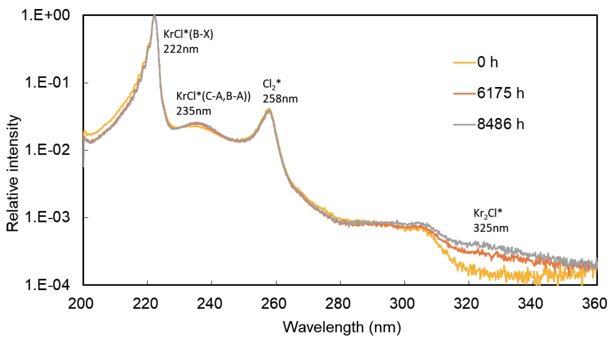


Fig. 9 Spectral changes of single lamp without filter.

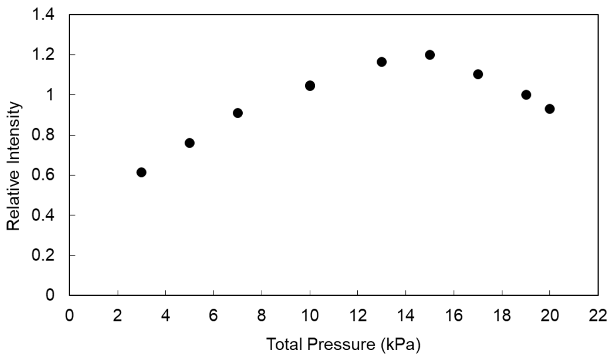


Fig. 10 Relationship between gas pressure and output power of 222 nm.

compensate for the Cl loss over time and to extend the product's lamp life, a higher initial pressure was chosen. The output of such a lamp over life is characterized by an initial small increase in output, after which it decreases.

Figure 11 shows the change in the Care222 quartz tube's transmittance, which decreased by approximately 5% after 4000 h of operation. The decrease in its transmittance near the 222 nm wavelength is mainly due to the absorption caused by the formation of the structural defects in the glass by UV irradiation.[19] Absorption by these defects is related to an absorption band with an E' center (≡Si·), which is an oxygen deficiency defect, centered at around 220 nm and generated by the UV–VUV irradiation of quartz. The tube material in Care222 was carefully selected to maintain transmittance at 222 nm and reduce such defects over extended operation time.

Figure 12 shows the discharge appearance of a Care222 module at 0 h and after 4000 h of operation. At 4000 h, the discharge appears longer, showing that it changed with operating time. The total output power is expected to increase with discharge length, which compensates to a certain degree the decline in output caused by other reasons. This effect can also be seen as a major advantage of this lamp design over other excimer lamp designs (e.g., cylindrical).

Band-pass filter (filter only)

Figure 13 shows the change in the transmittance of various filters (type A~E) at 222 nm. The vertical axis shows the relative transmittance at 222 nm compared to the initial transmittance. The changes in transmittance depend on the filter type, with a degradation of 5 ~ 20% after 8000 h. Fig. 14 shows the transmission spectrum of the B filter in Fig. 13 over the operating time of the Care222 lamp module. The transmission spectrum of around 222 nm decreased; no change in the blocking band above 235 nm was observed in the transmission spectrum. We infer that the decrease of transmittance at 222 nm over time is due to UV damage.

Regarding the filter, which is a critical part of the Care222, various filter durability tests were conducted at high-tempera-

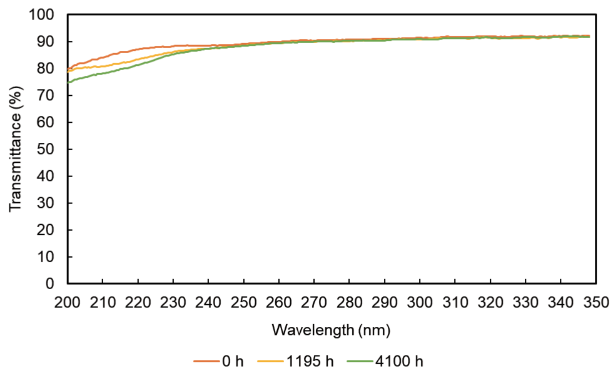


Fig. 11 Transmittance of Care222 quartz tube at various operating times.

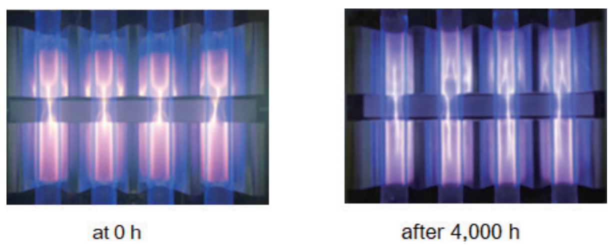


Fig. 12 Discharge status of Care222 (left: operating at 0 h, right: operating at 4,000 h).

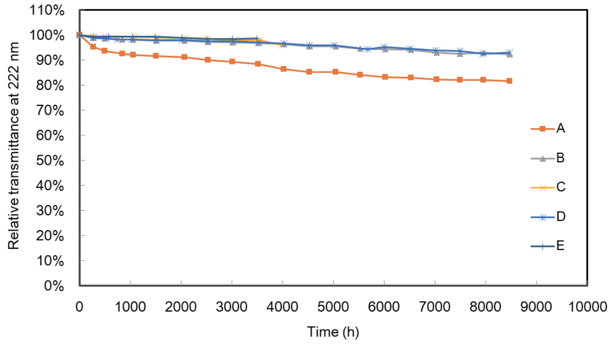


Fig. 13 Relative transmittance changes of filter at 222 nm.

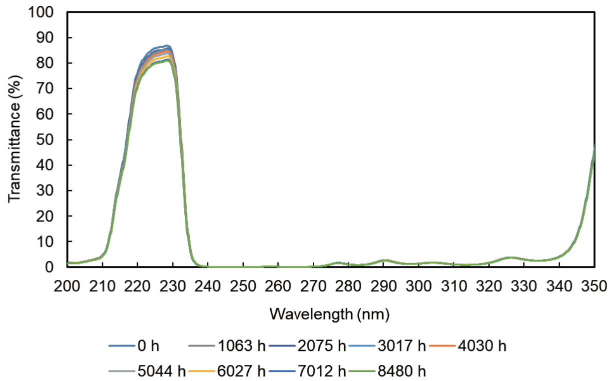


Fig. 14 Transmittance of Care222 filter at various operating times.

ture and high-humidity (temperature 85 °C, humidity 85% over 24 h), thermal shock tests (two cycles of 300 °C for 30 min and room temperature), and pure water boiling tests (10 min in pure boiling water). Depending on the filter manufacturing technology, we observed changes in the transmission spectrum that depend on humidity and other factors. The mass-produced filters for Care222 do not exhibit such spectral changes. However, the transmission spectrum changes in filters with a low-density multilayer filter using the conventional vacuum vapor deposition method. Fig. 15 shows an example of a filter whose transmission spectrum changes. The low density causes water absorption on the film, and when the lamp is operated in this state, the water is removed, and the cut wavelength shifts to the short wavelength side. Storage in an area with high humidity induces another shift to the long-wavelength side; this shift is dangerous. It can be prevented with a coating method that can form a high-density film instead of the conventional vacuum vapor deposition method.

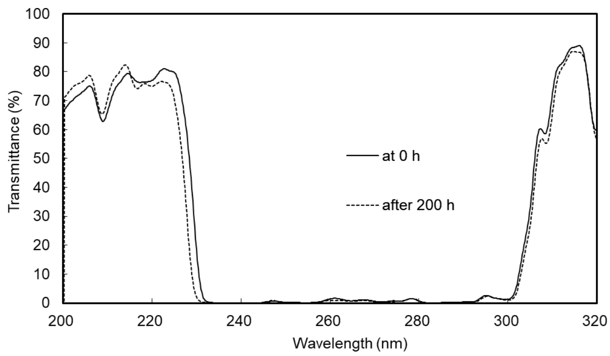


Fig. 15 Transmittance of a filter made with different manufacturing technology where wavelength shifted.

3.3 Spectral variation during manufacturing

We implemented a full spectral characterization of each lamp module in production to minimize the variations between lamp modules as much as possible and guarantee the specified output characteristics. Fig. 16 shows an example of the test results over the course of four months. Even the worst harmful UV ratio in all the lamp modules was less than 0.7%. As an ACGIH TLV-compliant lamp, the current standard should have a harmful UV ratio (235 ~ 320 nm integrated intensity/200 ~ 230 nm integrated intensity) of less than 1.5%, whereas the actual value of the present lamp is less than 0.7%. Based on the increased ACGIH TLV values, the lamp specifications and tolerance will be updated accordingly.

3.4 Next-generation lamp module

We showed the long-term stability in the Care222 lamp module, although the following issues remain with it: the beam angle of the current lamp module is narrow and the currently used on-off control in applications to control doses below ACGIH TLV 22 mJ/cm^2 is not optimal for certain target applications. We developed a next-generation lamp module and an inverter that can resolve these problems.

Table 2 compares the light distribution and performance between the current (B1) and next-generation model (B2) of the Care222 modules. The light distribution of the next model approaches perfect isotropic (Lambertian) radiation distribution using diffusing material after the spectrum filter. We also developed a new driver that allows dimming of the module and continuous operation in most applications when operating within ACGIH TLV dose limits.

4. Summary

We investigated the causes of lamp degradation and filter wavelength transmission characteristics over a continuous operating period of over 8000 h and showed that no spectral changes occurred over 8000 h of operating time. Lamp life tests

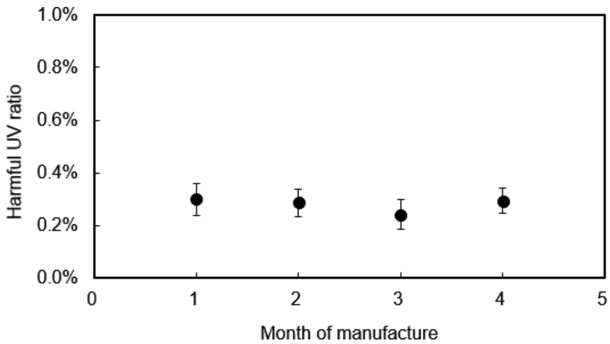
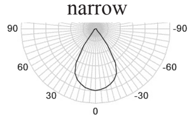
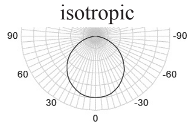


Fig. 16 Variation in harmful UV ratio during manufacturing.

Table 2 Comparison between current (B1) and next generation (B2) Care222 module.

Characteristic	B1	B2
Light distribution	narrow 	isotropic 
1/2 beam angle [degrees]	64	108
Irradiation @100 cm [µW/cm^2]	14.0	3.2
Operation	Non-dimming	4-step dimming (20, 40, 70, 100%)

continue, and we anticipate an even longer lamp life. If the lamp module is operated with on-off control to stay within the ACGIH TLV limits, the total on-time of 8000 h equals 27 years when irradiated eight hours a day. Furthermore, the filter underwent not only long-term irradiation tests but also high-temperature and high-humidity tests, confirming its stability. Variations in the lamp and filter during the manufacturing process have been minimized, and the spectrum is being checked for all lamp modules. We conclude that our filtered 222 nm lamp is safe for environmental infection control, has little variation among lamps, provides extreme stability over its life, and can be mass produced. Finally, the remaining improvements of light distribution and dimming functionality will be addressed with the next-generation lamps. The isotropic light distribution with continuous irradiation enabled by this new dimming functionality will enable new applications in the near future.

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