Design and implementation of water purification system based on deep ultraviolet light emitting diodes and a multi-pass geometry reactor

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ABSTRACT

A novel reactor was designed and implemented for water purification using deep ultraviolet light emitting diodes (LEDs). The focus was on minimizing the number of LEDs required for effective germicidal action. Simulation studies were carried out on the flow of water as well as the irradiance of UV. Variation was made in the beam divergence of the UV sources and reflectivity of optical coatings used for photon recycling. Based on optimized reactor designs, water purification was carried out both in the static and flow-through configuration. Water from various sources was spiked with a known bacterial strain, exposure studies were carried out and germicidal effect was determined. Our results indicate that under optimal design, a 3 mL volume of water shows a three order inactivation using a single UV-LED in a static reactor in 180 s. For a flow-through geometry, only three LEDs were used in the reactor implementation, and a multi-pass procedure was used to purify 150 mL of water from an *Escherichia coli* CFU count of 4.3×10^4/mL to 12/mL. While slow, this process requires less than 2 W, and can be powered from rechargeable sources. Faster processes can be implanted using multiple such reactor units in parallel, and can be optimized to the requirement and power levels.

Key words | deep UV LEDs, water purification

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INTRODUCTION

While the use of ultraviolet radiation to de-activate pathogenic microorganisms using mercury lamps for large-volume water purification has been implemented for a long time (Wolfe 1990; Zimmer & Slawson 2002), the more recent development of UVC light emitting diodes (LEDs) (Crawford *et al.* 2005) has raised the prospect of small-volume, low-power, point-of-use water purification. This has opened up the possibility of personalized container-based water purifiers employing UV-driven germicidal action, powered by rechargeable and renewable electrical energy sources.

Ultraviolet light in a range of wavelengths has been tested by various groups, either singly or in combination, in order to determine their effectiveness in germicidal action (Vilhunen *et al.* 2009; Wurtele *et al.* 2011; Almquist *et al.* 2017; Oguma *et al.* 2018; Song *et al.* 2019). Wurtele *et al.* (2011) report that the germicidal efficiency peaks at 265 nm and radiation at this wavelength is most effective for arresting the multiplication of microorganisms. However, the wavelength of choice for water purification was found to be 275 nm (Bettles 2011) if both germicidal action and the efficient transmission of UV light through water were considered.

UV-LEDs in the wavelength range of 265–280 nm have been under development for over two decades (Mbonimpa *et al.* 2018), and have recently been commercialized. Typical UV-LED based germicidal reactors reported in the literature consist of a chamber with a number of such devices illuminating from the top, with a mechanical stirrer to ensure effective irradiation of the entire fluid (Lui *et al.* 2016;

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Nyangaresi *et al.* 2018; Jarvis *et al.* 2019). Disinfection studies based on 285 nm LEDs were extended for different water qualities and flow rates (Hessling *et al.* 2016). Furthermore, enhancement of efficiency with optimization of the geometrical arrangement of UVC-LEDs within the disinfection apparatus was also studied (Bowker *et al.* 2011; Oguma *et al.* 2016).

An important consideration that must be taken into account during the development of LED-based water purifiers is that despite concentrated effort by many groups, current germicidal UV-LEDs still produce power outputs in low mW levels, and the overall efficiency is far below that for blue or white LEDs (Hirayama *et al.* 2015; Moustakas 2016; Kneissl *et al.* 2019). This is due to inherent challenges associated with the III-Nitride materials, especially high Al-containing AlGaN alloys on which these LEDs are based. There are no native substrates for these materials, and heteroepitaxial deposition on to sapphire – the widely used substrate – leads to very high defect levels, thereby reducing wall-plug efficiency. Currently, AlN or GaN free-standing substrates are available, but they are smaller and significantly more expensive than sapphire. All of these make it necessary to design water purification systems that employ a smaller number of UV sources, and focus on optimal recycling of unused photons.

For water purification units designed for small-volume, low-power applications, such as personal portable water purifiers for field-locations powered by solar or other energy harvesting systems, a careful optimization of the overall reactor system design is necessary, taking into account not only the relatively low efficiency of UV-LEDs but also the power requirements for fluid circulation, system control, etc. In this paper we propose a versatile and scalable system for water purification based on UV-LEDs, where the throughput can be automatically matched to the available power.

EXPERIMENTAL METHODS

UV LEDs from LG Innotek (6060 series) nominally emitting at 278 nm with FWHM of 12 nm and a divergence angle of 121° were used as source for germicidal radiation in this work. The LED devices were mounted on a PCB using standard reflow solder and their electrical characteristics were measured using a Keithley 236 Source Measure Unit. Spectral response of the LEDs was determined using a calibrated setup including an Ocean optics USB 2000+ spectrometer sensitive to the 200–400 nm range and an integrating sphere. During this measurement a constant current source Keithley 6221 was employed to operate the LED. The angle-dependence of the output was measured by mounting the LED on a Newport rotational stage and collecting the emission at various angles using an optical fiber.

In this work, UV irradiations were performed for both static and circulating volume of water. Pure water (deionized (DI), 18 MOhm) was generated by a Millipore MilliQ system and further processed using a standard autoclave system. Five hundred μL of DH5 alpha strain of *E. coli* was mixed in 50 mL of Luria–Bertani (LB) broth and allowed to incubate at 37 °C at 110 rpm for 24 h before plating and further incubating for 24 h. A single colony (pure) was collected from the plate, mixed with 50 mL of LB broth and allowed to incubate for 24 h at 37 °C. To form the test-sample, 15 mL of pure water was spiked with 20 μL of this bacterial-broth and placed in the reactor to be subjected to UV irradiation.

Two sets of germicidal measurements were carried out during this work. The reactor A, as shown schematically in Figure 1(a), was based on a quartz cuvette of 3 mL capacity with four clear walls (MCQ-4). Optical transmitivity of water samples from different sources with varying level of dissolved and suspended impurities was measured using a Perkin Elmer 1050+ UV/Vis/NIR spectrophotometer. For reactor A, the cuvette was enclosed within an optical cavity formed by walls coated with AVIAN B paint from Avian Technologies. A single UV LED was placed on the top-opening of the reactor and the fluid contents directly irradiated. In this technique the photons were reflected back into the liquid both by the quartz–liquid interface as well as the UV-reflective paint.

For determination of effectiveness of the germicidal action, a 3 mL volume of the bacteria-spiked test-sample was filled in a reactor and exposed to UV radiation for bacterial inactivation, where the depth of water was 3 cm. At different times during the exposure, a test-sample of 250 µL was drawn, plated and incubated for CFU counting. Post incubation (incubation time 24 h), the number of

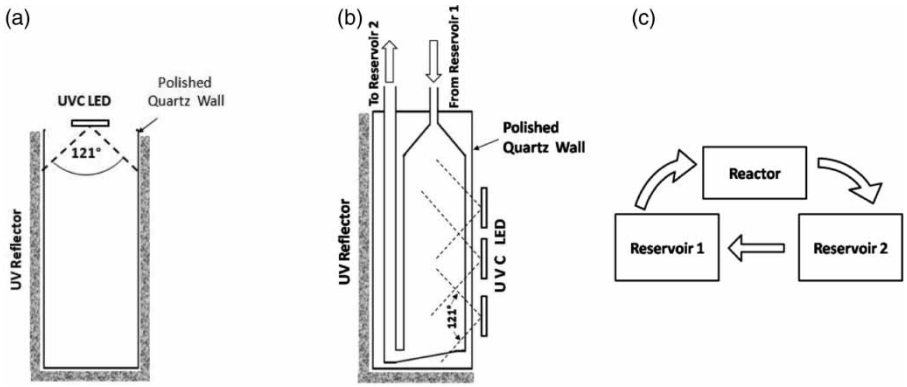

Figure 1 | Schematic of (a) Reactor A, (b) Reactor B (multi-pass geometry) and (c) purification scheme.

colonies per plate is a direct measure of the degree of disinfection by the UVC LED reactor. In order to study the effect of water quality on the germicidal action, water samples collected from various sources were irradiated and the results were compared.

In order to evaluate the effect of UV irradiation on water in a flow-through geometry, a germicidal Reactor B, whose configuration is shown schematically in Figure 1(b), was employed. An off-the-shelf flow-cell (STAR-71/Q/10) from Starna was used for this work. This system incorporates two ports for fluid input and output placed on the top face. The inlet consisted of a quartz tube with an inner diameter of 2.2 mm, which on entering the cuvette expanded through a pyramidal diffuser to over 8 mm. The fluid returns through a narrow channel of 10×1 mm cross section. Since we have restricted our focus on low-cost and portable solutions, which makes use of a very small chamber and small number of illumination devices, the water can be circulated through the chamber multiple times, and the germicidal action is based on the cumulative effect of the irradiation over a number of passes.

The passage of UV radiation through the system was simulated using a Monte-Carlo ray-tracing software TracePro. In order to understand the effect of the UV irradiance from different commercial LED systems, three different divergence angles (DAs) of 30, 60 and 120° were considered for simulation studies. To recycle photons and eliminate losses through the sidewalls, the coating of the back-wall was chosen to be either a white diffuse reflector or a 'perfect' mirror, and its effect on the irradiance was estimated. In addition, the flow of water through the complex path within Reactor B was simulated using COMSOL Multiphysics, which is a Finite Element Analysis solver.

In the experimental studies on flowing water, three LEDs emitting at 278 nm were connected in series to form the array and positioned on one polished and UV-transparent wall of the reactor. Photon recycling was enhanced by placing a diffused UV reflective coating on all the walls, as shown schematically in Figure 1(b). Water flow was carried out using a food-grade peristaltic pump, driven by a controlled DC voltage. In order to enhance the germicidal action a multi-pass process was used for this measurement. The same volume of water was passed through the reactor in multiple passes, while ensuring that no unexposed water remained at the end of each pass. A time gap of a few minutes was maintained between each pass, during which the UV LEDs were left in the on-state. In this method, we avoid the use of a mechanical stirrer, or a large UV-LED array. The water purification process is shown schematically in Figure 1(c).

The test for germicidal action was again carried out using water spiked with *E. coli* bacteria as described previously. A sample of 150 mL of water was mixed with 200 μL of bacterial broth, and passed through the system at a rate of 10 mL per min. After passing the entire volume of the test sample through the reactor, a volume of 250 μL was drawn for evaluation before repeating the entire exposure process. This was carried out for up to 10 passes, and samples drawn at the end of a pass were plated, incubated and analyzed for CFU counting. Thus, log inactivation after each pass was evaluated. This CFU counting technique was benchmarked using results from

the commercial water testing company Qualissure Laboratory Services (Kolkata, India).

RESULTS AND DISCUSSION

Simulation results

The flow Reactor B, as shown in Figure 1(b), was simulated using commercial software TracePro and COMSOL for the UV irradiance and the fluid flow respectively. The 3D model of the reactor is shown in Figure 2(a), indicating the important components. It is based on a rectangular quartz enclosure incorporating a water flow circuit where both the input and the output occur through tubes attached to the top face. The input chamber 'I' is $10 \times 40 \times 5$ mm in dimension and it is here that the germicidal action takes place in the presence of UV light. The output channel is a narrow tube and is marked 'O'.

The COMSOL microfluidics module was used to simulate the water flow through the reactor. A 'non-slip wall' was used as the boundary condition. The slice representation of flow rate though the entire path is presented in Figure 2(b). It was observed that while there is a high velocity and very strong non-uniformity within the narrow return tube, the flow in the inlet chamber is slow and very uniform. This is also supported by the 'streamline' data as obtained in Figure 2(c). The results allow a uniform residing time across the cross-section of the inlet chamber; and dosage can be calculated as a direct function of time for a fixed flow rate.

Figure 2(d) shows the 3D irradiance obtained from TracePro simulation results. The wall material was taken to be fused silica and the LED front surface was taken as a Lambertian source. The intensity distribution of three LEDs with beam divergences of 30, 60 and 120° has been studied. It should be noted that the UV-LEDs must be placed at a certain distance from the water, given by the thickness of the walls as well as the gap needed for their electrical isolation for safe operation. Thus, there is a considerable beam divergence by the time the UV light reaches the water surface in the form of a cone with a circular cross section. The results indicate that only the LED with 120° divergence creates a circular spot that fully spans the front face, and forms a high intensity cone, given in red, that nearly covers the entire depth of the water chamber. It can be also visualized that if all three LEDs had a divergence angle of 120°, they would together cover the entire front surface.

In this study the effect of the coating of the opposite wall on the irradiance properties has also been examined. Different optical coatings were placed on the wall opposite to the UV illumination, and its effect is seen by comparing Figure 2(d) and 2(e). It is observed that in the former case where a 'perfect' reflector was used in TracePro, the regions away from the direct path of the incoming beam were mostly dark. In contrast, when a 'diffused white paint' was used in Figure 2(e), the irradiation also reaches the 'shadow' areas and the overall uniformity is improved. It is clear that in the case of a smooth reflecting opposite wall, recycling of photons only occur by specular reflection. In the case of a rough reflecting opposite wall, the diffused reflection enhanced the uniformity of ray distribution within the entire reactor and hence a shadow zone was eliminated.

Experimental results

Testing of UV-LED and UV reflective paint

The UV LEDs (LEUVA66B00HF00) were tested electrically and optically before incorporation into the water purification systems. They exhibited a turn-on voltage of 5 V and an emission peak at 278 nm. Angle dependent studies indicated the LED viewing angle was 121° and the radiation pattern showed maxima at an angle of 30° from normal. An injection current of 10 mA was obtained at 6.5 V bias voltage, and this was used for all studies reported in this work. For this injection level – that is for an input power of 65 mW – the output UV optical power was measured using a calibrated spectrometer-based system and was found to be ~0.7 mW.

After coating the Avian B paint on an optically flat surface, a high diffuse reflectivity of 53–61% in the range of 250–280 nm was obtained using a spectrophotometer.

Reactor A (static water)

The germicidal action of UV radiation on static water was tested using Reactor A. Using the procedure described previously, a 3 mL pure water sample was spiked with *E. coli* and placed in the reactor. The effect of the UV irradiation

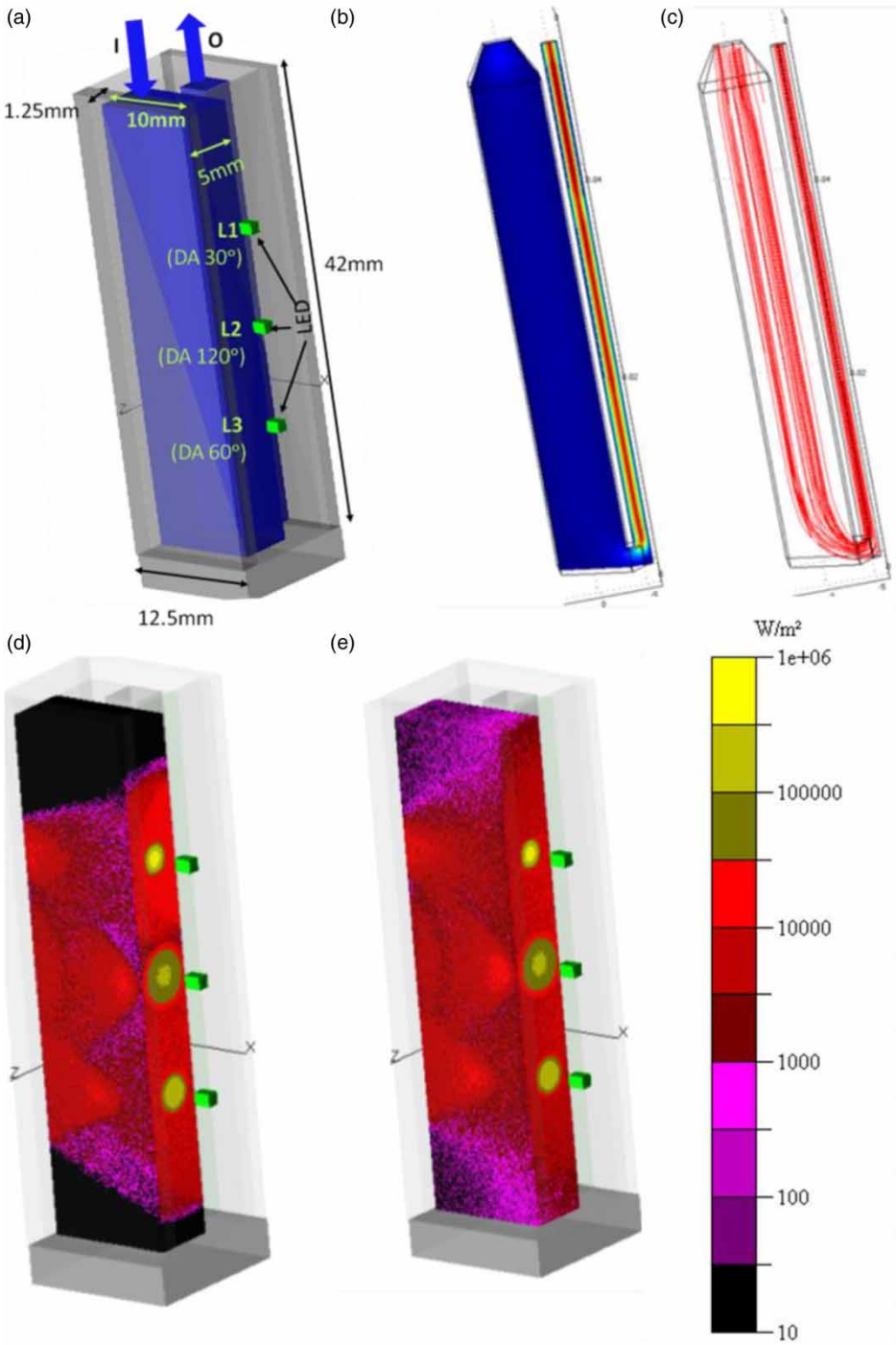

Figure 2 | (a) Schematic of UV irradiation Reactor B, (b) slice and (c) streamline representation of water flow using COMSOL Multiphysics; TracePro 3D irradiance results are shown for perfect rear mirror (d) and diffused white coating (e). Please refer to the online version of this paper to see this figure in colour: http://dx.doi.org/10.2166/wh.2020.008.

was tested by withdrawing samples periodically and carrying out standard CFU counting process. The results are shown in Figure 3.

While before exposure the CFU/mL count was found to be as high as 10^5, it was observed that only a 3 min exposure time is sufficient to reduce the count to 10^2 CFU/mL. From

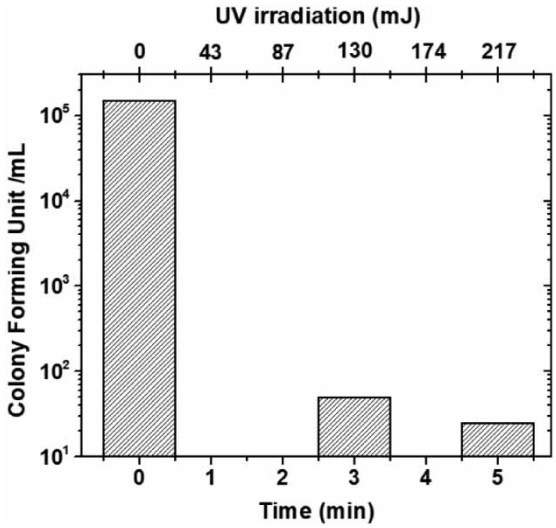

Figure 3 | Plating results as function of UV irradiation time for static autoclave/DI water spiked with DH5 alpha strain of *E. coli* in Reactor A.

the electrical and optical measurements on the LED devices, we calculate that an exposure of 130 mJ of UV radiation was sufficient to reduce the bacterial count of 3 mL of DH5 alpha spiked water sample by three orders of magnitude, and the electrical power requirement was 65 mW. Continued exposure causes a further reduction of bacterial colony counts, but the error margin in our measurements is too high to make a quantitative comparison for longer times.

It should be noted that the original water sample used in this experiment before spiking with bacteria was obtained from a pure source, i.e. 18 MOhm DI water, which was autoclaved before use. UV irradiation experiments were carried out on water samples collected from various sources, and the results are given below. These include: (a) DI/autoclave water, as before, (b) domestic tap water and (c) unfiltered river water. UV-transmitivity measurements were initially carried out using DI/autoclave water as the reference and the results are shown in Figure 4(a). It was observed that the UV-transmitivity of tap water was about 96% of that for DI/autoclave water at 278 nm, while that for unfiltered river water this value reduced to 67%. This can be attributed to different levels of dissolved solids, suspended particles and biological contamination.

Test-samples were formed upon spiking DH5 α bacterial strain of *E. coli* into these three types of water and were UV irradiated using the static Reactor A. As shown in Figure 4(b), it was observed that after plating and incubation the log inactivation for different water samples were not uniform. Reduction by a factor of 419 was obtained for autoclave water, but only by a factor of 7 for unfiltered tap water, and 14 for river water. These results may be linked to the presence of particulates and other contaminants which affect the transmission of UV light and therefore affect germicidal action. Even though the overall transmitivity was measured to be 96% for tap water, strong local variations may occur leaving unexposed volumes. The results comparing domestic tap water to river water need to be explained by other factors such as the possible presence of natural anti-bacterial elements. These results indicate that an initial particle filtration should be an integral part of the water purification system based on such weak UVC-LEDs.

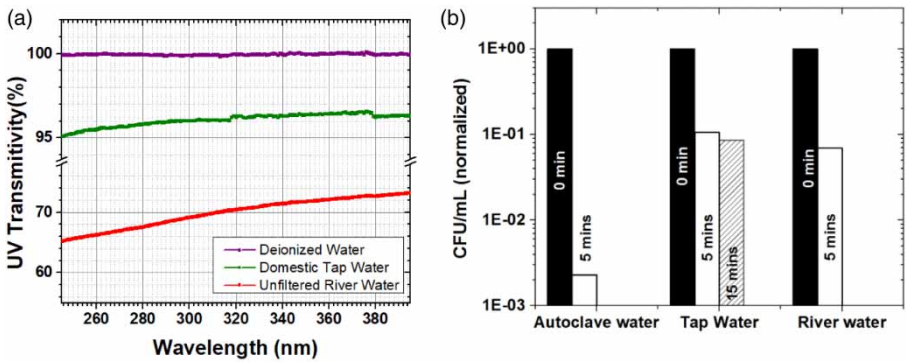

Figure 4 | (a) UV transmitivity through water from different sources and (b) CFU/mL values as function of UV irradiation time for three different types of water spiked with DH5 alpha strain of *E. coli*.

Table 1 | Culture results as function of UV irradiation passes for autoclave/DI water spiked with DH5 alpha strain of *E. coli* in a multi-pass geometry reactor

Sl. no.	Sample details	Test method	Characteristic	Results
1	Initial	APHA 23rd edn. 2017, 9222 I	*E. coli*/mL	4.3×10^4
2	Two passes			4.4×10^2
3	Six passes			3.8×10
4	Ten passes			1.2×10

Reactor B (flowing water)

The effect of UV irradiation on flowing water was tested using Reactor B, as described in the experimental methods above. The UV exposure was carried out for a total of 10 such passes, each pass consisting of about 13 min with a gap of a few minutes between passes, taking care that no fluid was retained in the lines at the end of each pass. The test sample was 150 mL of water spiked with DH5α and three UV-LEDs were used in the system. The electrical power was maintained at the same level during the entire irradiation process.

Samples were collected at the completion of the passes, which were then tested both in-house by the plating technique described previously and by M/S Qualissure Laboratory Services, which is a certified testing facility at Kolkata. Their report, which correlates well with in-house results, is shown in Table 1. The *E. coli* count per mL reduced from 4.3×10^4 to 4.4×10^2 in two passes, and finally to 12 in 10 passes, taking a total of 160 min. We have to stress that only three LEDs were employed and the total electrical power requirement was only 300 mW.

SUMMARY AND CONCLUSIONS

In this work reactors for static and flow-through geometry were implemented for the purpose of UV LED based germicidal action on *E. coli* bacteria-spiked water samples. The initial study of UV irradiation was performed on 3 mL pure (DI/autoclave) water inside static geometry and a reduction of three orders was obtained for UV irradiation of 130 mJ from a single LED source. These results were extended to study water samples from different sources using the same reactor geometry. Subsequently, the complexity of the reactor design was expanded for a flow-through geometry using a multi-pass process. Our experimental results indicate that for 150 mL of water deliberately spiked with a DH5 alpha strain of *E. coli*, a reduction from 10^4 to 12 CFU/mL can be obtained using only three LEDs.

This multi-pass geometry has many advantages, especially its low cost, effective mixing, stability, portability, and low power requirement. The limitation is that it takes longer for the process, which can be easily scaled down using additional LEDs, and a cost-time balance can be achieved, depending on the particular requirements. Furthermore, additional units can be placed in parallel to further enhance the process rate. This method has the advantage of real-time modification of process parameters by turning on or off these additional reactor modules according to the requirement and the electrical power available. This property of auto-scaling is important as it adds flexibility to the system, which can switch between a high-power high-throughput or a lower-power low-throughput mode depending on the level of disinfection needed and the electrical power available.

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