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Enhanced inactivation of *E. coli* by pulsed UV-LED irradiation during water disinfection

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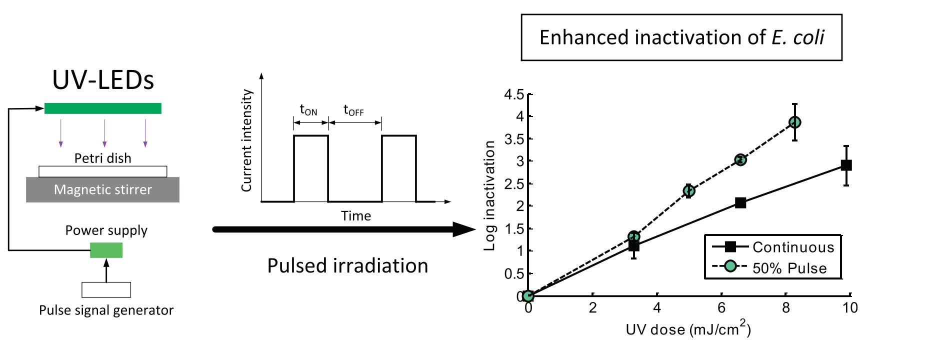
HIGHLIGHTS

- *E. coli* inactivation was enhanced using pulsed UVC-LED irradiation.
- Log-inactivation increased substantially as duty cycle decreased from 100% to 5%.
- Inactivation enhancement of pulsed UV were similar for 280 and 265 nm LEDs.
- High current pulsed irradiation showed remarkable inactivation enhancement.

GRAPHICAL ABSTRACT



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ABSTRACT

Pulsed ultraviolet (UV) irradiation has presented enhanced inactivation efficiency in water disinfection and food decontamination. As an emerging UV source, UV light-emitting diodes (UV-LEDs) are an attractive alternative for pulsed irradiation because they can be turned on and off with a high and adjustable frequency. In this study, disinfection efficiencies of pulsed and continuous UV-LED irradiation were compared for *Escherichia coli* (*E. coli*) inactivation in water using a high power 285 nm LED and low power 265 and 280 nm LEDs. Factors including various duty cycles, pulse frequencies and UV irradiances were evaluated. The log-inactivation of *E. coli* increased substantially as the duty cycle decreased from 100% to 5% at the same UV dose. For 265 and 280 nm LEDs, the log-inactivation enhancements of pulsed UV irradiation were similar. When a higher irradiance was applied, the energy efficiency enhancement of pulsed UV irradiation became more obvious. The log-inactivation of *E. coli* enhanced remarkably using high current pulsed irradiation of 280 nm LEDs. Compared to continuous UV irradiation, pulsed UV-LED irradiation is an attractive alternative for *E. coli* inactivation in water considering energy efficiency.

1. Introduction

Disinfection is an important process for diminishing waterborne diseases to ensure the safety of drinking water. Ultraviolet (UV) irradiation

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is an effective approach to water disinfection, which presents high inactivation ability against a wide range of pathogenic microorganisms and has been generally applied worldwide (Hijnen et al., 2006; Song et al., 2016). However, there are some critical problems regarding the UV sources of traditional low pressure and medium pressure mercury lamps. These UV lamps contain toxic mercury, which may cause environmental problems if not disposed properly (Chevremont et al., 2013; Close et al., 2006). In addition, the wall plug efficiency of mercury lamps is limited (<35% typically) and the lifetime is relatively short (around 10,000 h) (Autin et al., 2013; Chatterley and Linden, 2010).

As a novel UV source, UV light-emitting diodes (UV-LEDs) present a chance to solve the problems mentioned above. The advantages of UV-LEDs include no mercury containing, more durable, no warm-up time, and potential for less energy consumption and longer lifetime (Würtele et al., 2011). It is estimated that the lifetime of UV-LEDs may reach 100,000 h in the near future (Song et al., 2016). Besides, UV-LEDs with wavelengths in the range from 210 to 365 nm can be manufactured by combining semiconducting materials in proper proportions (Taniyasu and Kasu, 2010), which is beneficial because the UV wavelength has a critical effect on water disinfection efficiency (Song et al., 2016; Vilhunen et al., 2009).

Another superiority of UV-LEDs is that they can be turned on and off with a high and adjustable frequency, making them an attractive alternative for pulsed irradiation (Song et al., 2016; Würtele et al., 2011). The conventional way to generate pulsed UV is using a xenon lamp, which has shown application potential in water disinfection and food decontamination. Bohrerova et al. (2008) reported that the inactivation of *Escherichia coli* (*E. coli*) and phage T4 and T7 in water were significantly faster applying pulsed UV irradiation compared with low pressure or medium pressure UV lamps at the same UV dose. Elmnasser et al. (2007) summarized that pulsed light can inactivate microorganisms in food rapidly as an athermal technology. However, due to concerns about overheating, xenon lamps are limited in adjusting the duty cycle and pulse frequency, which have essential effects on inactivation efficiency of pulsed irradiation (Wengraitis et al., 2013). By contrast, UV-LEDs can generate pulsed irradiation with a larger range of duty cycles and pulse frequencies, offering more possibilities for inactivation efficiency optimization. Although there are numerous studies on pulsed irradiation using xenon lamps, especially for food decontamination (Elmnasser et al., 2007), these results cannot be applied directly for pulsed UV-LED irradiation because the pulse patterns are quite different between the two UV sources (Song et al., 2016). Therefore, more research regarding pulsed UV-LED irradiation is necessary.

There are only few studies on pulsed irradiation disinfection using UV-LEDs, especially rare for water disinfection or using UVC-LEDs (UVC: <280 nm), which are the most effective in disinfection. Li et al. (2010) presented enhanced inactivation efficiency of pulsed UVA-LED (UVA: 315–400 nm) irradiation on *Candida albicans* and *E. coli* biofilms. Xiong and Hu (2013) established a UVA-LED/TiO_2 system and found enhanced inactivation on *E. coli* by pulsed irradiation. However, both studies applied UVA-LEDs with peak wavelength at 365 nm. UVC irradiation can induce the formation of cyclobutane pyrimidine dimers (CPD) in the DNA of microorganisms and cause direct damage, while UVA irradiation can barely cause CPD formation and is much less effective for disinfection than UVC (Song et al., 2016; Xiao et al., 2018). As a result, their disinfection processes lasted for >1 h even with relatively high irradiance, while UVC-LEDs only take a few minutes to inactivate *E. coli* with relatively low irradiance typically. One of the major disadvantages of the pulsed UV disinfection is the lower time efficiency compared with the continuous UV irradiation, and the pulsed UVA-LED disinfection is even less time efficient. Therefore, UVC-LEDs are more appropriate for pulsed UV disinfection than UVA-LEDs. Wengraitis et al. (2013) studied pulsed UVC-LED disinfection of *E. coli* on agar plates with 272 nm LEDs and reported higher inactivation efficiency than continuous irradiation. However, inactivation efficiency in water was not investigated in their study, and no information about UV dose response of the disinfection process was provided.

Therefore, water disinfection efficiency of pulsed UV-LED irradiation was studied in this research. A high power 285 nm LED and low power 265 and 280 nm LEDs were applied for inactivation of *E. coli*, a commonly used indicator organism for assessment of water disinfection performance (Li et al., 2017a, b). Effects of duty cycle, pulse frequency, UV wavelength, UV irradiance and high current pulse on disinfection efficiency were investigated. Energy efficiency and time efficiency under various duty cycles were analyzed as well.

2. Materials and methods

2.1. Pulsed UV-LED system

A high power (30 mW output) 285 nm LED was supplied by Nikkiso (Japan). Low power (approximately 1 mW output) 265 and 280 nm LEDs were supplied by Qingdao Jason Electric Co., Ltd. (Qingdao, China). For normal current experiments, the high power 285 nm LED module was equipped with 1 LED, while the low power 265 and 280 nm LED modules were equipped with 9 LEDs (3*3). For high current pulse experiments, a low power 280 nm LED module equipped with 4 LEDs (2*2) was used. The spacing of the UV-LEDs is approximately 1 cm.

The pulsed UV-LED system was composed of a quasi-collimated UV-LED module, a constant current power supply and a pulse signal generator (Fig. 1(a)(b)). Pulse frequency (Hz) and duty cycle (%) are essential parameters in a pulsed UV-LED system. Pulse frequency is the number of pulse periods per unit of time, and duty cycle is the turned on time proportion of LEDs during a pulse period, as defined in Eqs. (1) and (2).

$$\text{Pulse frequency} = \frac{1}{t_{ON} + t_{OFF}} \tag{1}$$

$$\text{Duty cycle} = \frac{t_{ON}}{t_{ON} + t_{OFF}} \tag{2}$$

where t_{ON} and t_{OFF} are the ON and OFF times of LEDs during a pulse period (Fig. 1(c)).

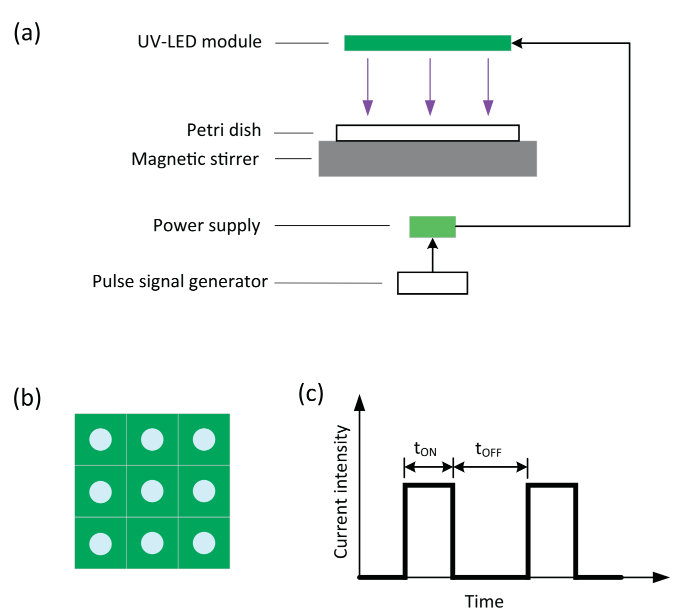

Fig. 1. Illustrations of (a) the pulsed UV-LED system (side view), (b) a UV-LED module (top view, 9 LEDs, 3*3), and (c) the pulse signal.