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Total Internal Reflection of Deep-Ultraviolet Light in a Water Waveguide and Its Application to Water Disinfection Technologies

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Abstract: We propose a new disinfection technology based on a novel concept involving the use of a small, deep-ultraviolet light-emitting diode (DUV-LED); the 265-nm DUV light is coupled to a running-water stream and is guided to a distant position without diffusion due to the total internal reflection of the DUV light inside the water stream. We demonstrate here the effect of the water waveguide disinfection technique by showing significant inactivation of a contaminated surface with indicator bacteria; this was verified by comparing the results of three disinfection methods: (i) disinfection with DUV light, (ii) disinfection with running water, and (iii) disinfection with the water-waveguide method. We believe that the marriage of the point-source nature of DUV-LED emission and the water-waveguide effect paves the way towards new applications such as water washing technologies that can reduce water consumption more than one order of magnitude without using additional chemicals in a simple manner.

Keywords: water; disinfection; bacteria; waveguide; deep-ultraviolet; light-emitting diode;

1. Introduction

Disinfection with ultraviolet (UV) radiation is a well-established technique that has been used widely, such as in the disinfection of water [1–10], room decontamination [11–15], and air purification. The wavelength of the UV radiation used is generally shorter than 280 nm, which places it in the UVC region (200–280 nm); this wavelength is selected because UVC radiation inactivates pathogenic bacteria, viruses and other microorganisms [16–22]. The inactivation is believed to occur via the formation of thymine dimers in deoxyribonucleic acid (DNA) by the absorption of UVC photons; the dimers prevent further replication of the DNA strains [23–29].

Low- or medium-pressure mercury vapor lamps have been widely used for many years for UV disinfection because they emit high-power UV radiation (on the order of 10 W) at a wavelength of 254 nm. This is close to the maximum absorption wavelength of DNA (approximately 260 nm) [1,3,30]. However, there are many drawbacks to using mercury vapor lamps; for example, the lamps contain highly toxic mercury, the lamps require fragile quartz glass tubes to seal in the mercury gas, a high operating voltage on the order of 1–10 kV AC is required, the lamps have a low plug efficiency of around 15–35%, long warmup times of approximately 10 min are needed, and the lamps generally have short lifetimes (on the order of 10,000 hours) [31].

Deep-ultraviolet light-emitting diodes (DUV-LEDs), where the emission occurs due to electron hole injection into the multiquantum well (MQW) semiconductor layer, have numerous advantages that may provide solutions to the above drawbacks of UV mercury lamps. Recently, advances have

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been made in DUV-LEDs based on aluminum gallium nitride (AlGaN) semiconductor materials [32,33], such as achieving a narrow emission spectrum that can be tuned between 210 nm (AlN) to 365 nm (GaN), a low operating voltage of the order of DC 10 V, a small emission area on the order of 1 mm², and instantaneous operation. However, the obtained output power (100 mW), external quantum efficiency (2–3%), and lifetime (10,000 hours) of the DUV-LEDs at the present stage have not yet reached their theoretical maxima; therefore, many studies have been conducted [31,34–36] to achieve an output power of 1 W, an external quantum efficiency of 50%, a lifetime of 100,000 hours, and a low price, as these characteristics are already available with GaInN blue LEDs.

Since the development of DUV-LED devices holds great promise, it is useful to consider not only their replacement of mercury vapor lamps but also their use in applications characteristic of LEDs. Various unique applications that typically use LEDs have been proposed for these devices [36–41], such as the sterilization of small medical devices [37,38] and germicidal DUV-LED lamps with multiwavelength radiations [39–41], as higher efficacies of inactivation of pathogens can be achieved with DUV-LEDs compared to the widely used mercury vapor lamps.

Here, we propose a new disinfection technology involving the use of a small DUV-LED device with 1 mW optical output power and running water. The concept is based on the optical coupling of DUV radiation into a water stream; by using the water waveguide effect, both physical disinfection by running water and photochemical disinfection by DUV light can be achieved simultaneously. (Here we note that the physical disinfection means that indicator bacteria are washed away from the contaminated surface.) The well-known water-waveguide effect can be seen in many educational and/or artistic video images of visible light traveling along with a water stream under internal reflection [42,43]. However, this effect is suitable not only for artistic demonstrations but also technically useful for disinfection. Because the absorption coefficient (α) of pure water in the DUV region is quite small ($\alpha = 0.013$ cm⁻¹ at 265 nm [44–46]); therefore, DUV light can be guided to a distant position without attenuation. Hence an intense DUV dosage can be obtained at a washing point with a low-power DUV-LED. Here, we demonstrate the effect of the water-waveguide disinfection technique by showing significant inactivation of a contaminated surface of a glass rod with indicator bacteria and show that small DUV-LED devices with output powers of the order of 1 mW are sufficient to inactivate the bacteria. We believe the marriage of the point-source character of DUV-LED light and the water waveguide effect paves the way towards various applications as a new water washing technology that could achieve significant reduction (more than 90%) in water consumption without using additional chemicals in a simple manner.

2. Materials and Methods

2.1. Culturing and Enumeration of Bacteria

Clinically isolated *Staphylococcus warneri* (*S. warneri*) was used for this germicidal experiment. The bacteria were cultivated in nutrient broth (E-MC63; EIKEN Chemical Co., Tokyo, Japan) at 37 °C for 20 hours. The concentration of 10^8 to 10^9 colony forming unit (CFU)/ml were used for the experiments. The head of a glass rod ($5\phi \times 270$ mm) was dipped in the suspension of *S. warneri*, and then this contaminated part was washed and/or disinfected by three methods: (i) disinfection with DUV light, (ii) disinfection with running water, and (iii) disinfection with the water-waveguide method. For enumeration, the disinfected part was streaked 80 times on nutrient broth agar plates. Colonies were counted after incubation for 24 hours at 37 °C. Plates yielding 1 to 500 CFU were considered for analysis. When the number of colonies were larger than 500 CFU, dilutions (10^{-1} – 10^{-2}) of the suspensions were made in order to obtain accurate analysis. All experiments were performed at least three times independently.

2.2. Theoretical Design and Experimental Setup for Water Waveguide System

The main components of the water waveguide disinfection system shown in Figure 1 are a TO-CAN type DUV-LED capped with a lens cap (VPT731 from Nikkiso Ltd. Tokyo, Japan), purified water supplied by a reverse osmosis (RO) system, and a rotating glass rod with a diameter of 5 mm.

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To perform the disinfections uniformly, the glass rod was rotated at 30 revolutions per minute during the disinfection tests (0–90 s). The emission wavelength of the DUV-LED was 265 nm with a spectral width (full width at half maximum) of 12 nm as measured by a spectrometer through an optical fiber (BIM-6002A, Brolight Technology Corporation, Hangzhou, China). The emission intensity of the LED was 0.5 mW at a forward voltage of 6 V and a driving current of 30 mA. The viewing angle of the 265 nm emission was determined by the DUV power as a function of distance between the LED and the irradiated area, and the power at the irradiated area was measured by a thermal surface-absorber-type power meter (PM16-401, Thorlabs Inc., Newton, NJ, USA). The viewing angle of the DUV emission was determined to be approximately 6°.

To introduce the 265-nm emission into the water stream, we used a T-shaped glass tube with an inner diameter of 6 mm. The flow rate of the purified water was maintained at 400 mL/min. Total reflection inside the water waveguide occurs at incident angles larger than 47° as estimated by Snell's law; the refractive index of water, n, of 1.36 for this wavelength region was used for the estimation [44–46]. Therefore, DUV-LEDs with viewing angles smaller than 120° can be used for the water-waveguide method. Here, we note that LEDs generally have randomly polarized emissions; therefore, the reflectance at the air-water interface becomes larger with increasing incident angle. However, due to the small value of the refractive index of water, only 6.5% of the light is reflected when the incident angle is 60°. Therefore, efficient optical coupling between the DUV-LEDs and the water waveguides can be achieved without difficulty. Another advantage of using the water waveguide is the small extinction coefficient (κ) of water, κ = 2.75 × 10⁻⁸, which corresponds to a small absorption coefficient α of 0.013 cm⁻¹ at an emission wavelength of 265 nm [44–46]. Therefore, almost all of the DUV emission can be guided; for example, theoretically, 85% of the DUV emission can be introduced to a disinfected glass rod 10 cm from the source.

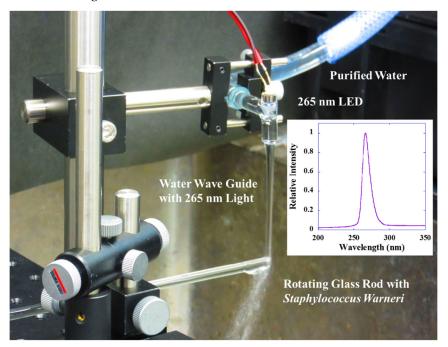


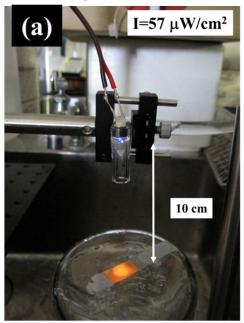
Figure 1. Water waveguide disinfection system. To introduce the 265-nm emission into the water stream, a T-shaped glass tube with an inner diameter of 6 mm was used. The emission wavelength of the deep-ultraviolet light-emitting diode (DUV-LED) was 265 nm, and its spectral width was 12 nm, as shown in the inset spectrum. Purified water supplied from a reverse osmosis system with a flow rate of 400 mL/min was used for the disinfection experiments. To perform the disinfection uniformly, each glass rod (after dipping the head in the bacterial suspension) was rotated at a speed of 30 revolutions per minute during the disinfection tests.

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3. Results and Analysis

3.1. DUV Fluence Measurements

The spectral intensity of 265 nm-DUV radiation at the head of the glass rod was measured by setting an optical fiber at the same place of the head and then the spectral intensity with or without the water stream was evaluated through the optical fiber and a spectrometer (BIM-6002A, Brolight Technology Corporation, Hangzhou, China). The DUV radiation power estimated by the intensity of the spectrum was 57 $\mu W/cm^2$ without the water waveguide and 143 $\mu W/cm^2$ with the water waveguide. The DUV radiation could be guided to a distant washing point (10 cm from the DUV-LED) with total internal reflection of the water stream; therefore, we could direct a 2–3 times higher DUV dose to the distant point compared to what could be achieved with simple DUV irradiation with the same disinfection time. The enhancement of the DUV light intensity due to internal reflection in the water flow is demonstrated by illuminating a phosphor tape (680MSH, 3M Japan Ltd., Tokyo, China) as shown in Figure 2.



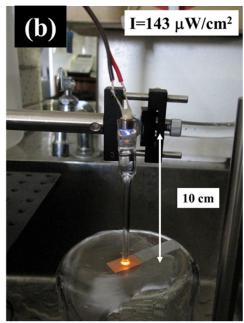


Figure 2. DUV fluence of the water-waveguide effect. The radiation power estimated by the intensity of the spectrum was (a) $57 \,\mu\text{W/cm}^2$ for DUV radiation without the water-waveguide effect and (b) $143 \,\mu\text{W/cm}^2$ for DUV irradiation with the water-waveguide effect. The enhancement of the DUV light intensity achieved with the water-waveguide effect is demonstrated by illuminating a phosphor tape.

3.2. Efficacy of Disinfection by the Water-Waveguide Method

To quantitatively investigate the disinfection rates of the water-waveguide DUV disinfection system, we performed three experiments: (i) disinfection with DUV light, (ii) disinfection with running water, and (iii) disinfection with the water-waveguide method. For each experiment, the head of a glass rod ($5\phi \times 270$ mm) was dipped in a suspension of *S. warneri*, and then, the head of the glass rod was disinfected by one of the above three methods. To perform the disinfections uniformly, the glass rod was rotated at 30 revolutions per minute during the disinfection tests (0–90 s).

Figure 3 shows the results of (a) the control (before the disinfections), (b) disinfecting with DUV light (57 μ W/cm²), (c) disinfecting with running water (400 mL/min), and (d) disinfecting with the water-waveguide method (143 μ W/cm² and 400 mL/min), where (b), (c), and (d) were performed for 1 min. The numbers of colonies counted were (a) 18,300 \pm 4831 CFU for the control plate, reduced to (b) 3510 \pm 1588 CFU by 1 min of DUV disinfection treatment, (c) 201 \pm 141 CFU by 1 min of disinfection with running water, and (d) 9.3 \pm 5.2 CFU by 1 min of water waveguide disinfection treatment. Here, the numbers reported are the means and standard deviations of CFU; dilutions (10-²-10-¹) of the

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suspensions for (a) and (b) were made for accurate counting. In particular, statistically significant differences were clearly observed between (c) and (d), and its probability value (P) was calculated as P = 0.034.

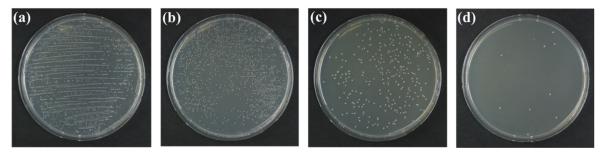


Figure 3. Results of disinfection for *S. warneri*; (a) control, (b) disinfecting by DUV light (57 μW/cm²), (c) disinfecting by running water (400 mL/min), and (d) disinfecting by the water-waveguide method (143 μW/cm² and 400 mL/min); the durations of treatments (b), (c), and (d) were 1 min. The number of colonies counted were (a) $18,300 \pm 4831$ colony forming unit (CFU) for the control plate, reduced to (b) 3510 ± 1588 CFU by 1 min of DUV disinfection treatment, (c) 201 ± 141 CFU by 1 min of the disinfection with running water, and (d) 9.3 ± 5.2 CFU by 1 min of water-waveguide treatment. Here, the numbers of CFU are reported as the means and standard deviations of at least three independent experiments. For counting the number of CFU of original (a) and (b) plates, dilutions (10^{-2} – 10^{-1}) of the suspensions for (a) and (b) were prepared. Significant differences were observed between (c) and (d), and the probability value (P) was found to be P = 0.034.

3.3. Theoretical Analysis of the Disinfection Rates

To quantitatively investigate the reduction of the disinfection rates as a function of duration, we plotted the relative ratio of the decrease in CFU (disinfected-CFU divided by control-CFU) caused by DUV light irradiation with an intensity of $57~\mu W/cm^2$ (purple circles), disinfection by running water at a flow rate of 400~m L/min (blue circles), and water-waveguide treatment with an intensity of $143~\mu W/cm^2$ and a flow rate of 400~m L/min (red circles), as shown in Figure 4. The reduction of the disinfection rates suggests that the decrease in CFU is not fitted by a mono-exponential reduction curve but is fitted by a multi-exponential curve (superposition of the mono-exponential reduction curve). It is evident that the multi-exponential reduction originates from variation between the environment of bacteria, because the DUV irradiation intensity and the adsorption strength that the bacteria sense are different from the position of the bacteria in the contaminated layer on the glass rod. These physical inhomogeneities affect the disinfection rates obtained by DUV light and/or running water. Thus, the relative rate of CFU reduction by the DUV irradiation, which is derived from the single-target model [47,48], can be expressed by the sum of the mono-exponential reduction curves with different reduction rate as

$$\frac{N_D(t)}{N_0} = \frac{1 - \exp(-\Gamma_D I t)}{\Gamma_D I t} \tag{1}$$

where N_0 is the number of initial CFU; $N_D(t)$ is the number of CFU obtained by the DUV irradiation; t is the duration of the disinfection process; Γ_D (cm²/mJ) is the photoinactivation rate, which depends on the environment of the bacteria and the irradiation wavelength; and I is the irradiation intensity of DUV light. Here we assume that the distribution of the reduction rates is uniform for the above calculation. To fit our results of the relative reduction rate of CFU by DUV irradiation, we used Γ_D = 2.10 cm²/mJ, and I = 57 μ W/cm². The theoretically derived reduction rate described by Equation (1) is shown as a solid purple line. We note here that the value of Γ_D obtained here is about three times smaller than the value previously reported [49]. We consider that this difference originates from the shadow region (back side of the glass rod) when the glass rod is irradiated by the DUV radiation.

The theoretical rate of CFU disinfection with running water can be analyzed based on the kinetics of surface cleaning [50–52]. By taking the inhomogeneity in the disinfection process of running water into account, we find that the theoretical rate can be expressed as

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$$\frac{N_W(t)}{N_0} = \frac{1 - \exp(-\Gamma_W J t)}{\Gamma_W J t} \tag{2}$$

where Nw(t) is the number of CFU obtained by the running water treatment; Γw (cm⁻¹) is the disinfection rate of bacteria adsorbed on the surface of the glass rod, and J (cm/s) is the velocity of the water flow. To fit the results of infectivity by the disinfection with running water, we used the values J = 34.0 cm/s and $\Gamma w = 0.05$ cm⁻¹. The theoretically derived relative rate of CFU obtained by these values is shown as a solid blue line.

The combined relative rate of the decrease in CFU by the water waveguide treatment can be expressed by the convolution function both of the photochemical disinfection and the running water given by Equations (1) and (2) as

$$\frac{N_{DW}(t)}{N_0} = \left(\frac{1 - \exp(-\Gamma_D I t)}{\Gamma_D I t}\right) \left(\frac{1 - \exp(-\Gamma_W J t)}{\Gamma_W J t}\right) \tag{3}$$

where $N_{DW}(t)$ is the number of CFU obtained by the water waveguide treatment. Here we used $I=143~\mu W/cm^2$ as the intensity of the DUV light. The same values, except this DUV intensity, were used for the fitting. The theoretically derived relative rate of the decrease in CFU obtained from these values is shown as a solid red line. The fitted curves agree well with the experimental results, and this agreement demonstrates that the water-waveguide is an efficient disinfection method that combines both physical disinfection with running water and photochemical inactivation.

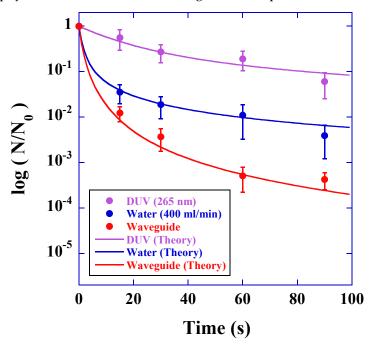


Figure 4. The relative ratio of the decrease in CFU for *S. warneri* (disinfected-CFU divided by control-CFU) achieved with DUV light irradiation with an intensity of 57 μ W/cm² (purple circles), running water treatment with a flow rate of 400 mL/min (blue circles), and water-waveguide treatment with an intensity of 143 μ W/cm² and a flow rate of 400 mL/min (red circles). The theoretically calculated lines express the rate of the decrease in CFU by DUV irradiation (purple line), running water treatment (blue line), and water-waveguide treatment (red line).

4. Discussion

The time requirements for 1-Log (10^{-1}) to 4-Log (10^{-4}) reduction levels by DUV light, running water, and water-waveguide methods obtained by the experimental setup (shown in Figure 1) were theoretically estimated based on the experimentally determined parameters of Γ_D and Γ_W . The calculated results are presented in Table 1. The results show that higher reduction levels can be realized in a short period of time by using the water-waveguide method. For example, in order to obtain 3-Log level reduction, the duration of 590 s is required for running water treatment; however,

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it can be shortened to 44 s by using the water-waveguide method, leading to save 90% water consumption. More water consumption can be saved for higher Log reduction levels by using the water-waveguide method. Therefore, the water-waveguide treatment is a promising way for obtaining both a higher reduction levels and less water consumption.

Table 1. Time requirements to obtain the 1-Log to 4-Log reductions by DUV light, running water, and water-waveguide disinfection methods for *S. warneri*. The unit used here is seconds.

Disinfection Methods	$Log (N/N_0) = -1$	$Log(N/N_0) = -2$	$Log(N/N_0) = -3$	$Log(N/N_0) = -4$
DUV Irradiation	8.3×10^{1}	8.3×10^{2}	8.3×10^{3}	8.3×10^{4}
Running Water	5.9	5.9×10^{1}	5.9×10^{2}	5.9×10^{3}
Water Waveguide	3.6	1.4×10^{1}	4.4×10^{1}	1.4×10^{2}

Here we note that special care should be taken so that the lens of the DUV-LED is not immersed in water. This is because the T-shaped glass tube for obtaining the water waveguide effect is made of borosilicate glass material whose absorption coefficient at 265 nm wavelength region is large, more than 10 cm^{-1} [53,54]. Figure 5a shows that when the lens is not immersed in water, the DUV emission with incident angle of 3° in air refracts to 2° in water. The first total reflection point (TRP) can be taken to a distant position (66 mm in our case), and there is no absorption material at this position, hence the efficient total reflection occurs. However, as shown in Figure 5b, when the lens is immersed in water, the viewing angle of DUV emission becomes wider (30°) and the TRP moves closer to DUV-LED (4.3 mm in our case). The reflection occurs at the interface between water and the borosilicate glass. In this case, almost all the DUV emission transmits to the glass side (98%), where the DUV emission is almost absorbed due to the large absorption coefficient of the glass at 265 nm wavelength region. Therefore, we cannot obtain the DUV light waveguide effect with the water stream when the lens is immersed in water.

We have presented a new cleaning technique involving the use of water flow as the waveguide of DUV emission. The developed technique can be applied to various point-of-use treatments that can be installed in small spaces due to the small size of a DUV-LED, such as the disinfection of drinking water from taps. However, when we consider disinfecting drinking water, the water must be disinfected quickly (the time it takes for the water to move from the emission point of the DUV-LED to the end of the water stream (water waveguide region)). Therefore, a much higher intensity of DUV light is required. For example, assuming that the water flow rate is 100 mL/s, the water waveguide region is 10 cm, and the DUV light intensity in the water waveguide is constant, to inactivate Cryptosporidium parvum oocysts, the dose required for 3-Log inactivation is approximately 10 mJ/cm² [1,5,20,55]. In this case, the required output power of the DUV light would be 85 mW at an emission wavelength of 265 nm (we determined these values based on the germicidal effectiveness curve [1,3,30]). These values can currently be achieved by using a high-power DUV-LED with a much wider spatial distribution and a viewing angle on the order of 120°. Total reflection inside the water waveguide occurs at angles larger than 47°, which corresponds to an incident angle of DUV-LED emission from air to water being approximately 60°; at this incident angle, reflectance loss is estimated as only 6%. Therefore, 94% of the DUV emission can be coupled to the water waveguide.

The water waveguide does not have a perfect smooth surface but a fluctuating shape; therefore, the condition of the total internal reflection is sometimes broken, leading to occur weak scattering of the DUV emission from inside to outside of the waveguide. Furthermore, the bacteria itself becomes the DUV scattering source. We have to take care of this leakage of the DUV emission, which would not exceed the threshold limit level (TLV) for UVC exposure. For example, in the United States of America, the TLV is 6 mJ/cm² over an 8-hour period [1,56]. However, this DUV scattering can be protected by using commonly available transparent thin films such as polyethylene terephthalate (PET), because these films have large absorption coefficients at this DUV wavelength region.

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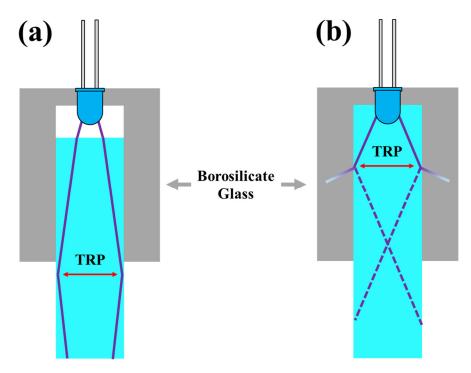


Figure 5. Change of the total reflection point (TRP) when the head of the LED-lens is (a) not immersed in water or (b) immersed in water. (a) When the lens is not immersed in water, the DUV emission with incident angle of 3° in air refracts to 2° in water. The first TRP point can be taken to a distant position (66 mm in our case), and there is no absorption material (borosilicate glass) at this position, thus the efficient total reflection occurs. (b) When the lens is immersed in water, the viewing angle of the DUV emission from the lens becomes wider (30°) and the TRP moves closer to the DUV-LED (4.3 mm in our case). The 98% transmission of the DUV emission occurs from water to the borosilicate glass and in this case we cannot obtain the water-waveguide effect of the DUV light.

5. Conclusions

We have successfully demonstrated a new water waveguide disinfection technique involving the use of a small, deep-ultraviolet light-emitting diode. The proposed water waveguide disinfection technique can be applied to many fields, such as washing techniques of skin, intraoral and internal-organ without the use of chemicals. The combination of the new technique demonstrated here powered by a small solar cell can provide a new convenient faucet for developing countries, which is possible to achieve efficient inactivation of a contaminated surface with less water consumption.

Author Contributions: T.M. is the co-first author. T.M., R.K and T.F. contributed to the design of the water waveguide disinfecting system and completed all the disinfecting experiments. I.T. and T.H. provided technical support and bacterial expertise for the bacterial growth and colony-forming experiments. T.M., I.T., and T.H. constructed the quantitative model of the disinfection results. R.K and T.F. performed the statistical analysis for the infectivity experiments. All authors read and approved this submitted manuscript

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Conflicts of Interest: The authors declare no competing interests.

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