Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2021.26625

212 (2021) 71–77
February

Disinfection of biologically treated sewage using AlGaN-based ultraviolet-C light-emitting diodes in a novel reactor system

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Received 10 May 2020; Accepted 28 September 2020

ABSTRACT

The application of ultraviolet (UV) disinfection systems in water and wastewater treatment plants is highly recommended due to their high inactivation efficiency and absence of disinfection by-products. UV light-emitting diodes (UV-LEDs) are alternatives for UV mercury lamps due to its longer lifetime, improved efficiency, robustness, lesser start-up time, and environment-friendly. In this research study, the log inactivation of total coliform, fecal coliform, *Escherichia coli (E. coli)*, and fecal streptococci were determined in biologically treated sewage using AlGaN-based UVC-LED reactor system operated at a wavelength of 275 nm. A novel reactor system with a baffled arrangement of UV-LEDs was developed and evaluated to improve the inactivation efficiency. The performance of the laboratory-scale UVC-LED reactor was evaluated under batch mode. AlGaN-based UVC-LED reactor yields high log inactivation in terms of coliform removal, even in the biologically treated sewage of heterogeneous character. The experimental results showed that the log inactivation of total coliform, fecal coliform, *E. coli*, and fecal streptococci was 5.25, 5.45, 5.00, and 4.70 log, respectively. With improved reactor design, increased output power, and reduced cost, AlGaN-based UVC-LEDs could be scaled-up for full-scale application in sewage treatment plants.

Keywords: Biologically treated sewage; Coliform inactivation; Kinetics; UVC-LED disinfection; Wastewater reclamation and reuse

1. Introduction

Domestic wastewater treatment and reuse have been practiced in many countries and it is increasing significantly in recent years due to the scarcity of freshwater. The development of advanced treatment technologies for reuse of treated wastewater is essential to meet the demands of growing urbanization and industrialization [1]. Moreover, the stringent reuse standards mandate to reuse the treated wastewater for various purposes viz. irrigation, toilet-flushing, vehicle washing, fire protection, and recharge of aquifers. As per Indian standards, the permissible limit of fecal coliform for toilet-flushing and gardening (non-edible crops) should be nil and 230 CFU per 100 mL, respectively [2]. World Health Organization (WHO) standards for wastewater reuse (2006) say that the permissible limit of fecal coliform in treated sewage used for irrigation of crops likely to be eaten uncooked, sports fields, and public parks should be in the range of 200–1,000 CFU per 100 mL. Many wastewater treatments use biological technologies namely activated sludge process, sequencing batch reactor for the treatment of raw sewage [3]. After biological treatment followed by filtration, the treated sewage containing disease-causing pathogens must be disinfected before reuse.

Ultraviolet (UV) disinfection has been considered as a promising technology for the inactivation of pathogens due to its broad-spectrum [4]. UV disinfection is used in both centralized and decentralized water and wastewater treatment plants [5,6]. UV disinfection is recommended as

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a substitute for chemical disinfectants for water and wastewater treatment [7]. The main advantages of UV radiation over conventional chemical disinfection are the absence of harmful chemicals, no formation of disinfection by-products, no change of color, taste, and odor, no danger of overdosing, and low maintenance [5–9]. The major source of UV light inactivating bacteria, fungi, viruses, and protozoa is low or medium-pressure mercury vapor lamps; however, it has limitations viz., the toxicity of mercury, fragility, shorter lifetime (8,000–10,000 h), high operation cost, and fouling of UV lamps 10,11]. Therefore, an alternative and new UV light source – UV light-emitting diode (UV-LED) has been developed in the past decades to overcome the shortcomings of UV mercury lamps.

The light-emitting diode (LED) is a semiconductor device, made up of semiconducting materials to form *p–n* junction (hole and electron). The electrons and holes combine at the junction, produce electroluminescence, and emit board spectrum of light [6,12]. The wavelength of light emitting from LED depends on the type of semiconducting materials used. UV-LED has many advantages over mercury lamps namely environmental friendly (mercury-free), longer lifetime (1,00,000 h), high efficiency, less energy-intensive, multiple wavelengths, negligible warm-up time, flexibility in designing novel reactors, compact, and robust [6,10,13–16]. These aspects of UV-LEDs provide an attractive proposition over conventional mercury lamps for disinfection. Zhou et al. [6] reported that the maximum electrical to microbial inactivation efficiency could be achieved in the UV-LED system was 75%. UV-LEDs can be manufactured at different desirable wavelengths using semiconducting materials namely gallium nitride (GaN), aluminum gallium nitride (AlGaN), aluminum nitride (AlN). UV-LEDs containing GaN emit radiation at a wavelength of 365 nm (near UV), AlN emit radiation at 210 nm (deep UV) and AlGaN emit radiation at a wavelength range from 210 to 365 nm [15,17]. The unique feature of UV-LED is its ability to emit radiation at multiple specific wavelengths.

AlGaN based UV-LEDs can be utilized in water disinfection for effective and efficient microbial inactivation, as DNA (deoxyribonucleic acid) destruction is more favorable at the UV-C region [5]. UV-A (315–400 nm) radiation inactivates micro-organisms by producing reactive hydroxyl radicals through photo-oxidation of oxygen, whereas UV-C (200–280 nm) radiation destroys DNA directly by producing cyclobutane pyrimidine dimers (CPD) [5,13,18]. Therefore, the selection of wavelength is an important factor for germicidal inactivation; however, efficiency depends on the UV sensitivity of micro-organisms. The disinfection mechanism of UV mercury-vapor lamp and UV-LED is similar. U.S. Environmental Protection Agency (EPA) [7] and Vihunen et al. [19] suggested that UVC-LEDs having wavelength around 260–280 nm are most suitable for effective water disinfection as they not only destroy DNA but also damage enzymes responsible for DNA repair. The water quality parameters, namely turbidity, and total suspended solids (TSS) concentration influence the inactivation efficiency. The suspended solids shield/protect micro-organisms from UV irradiation, which can be removed by simple filtration [20,21]. Bowker et al. [14] determined the microbial UV fluence-response of three surrogate micro-organisms such as *Escherichia coli* (*E. coli*), male-specific coliphage (MS2), Phage T7 using a novel UV-LED system for 255 nm UV-LEDs, 275 nm UV-LEDs, and 254 nm low-pressure mercury lamps. They have reported that 275 nm UV-LEDs are more effective in microbial inactivation than 255 nm UV-LEDs. Nguyen et al. [22] evaluated the efficiency of the flow-through UV-LED (285 nm) system in domestic wastewater disinfection. It has been reported that 3.7 log MS2 coliphage inactivation was achieved at a flow rate of 10 mL/min. Several UV-LED light arrangements were studied for efficient reactor performance in water systems [23]. This study motivates to design a novel and compact reactor for disinfecting treated sewage for reuse. By considering the following parameters namely reactor hydrodynamics, radiation distribution patterns, and reactor kinetics, UV-LEDs are used to design novel UV disinfection systems for achieving maximum germicidal efficiency.

The objective of this research study is to develop and demonstrate a novel UVC-LED reactor system for disinfecting secondary treated sewage for reuse. The performance of the UVC-LED reactor system was investigated by considering the following bacteriological parameters viz., total coliform, fecal coliform, *E. coli*, and fecal streptococci.

2. Methods

2.1. Characterization of secondary wastewater treatment effluent

Secondary treated sewage was collected from the outlet of a pilot-scale biological treatment system at Anna University, Chennai, India. Total coliform, fecal coliform, *E. coli*, and fecal streptococci are considered as fecal contamination indicators. The samples were characterized as per the test procedure recommended in the Indian standard IS 1622:1981 [24]. The characteristics of biologically treated sewage after settling are presented in Table 1. Samples were analyzed for various water quality parameters, namely pH, TSS, turbidity, and bacteriological parameters namely total coliform, fecal coliform, *E. coli*, and fecal streptococci. The performance of the UVC-LED reactor was evaluated for a period of 1 month, out of which 20 d samples were presented in the graphs. All the samples were repeated for three times and obtained the results with a standard deviation error of less than 5%.

2.2. Reactor set-up and operation

The schematic representation of the AlGaN-based UVC-LED reactor is illustrated in Fig. 1. A lab-scale reactor was designed and fabricated using stainless steel material for a total capacity of 6.0 L. The dimension of the UVC-LED reactor was 40 cm × 15 cm × 10 cm. The working volume was 5.0 L. Four quartz tubes were inserted in the reactor system perpendicular to the direction of water flow. Each quartz tube contained 8 UVC-LEDs with a radiant flux of 12 mW, was connected to a power supply. The reactor was designed in such a way that the maximum light photons are to be used for microbial inactivation. UVC-LEDs were procured from Bytech Electronics Co., Ltd., China. The peristaltic pump was used only for filling the treated sewage into the reactor. All the experiments were carried out

Table 1
Characteristics of raw sewage and biologically treated sewage

Parameters	Raw sewage	Biologically treated sewage (before filtration)	Biologically treated sewage (after filtration)
pH	6.8–7.4	7.5–8.8	7.5–8.8
BOD (mg/L)	250–600	3–9	3
TSS (mg/L)	120–570	3–16	<2
Turbidity (NTU)	40–55	<5	<1
Transmittance		60%	88%
Total coliform (CFU/100 mL)	10^8	10^7	10^7
Fecal coliform (CFU/100 mL)	10^7	10^6	10^6
E. coli (CFU/100 mL)	10^6	10^5	10^5
Fecal streptococci (CFU/100 mL)	10^6	10^5	10^5

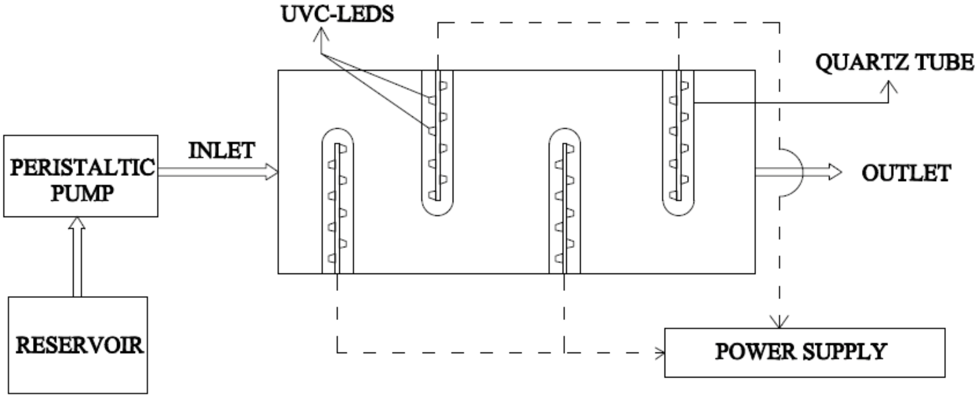

Fig. 1. Schematic representation of AlGaN-based UVC-LED reactor.

under batch mode. The biologically treated sewage samples were exposed to UVC-LED rays for 0, 30, 60, 90, 120, 150, 180, 210, and 240 s to achieve UV dose of 0, 5, 10, 15, 20, 25, 30, 35, and 40 mJ/cm^2, respectively. The disinfected samples were collected through an outlet port and were then characterized immediately. The technical specifications of the UVC-LED are presented in Table 2.

2.3. Characterization of UV-C LED

Fig. 2 illustrates the emission spectrum of AlGaN-based UVC-LED measured using a Stellarnet spectrometer (IIT Madras, Chennai, India); to investigate the ability of LED to perform disinfection studies. The peak wavelength of the emitted UV light was clearly observed at 275 nm. The FWHM of emission spectra was found to be in the range of 10–12 nm. The inactivation efficiency may depend not only on the UV light but also on the relaxation processes of the excited states, which in turn depend on the wavelength [25].

2.4. Analytical methods

The performance of the AlGaN-based UVC-LED reactor was assessed based on the log inactivation of total coliform, fecal coliform, *E. coli*, and fecal streptococci. The samples were characterized before and after irradiation by UVC-LED as per the test procedure recommended in IS 1622:1981 [24]. Microbial inactivation efficiency was experimentally determined by logarithmic inactivation, often expressed as $\log_{10}(N_0/N)$, where N_0 and N represents the number of micro-organisms present before and after UVC-LED irradiation, respectively. The microbial inactivation by UV irradiation could be described with first-order kinetics by carrying out the collimated beam tests, where optimal UV dose rate could be determined [4]. The inactivation kinetic parameter namely inactivation rate constant "*k*" (cm^2/mJ) was calculated using a disinfection model called Chick–Watson first-order linear model (Eq. (1)), which

Table 2
Technical specifications of UVC-LED

Parameters	Characteristics
Measured peak wavelength	275 nm
Full width half maximum (FWHM)	9.2 nm
Radiant flux	12 mW
Forward voltage	5–7 V
Thermal resistance	<10°C/W

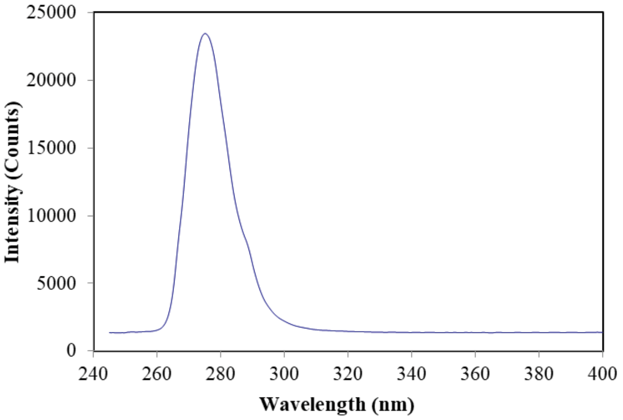

Fig. 2. Emission spectrum of UVC-LED showing a peak wavelength of 275 nm, measured using the Stellarnet spectrometer.

describes the linear relationship between log inactivation and UV dose applied [4,11].

$$\log_{10}\left(\frac{N_0}{N}\right) = -(k)D \tag{1}$$

where N_0 and N represent the number of micro-organisms present before and after UVC-LED irradiation, respectively, k represents inactivation rate constant, and D represents UV dose rate (product of the intensity of UVC-LED and exposure time). Some researchers observed two major deviations in the first-order UV disinfection model [26,27]. They observed no microbial inactivation at lower UV dose rates (offset), and a log-linear increase at intermediate UV dose rates. The microbial inactivation appears to be insignificant at higher UV dose rates (tailing effect) due to the presence of suspended solids, which shields micro-organisms. These discrepancies can be overcome by using another disinfection model called shoulder model (Eq. (2)) [4,15].

$$\text{DRF} = -\left[(k)(D)\right] - b \tag{2}$$

where DRF represents decimal reduction factor ($\log_{10}(N_0/N)$) and b represent negative y-intercept value.

2.5. Electrical energy efficiency

The electrical energy ($E_{E,N}$) is an important factor used to evaluate the performance of UV or UV-LED disinfection systems based on the consumption of electrical energy, which is defined as the amount of electrical energy (kWh/m^3) required to inactivate the pathogens by one order of magnitude [28,29]. The electrical energy ($E_{E,N}$) for a specific N-log reduction of microbes can be calculated using Eq. (3) [29].

$$E_{E,N} = \frac{A \cdot F_N}{3.6 \cdot 10^3 \cdot V \cdot C \cdot \text{WF}} \tag{3}$$

where $E_{E,N}$ denotes the electrical energy per specific N-log reduction (kWh/m^3/N-log reduction), A denotes the irradiant surface area (cm^2), F_N represents the UV dose required for N-log reduction (mJ/cm^2), V is the volume of the sample used (mL), C denotes the wall plug efficiency given by manufacturer, WF represents the water factor and the value of 3.6×10^3 is a unit conversion constant.

3. Results and discussion

3.1. Optimization of UVC-LED dose

Fig. 3 shows the log inactivation of total coliform, fecal coliform, *E. coli*, and fecal streptococci at different UVC-LED doses recorded at a wavelength of 275 nm. The tailing effect was observed for all four bacteriological parameters at a higher UV doses. The UVC-LED dose rate required for achieving maximum log reduction of total coliform, fecal coliform, *E. coli*, and fecal streptococci was 35, 35, 20, and 25 mJ/cm^2, respectively. The result shows that the log inactivation increased gradually with an increase in UVC-LED dose rate and it is observed that there is no further increase in log inactivation at higher UVC-LED dose rates, indicating a tailing effect. Zhou et al. [6] reported that longer irradiation time results in a higher UV dose rate, which could impart a lethal effect on the bacteria, thus improves the inactivation efficiency.

The UV sensitivity of the micro-organisms is described by inactivation kinetics. The most commonly used disinfection model for chemical disinfectants is the first-order Chick–Watson (1908) model, and the same model can be used for UV disinfection studies [4]. Based on the first-order model, the log-linear relationship between UV dose rate and log inactivation was developed. The inactivation rate constant k (cm^2/mJ) arrived from the disinfection models are summarized in Table 3, which could be used to design a full-scale UVC-LED disinfection system.

3.2. Disinfection efficiency of total coliforms

The variation of total coliform concentration in influent and effluent is presented in Fig. 4. The experiments were carried out for a period of one month to study the long-term performance of the UVC-LED reactor. The results showed that the AlGaN-based UVC-LED system is efficient and effective in inactivating the coliforms. The biologically treated sewage (before filtration) had suspended solids concentration of 3–16 mg/L and turbidity of <5 NTU. The presence of organic and inorganic particles in the UV feed could shield the microbes from UV irradiation, which decreases inactivation efficiency [6,30]. Thus, before UV irradiation, the treated sewage was passed into activated carbon filter at a flow rate of 30 mL/min, to reduce suspended solids concentration and turbidity to <2 mg/L and <1 NTU, respectively. The transmittance of treated sewage measured at 275 nm before and after filtration was 60% and 88%, respectively. The initial concentration of total coliforms present in the biologically treated sewage was found to be 10^7 CFU/100 mL. At the optimal UVC-LED dose of 35 mJ/cm^2, the average concentration of the total coliforms present in the effluent was 448 CFU/100 mL and the average log inactivation of the total coliforms were found to be 5.25 log. The concentration of total coliform in the effluent is found to be consistent over a month of testing. UV reactor design and emitted wavelength play a crucial role in microbial inactivation efficiency. In a conventional reactor, UV light source is placed at the top (away from water sample), the distance between water sample and UV-LED requires high output power to compensate the losses due to non-homogenous output power [15]. Therefore, in this study, a UVC-LED reactor was designed in such a way that the losses due to non-homogenous output power were eliminated by placing UVC-LEDs (inside quartz tube) within the reactor, having direct contact with the water sample. Quartz tubes containing UVC-LEDs are placed in a baffled manner and are perpendicular to the direction of water flow. This reactor design enables to inactivate the maximum number of micro-organisms, resulting in low total coliform concentration in the effluent. This new reactor design for efficient disinfection is the significant contribution of this study with respect to the conventional UV-LED systems.

3.3. Disinfection efficiency of fecal coliforms

Generally, fecal coliforms are considered as microbial contamination indicators. Fig. 4 illustrates the variation in fecal coliform influent and effluent concentrations recorded in the AlGaN-based UVC-LED reactor throughout the experimental period. The average concentration of the fecal coliforms in the effluent was 14 CFU/100 mL, and the log reduction of the fecal coliforms was 5.45 log at an optimal UVC-LED dose of 35 mJ/cm². The UVC-LED reactor system was found to be effective even there was a fluctuation in the influent characteristics. The negative effect due to suspended solids settling down in the reactor could be eliminated by proper filtration before irradiation, which helps to improve the performance of the reactor. In this UVC-LED reactor system, 5 log fecal coliform reductions were achieved at an exposure time of 3.5 min and a wavelength of 275 nm. Andreadakis et al. [30] attempted to inactivate the residual fecal coliform present in the effluent due to TSS concentration at higher UV doses, but no inactivation was observed at higher UV dose. They have reported that the inactivation rate of the filtered sample is 2.5 times greater than the unfiltered sample, indicating that suspended solids protect micro-organisms. The concentration of fecal coliform in the effluent is within the permissible limit recommended by Indian standards for treated sewage reuse (230 CFU/100 mL).

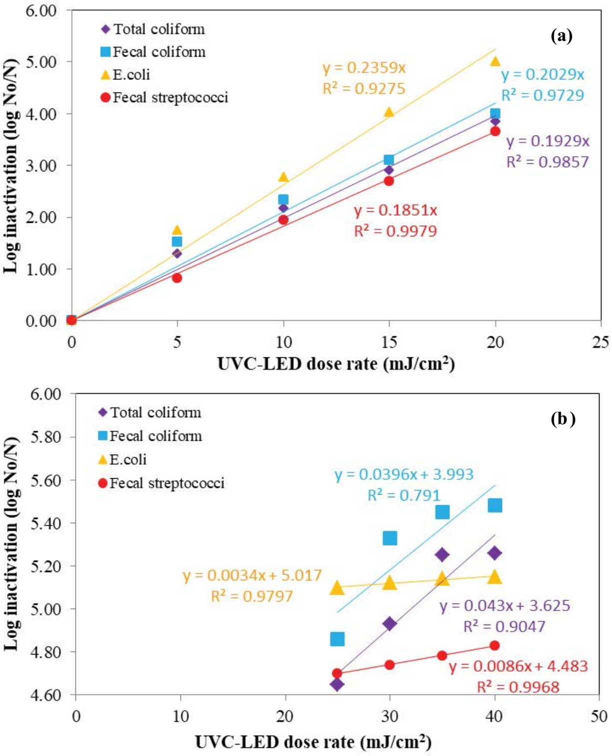

Fig. 3. Inactivation responses at different UVC-LED doses (a) linear Chick–Watson model and (b) non-linear shoulder model.

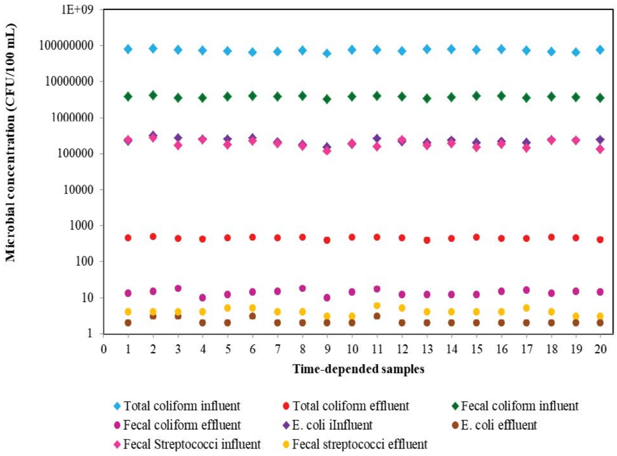

Fig. 4. Microbial concentrations in influent and effluent.

Table 3
Kinetic analysis of Chick–Watson linear model and shoulder model

Microbial parameters	Chick-Watson linear model		Shoulder model		
	k (cm²/mJ)	R^2	k (cm²/mJ)	b	R^2
Total coliform	0.1929	0.9857	0.0430	–0.2154	0.9047
Fecal coliform	0.2029	0.9729	0.0396	–0.2386	0.7910
E. coli	0.2359	0.9275	0.0034	–0.4423	0.9797
Fecal streptococci	0.1851	0.9979	0.0086	–0.3440	0.9968

3.4. Disinfection efficiency of E. coli and fecal streptococci

The most common fecal contamination indicators are *E. coli* and fecal streptococci. However, recent studies show much interest in developing an effective and efficient UVC-LED reactor system to evaluate its inactivation ability against other pathogenic organisms. In this study, in addition to fecal coliforms, the performance of UVC-LED reactor on *E. coli* and fecal streptococci inactivation was studied. The variation in *E. coli* and fecal streptococci influent and effluent concentrations recorded in AlGaN-based UVC-LED reactor at a wavelength of 275 nm is presented in Fig. 4. The average concentration of *E. coli* and fecal streptococci in the effluent was 2 and 4 CFU/100 mL, respectively. With an exposure time of 2 min and an optimal UVC-LED dose of 20 mJ/cm^2, the average log reduction of *E. coli* was calculated to be 5.00 log. The average log reduction of fecal streptococci was calculated to be 4.70 log at an exposure time of 2.5 min and an optimal UVC-LED dose of 25 mJ/cm^2. As UVC-LEDs are capable of damaging the DNA directly by producing CPD, it takes only a few minutes to inactivate *E. coli* and fecal streptococci [18]. There is no permissible limit available for *E. coli* and fecal streptococci discharge or reuse in Indian standard. The graphs show that the concentration of *E. coli* and fecal streptococci in the effluent is very low. Chrtek and Popp [31] evaluated the performance of UVC reactor using secondary treated wastewater and reported that the log inactivation of total coliform, fecal coliform, and fecal streptococci was 4.2, 4.3, and 3.8 log, respectively, under UVC irradiation. Therefore, the AlGaN-based UVC-LED reactor system has a strong inactivation ability for total coliform, fecal coliform, *E. coli*, and fecal streptococci. Nyangaresi et al. [32] investigated the inactivation efficiency and photo-reactivation of *E. coli* using the UVC-LED (275 nm) system under batch mode. It was reported that 275 nm UV-LED exhibited good persistence against photo-reactivation as the damage of protein occurs at 275 nm. Therefore, the significance of employing 275 nm UVC-LED is to deliver high output power, attain maximum microbial inactivation efficiency, and prevent photo-reactivation. The inactivation rate of *E. coli* at higher UV dose and shorter irradiation time is found to be more effective than lower UV dose with longer irradiation time [12,14].

3.5. Electrical energy efficiency

The selection of UVC-LEDs at various wavelengths based on inactivation efficiency and kinetics can cause ambiguity in results. To make the economically convincing decision, it is necessary to evaluate the electrical energy efficiency of UVC-LEDs used for inactivating the pathogens [32,33]. In this research study, the peak wavelength of UVC-LED used was 275 nm, and the maximum UVC-LED dose rate required to inactivate total coliform, fecal coliform, *E. coli*, and fecal streptococci was 35, 35, 20, and 25 mJ/cm^2, respectively. The amount of electrical energy consumed for inactivating total coliform, fecal coliform, *E. coli*, and fecal streptococci was determined by using Eq. (3). The electrical energy consumed per 5.25, 5.45, 5.00, and 4.70 log inactivation for total coliform, fecal coliform, *E. coli*, and fecal streptococci respectively was calculated to be 0.204, 0.204, 0.117, and 0.146 kWh/m^3, respectively. It can be observed that the consumption of electrical energy was lower for *E. coli* inactivation than that of other bacteriological parameters. This is due that inactivating *E. coli* required only 20 mJ/cm^2 of UVC-LED dose rate. The electrical energy ($E_{E,5}$) required to inactivate *E. coli* at 275 nm was much lower than (0.117 kWh/m^3) the results reported in a study by Nyangaresi et al. [32], where the 275 nm UV-LED required 0.1367 and 0.2219 kWh/m^3 for $E_{E,1}$ and $E_{E,2}$, respectively in inactivation of *E. coli*.

4. Conclusion

In this study, the microbial inactivation ability of AlGaN-based UVC-LED reactor was evaluated in secondary treated sewage disinfection for bacteriological parameters namely total coliform, fecal coliform, *E. coli*, and fecal streptococci at a wavelength of 275 nm. The new reactor design incorporating a baffle arrangement of UVC-LEDs has achieved maximum germicidal inactivation efficiency at minimum electrical energy consumption. The log inactivation by AlGaN-based UVC-LED fitted good with the first-order Chick-Watson and Shoulder log-linear disinfection models. The electrical energy consumed for 5 log reduction of *E. coli* at 275 nm was lower than that of other bacteriological parameters reported, which was due to its lower UVC-LED dose rate. This new and compact reactor system in combination with biological sewage treatment methods is proposed to be an efficient green technology for wastewater treatment facilities.

Acknowledgments

This research work was financially supported by M/s L&T Construction, Chennai, India [Grant No. E8722WOD7000116]. Contributions of Mr. S. Jagannathan, CTO, WET IC, L&T Construction, Mr. P. Ganesh Kumar, Sr. Engineering Manager, WET IC, L&T Construction, and Mr. Mohamed Ahmed Salah, Water Technologist, WET IC, L&T Construction are acknowledged.

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