Water Research 147 (2018) 331–341

Contents lists available at ScienceDirect

Water Research

journal homepage: www.elsevier.com/locate/watres

Effects of single and combined UV-LEDs on inactivation and subsequent reactivation of *E. coli* in water disinfection

Paul Onkundi Nyangaresi [a], Yi Qin [a], Guolong Chen [b], Baoping Zhang [a, *], Yinghua Lu [c], Liang Shen [c, **]

[a] Department of Electronic Engineering, Laboratory of Micro/Nano-Optoelectronics, Xiamen University, Xiamen, Fujian, 361005, China
[b] Department of Electronic Science, Fujian Engineering Research Center for Solid-State Lighting, Xiamen University, Xiamen, 361005, China
[c] Department of Chemical and Biochemical Engineering, College of Chemistry and Chemical Engineering, Xiamen University, Xiamen, Fujian, 361005, China

ARTICLE INFO

Article history:
Received 6 August 2018
Received in revised form
2 October 2018
Accepted 5 October 2018
Available online 8 October 2018

Keywords:
UV-LED
Disinfection
Inactivation
Reactivation
Synergistic effect
Electrical energy efficiency

ABSTRACT

Ultraviolet light emitting diodes (UV-LEDs) have shown a potential to replace traditional Ultraviolet (UV) pressure lamps for water disinfection. However, the research is not sufficient and hence, it is still difficult to make any logical conclusions. In this work, UV-LEDs with peak emissions at 267, 275, 310 nm and combined emissions at 267/275, 267/310 and 275/310 nm were applied to a batch water disinfection system. Under either single- or combined-wavelength situation, the inactivation efficiency, reactivation (due to photoreactivation and dark repair) after UV irradiation and electrical energy consumption were evaluated by way of the model bacterium *Escherichia coli*. It was found that, the 267 nm UV-LED had the highest inactivation efficiency than other UV-LEDs. Although reactivation occurred after 267, 275, 267/275 and 275/310 nm UV-LEDs' irradiations, it occurred to a lesser extent in dark repair than in photoreactivation, demonstrating that photo-effect is the dominant mechanism of reactivation. In addition, decay phase was more prominent than reactivation in dark repair. However, the irradiation by the 275 nm UV-LED showed a better persistence against reactivation which could be attributed to protein damage at 275 nm. No synergistic effect for combined wavelengths was observed in this study. The electrical energy consumption was lower for the 275 nm UV-LED than the other UV-LEDs which was attributed to its higher wall plug efficiency. This study showed the variation principle between the single and combined UVB/UVC-LEDs in inactivation efficiency, inhibition of reactivation, synergistic effect and electrical energy consumption in treatment of *E. coli*, which is useful for the reasonable exploitation of UV-LEDs in water disinfection systems.

1. Introduction

UV irradiation with wavelengths in the germicidal range (200–320 nm), is the latest method of modern water and wastewater treatment (Kowalski, 2009). Currently low pressure (LP) and medium pressure (MP) mercury lamp emitting monochromatic emission at a wavelength of 253.7 nm and polychromatic emission light at a broad range of wavelengths, 200–600 nm respectively, are widely employed as a UV source in drinking water and wastewater treatment plants (Bolton and Cotton, 2011). The efficacy and the doses needed using these mercury lamps are well established for various pathogens which include bacteria, protozoan parasites, and viruses. (Abbaszadegan et al., 1997).

The newly emerging ultraviolet light-emitting diode (UV-LED) is a potential alternative of traditional UV mercury lamps because of the advantages such as: diversity in wavelengths, environmental friendliness (no mercury), compactness, robustness, faster start-up time (excluding warm-up), potentially less energy consumption, longer lifetime, and the ability to turn on and off with high frequency (Würtele et al., 2011). In addition, UV-LED reactors can best be utilized in small scale, which is especially convenient in remote areas in view of cost (Crawford et al., 2005; Lui et al., 2014). Such UV-LEDs are wide band gap semiconductors composed mostly of gallium nitride (GaN) and aluminum gallium nitride (AlGaN). Although the wall plug efficiency (WPE) of UV mercury lamps (15–35%) is higher than that of UV-LEDs (1–3%), the latter is

* Corresponding author.
** Corresponding author.
E-mail addresses: bzhang@xmu.edu.cn (B. Zhang), shenliang@xmu.edu.cn (L. Shen).

https://doi.org/10.1016/j.watres.2018.10.014

expected to be improved significantly, being similar to the case seen in visible LEDs (Harris et al., 2013).

For UV-LEDs to be applied as a practical option, several studies have examined their application in inactivation of pathogens for water disinfection. Several of these studies have reported that UV-LEDs at wavelength around 265 nm have a relatively higher inactivation of microorganisms than other wavelengths in the UVB and UVC (200–300 nm) range (Chatterley and Linden, 2010; Bowker et al., 2011; Chevremont et al., 2012a; Oguma et al., 2013; Beck et al., 2017; Rattanakul and Oguma, 2018; Li et al., 2017). The inactivation by the UVB and UVC radiation is effected through the formation of lesions in the genomic DNA of the organisms. The major DNA lesions induced by the UV light are cyclobutane pyrimidine dimers (CPDs), pyrimidine 6-4 pyrimidone photoproducts (6-4 PPs), and their Dewar isomers (Ravanat et al., 2001; Sinha and Häder, 2002; Cadet et al., 2005; Friedberg et al., 2006). The presence of these UV-induced lesions would inhibit the normal replication of DNA resulting in inactivation of the microorganisms.

However, some microorganisms, particularly bacteria, are capable of reactivating by repairing their damaged DNA after UV irradiation by mechanisms such as photoreactivation and excision repair (dark repair) (Friedberg et al., 1995; Harm, 1980). This will greatly decrease the final inactivation result and thereafter health risks of infection, when UV radiation is used for microbial disinfection in water. Photoreactivation is a process where microorganisms utilize light in the wavelength range of 330–480 nm to activate a photolyase enzyme, which binds specifically to the CPDs (CPD photolyase) or 6-4 PPs (6-4 photolyase) and directly monomerizes the cyclobutane ring of the pyr <> pyr and protects the genome from deleterious effects of UV radiation whereas excision repair (dark repair) is a multistep, where an abnormal or damaged base is removed by two major subpathways: (i) base excision repair (BER) and (ii) nucleotide excision repair (NER) (Rastogi et al., 2010).

Previous studies have indicated that, medium pressure (MP) UV lamps have the ability to repress photoreactivation of *Escherichia coli (E. coli)* (Oguma et al., 2004). It has been suggested that, the repressed photoreactivation is due to the irreversible oxidative damage to photolyase by MP UV (Quek and Hu, 2008). Another study also assumed that the MP UV irradiation resulted in less photoreactivation due to induced damage to proteins other than DNA itself (Kalisvaart, 2004). In addition, MP UV emission wavelengths of 220–300 nm was reported to reduce the subsequent photorepair of *E. coli* by causing a disorder with endogenous photolyase a DNA repair enzyme (Oguma et al., 2002). The DNA of most of the microorganisms is believed to have an absorption maximum between 260 and 270 nm (LeChevallier and Kwok-Keung, 2004; Gates, 1930). Meanwhile the proteins that are responsible for infection usually show absorption maximum between 275 and 280 nm which is caused by the absorbance of aromatic amino acids tryptophan, and tyrosine and cystine (i.e. of disulfide bonds) (Schmid, 2001). Therefore, repression of photoreactivation can be attributed to protein damage at a wavelength between 275 and 280 nm, though more studies should be conducted to confirm this hypothesis.

Since effectiveness of UV light for inactivating microorganisms is in the UV-B and UV-C ranges of the spectrum (200–310 nm), similar to MP UV lamp, repression of the reactivation by UV-LEDs is possible, which has also been reported in the recent study. Specifically, the 280 nm UV-LED was found able to significantly repress reactivation (Li et al., 2017). However, the UV-LEDs are characterized by diversity in wavelengths, whereas MP UV lamp emits a continuous and broad spectrum of germicidal wavelengths. Therefore, the combination of UV-LEDs of different wavelengths may be necessary to suppress effectively the reactivation. This raises the study on the inactivation effect using the combined UV-LEDs. The synergistic effect is normally determined by comparing the results of log inactivation by combined disinfection treatments and the results from the sum of log inactivation by individual treatments (Koivunen and Heinonen-Tanski, 2005). Till now, a synergistic effect after application of UV-LEDs disinfection systems has only been reported in quite few references (Chevremont et al., 2012a; Nakahashi et al., 2014; Green et al., 2018). Some studies even reported contrasting finding of no synergistic effect (Oguma et al., 2013; Beck et al., 2017; Li et al., 2017). Nevertheless, the effectiveness of both inactivation and reactivation in cases of the combined UV-LEDs is of great importance for water disinfection.

Electrical energy efficiency is another factor involved in making an economically reasonable decision when designing disinfection systems. It is characterized by a parameter known as electrical energy per order (E_{EO}) which has been previously used for interpreting collimated beam data to estimate electrical efficiencies of LP UV and MP UV lamps for large-scale treatment of chemical contaminants (Sharpless and Linden, 2005). Therefore, the same parameter of electrical energy per order (E_{EO}) can be applied in UV-LED disinfection systems to determine their electrical energy efficiency. However, up to date only a few studies have considered both inactivation and electrical energy efficiency. In one study, the inactivation and electrical energy efficiency of 260, 280 and 260/280 nm UV-LEDs were compared (Beck et al., 2017). For *E. coli* inactivation in particular, the results of 260 nm and 280 nm UV-LEDs were not statistically different, which contradicts the earlier findings that the relative peak of bactericidal effectiveness is between 260 and 270 nm (Gates, 1930). However, it was reported that, the electrical energy efficiency of the 280 nm UV-LED and the 260/280 nm UV-LED combination was less than the 260 nm UV-LED. In another study, the 265, 280 and 300 nm UV-LEDs were used in inactivation of *E. coli*, *Bacillus subtilis spores*, *Bacteriophage Qβ*, *Pseudomonas aeruginosa* and *Legionella pneumophila* (Rattanakul and Oguma, 2018). It was reported that, although the 265 nm UV-LED had higher inactivation efficiency, the 280 nm UV-LED showed the lowest energy consumption. The differences in the electrical energy consumption were attributed to difference in wall plug efficiency. Note that, the wall-plug efficiency (radiant efficiency) is the energy conversion efficiency with which the system converts electrical power into optical power. In other words, the wall-plug efficiency is defined as the ratio of the radiant flux (total optical output power of the device) to the input electrical power (Barnes, 2007). However, we think that, not only does the electrical energy consumption depend on wall plug efficiency, but also on the water factor and the sensitivity of the microorganism at a particular wavelength.

As shown above, the studies on application of UV-LEDs for water disinfection are insufficient and from the few published ones, discrepancies exist on important factors such as inactivation efficiency, repression of reactivation of microorganisms after UV irradiation, combined effect of different wavelengths and electrical energy efficiency. Therefore, in this study, these factors are evaluated, with special attention on the potential synergistic effect of combined UV-LED wavelengths. We believe that, the results from this work will provide additional and beneficial information for consideration in application of UV-LEDs for water disinfection systems.

2. Materials and methods

2.1. Culturing and enumeration of microorganisms

In this study, *E. coli* was chosen as a model microbe because the presence of *E. coli* itself is a good indication on the presence of pathogens and they are a good measure of overall water quality