EXPERIMENTAL AND NUMERICAL EVALUATION OF A UV-LED POINT OF USE DEVICE

Richard Matthew Jenny* rmjenny@ncsu.edu  
Dr. Otto D. Simmons III† odsimon@ncsu.edu  
Dr. Joel J. Ducoste* jducoste@ncsu.edu  

*Department of Civil, Construction, and Environmental Engineering, North Carolina State University, Campus Box 7908, Raleigh, NC 27695-7908, USA  
†Department of Biological and Agricultural Engineering, North Carolina State University, Campus Box 7625, Raleigh, NC 27695, USA

ABSTRACT

The use of ultraviolet (UV) light for water treatment disinfection has become increasingly popular due to its ability to inactivate chlorine-resistant microorganisms without the production of known disinfection by-products. Currently, mercury-based lamps are the most commonly used UV disinfection source; however, these lamps are toxic if broken during installation or by foreign object strike during normal operation. UV light emitting diodes (LEDs) offer an alternative, non-toxic UV source that will provide design flexibility due to their small size, longer operating life, and fewer auxiliary electronics than traditional mercury-based lamps. Modeling of UV reactor performance has been a significant approach to the engineering of UV reactors in drinking water treatment. Yet, no research has been performed on the experimental and modeling of a continuous flow UV-LED reactor. A research study was performed to validate a numerical computational fluid dynamics (CFD) model of a continuous flow UV-LED water disinfection process. Results showed good agreement between numerical simulations and experimental testing. Accuracy of fluid velocity profile increased as flow rate increased from 109 mL/min to 190 mL/min, whereas chemical actinometry saw better agreement at the low flow rate. Biodosimetry testing was compared only at the low flow rate and saw good agreement for log inactivation of bacteriophages Qβ and MS-2 at 92% and 80% UV transmittance (UVT).

KEYWORDS

Ultraviolet light disinfection; Light Emitting Diode; Computational fluid dynamics; Continuous flow

1. INTRODUCTION

The application of UV light for drinking water disinfection has generated increasing amounts of interest due to many of its distinct advantages over traditional, chlorine-based disinfection processes. These advantages include its ability to inactivate the chlorine-resistant protozoa Cryptosporidium and Giardia, no known disinfection by-product (DBP) formation, no resulting taste/odor issues, and over-dosing will not compromise public health (Chatterley et al., 2010).

Presently, the majority of UV light technologies used for water treatment are generated by mercury-based lamps. The three most common mercury-based UV sources are low-pressure (LP), low-pressure high output (LPHO), and medium-pressure (MP) lamps. LP and LPHO lamps emit monochromatically at 253.7nm and generate low to medium power outputs. Conversely, MP lamps emit polychromatic light (200-400nm) and operate at much higher temperature and intensity than LP and LPHO lamps. Although MP lamps emit higher output energy than LP and LPHO lamps, the broadened wavelengths emitting from the MP lamp reduces its germicidal efficiency (USEPA, 2006).

Recent technological advances has allowed for the use of small Light Emitting Diodes (LEDs) between 5-9 mm in diameter that generate light in the germicidal range (Chatterley et al., 2010; Bowker et al., 2010). Currently, UV-LEDs operate in the range of 247-365 nm (Gaska, 2007). LEDs can be manufactured to produce a desired wavelength of light, which can offer a significant increase in design flexibility over traditional lamp technologies (Shur and Gaska, 2008). The additional design flexibility offered by UV-LEDs provides an opportunity to engineer a more efficient disinfection process. Further drawbacks to
traditional UV lamps include its toxic components. Mercury is hazardous to both human health and the environment, and if the lamps are broken during installation, maintenance, disposal, or by foreign-object strike, mercury vapor may enter the drinking water supply or may expose plant personnel (USEPA, 2006). LEDs provide significant benefits, as they have been recognized as a system that saves energy (input power is in range of mW), lowers maintenance cost, lengthens replacement intervals, and are non-toxic (Chatterley et al., 2010). For all these reasons, LEDs have the potential to replace fluorescent lamps for point-of-use drinking water disinfection systems.

Although an increasing amount of experimental research has been performed using UV-LEDs, no research has combined the results from a computational fluid dynamics (CFD) model and completely validated the data with experimental testing. This is in part due to the complex nature of the multiple physics that occur inside a UV reactor. Nonetheless, CFD is considered as a powerful and well-established tool for UV reactor modeling and design (Liu et al., 2007; Wols et al., 2010). Several authors used CFD calculations to validate reactor hydraulics, and in most of the cases a relatively good agreement was established (Ducoste et al. 2005; Sozzi and Taghipour, 2006; Zhao et al., 2009; Wols et al., 2010). Knowledge of the reactor hydraulics, irradiance, and microbial response of the reactor is necessary for validation. The hydraulic validation provides comprehensive information about the flow field, pressure distribution, and mixing behavior of the fluid inside a reactor (Wols et al., 2010). Chemical actinometry is a strong indicator of the average irradiance inside the reactor at various operating conditions (flow rate, water transmittance, and lamp power output). For a given set of reactor operating conditions, the hydraulics and average irradiance can provide information about the average dose within the reactor. The final validation component involves the prediction of the microbial log inactivation. These validation approaches, although performed separately, are combined to provide important understanding of the model performance to predict the UV reactor behavior.

2. METHODOLOGY

2.1. Hydraulic Characterization

In this study, a positive step tracer test was conducted where a NaCl (127μS/m) (Certified ACS Crystalline S271-1, Fischer Scientific Pittsburg PA) solution was used as a non-reactive tracer. NaCl solution was injected as a continuous input at the inlet and samples were collected at the outlet at an interval of 5% of theoretical residence time. Conductivity of all the samples was measured with a digital conductivity meter (PH/CON 2700 MTR W/PROBE, Cole-Parmer, Court Vernon Hills, Illinios). Tracer simulations were performed using COMSOL Multiphysics (version 4.3a, COMSOL, Inc., Burlington, MA). The transport of the tracer chemical was performed using the continuity equation, Navier Stokes equations, and the scalar convective-diffusion transport equations (Liu et al., 2007). The back ground fluid transport was performed at steady state while the tracer transport simulation was performed under transient conditions to replicate the experimental conditions.

2.2. Fluoence rate characterization

The fluence rate characterization was performed using chemical actinometry. Standard actinometry solution was created using potassium iodide, potassium iodate, and borax. The concentration of iodide, iodate, and borax used was 0.6M, 0.1M and 0.01M, respectively. When the iodate and iodide are exposed to UV light, the chemical species react to form triiodide. The triiodide concentration was measured using spectroscopy where the excitation wavelength is 352 nm. The absorbance value was converted to a molar concentration of triiodide using the following equation (1):

\[
\text{Triiodide [M]} = \left( \frac{\text{Final Absorbance} - \text{Initial Absorbance}}{26400} \right)
\]

(1)

The reactor was operated with the UV LEDs on for approximately 11.8 minutes and 20 minutes for the 190 mL/min and 109 mL/min flow rates, respectively. These time periods were used to yield the maximum triiodide concentration at the outlet and to ensure that the flow had achieved steady state conditions. A 2.25L actinometry batch solution was used as the stock feed to achieve steady state and make replicate measurements in the reactor effluent. Continuous flow-through experimental tests were performed at two different UV LED light configurations, 25 and 30 active LEDs (260nm).
A model of the iodide/iodate/triiodide chemical reaction was used to predict the effluent concentration of triiodide using CFD. The chemical equations involved in the transformation of iodide and iodate to triiodide in the presence of UV light, as described by Rahn (1997), was incorporated into the CFD code. Bench scale actinometry tests were performed to determine the empirical reaction rate constant used in the CFD model at different UV doses.

2.3. Microbial inactivation characterization
Microbial inactivation kinetic data for the reactor was generated using bacteriophages MS-2 and Qβ, both of which are model indicator organisms representing single-stranded human RNA viruses, such as Hepatitis A virus and human Noroviruses. Each virus stock used for collimated beam and continuous flow experiments were propagated and spiked at $10^8$ pfu (plaque forming unit)/mL such that a 5 log$_{10}$ inactivation could easily be followed for each virus. Virus inactivation trials for each bacteriophage were conducted independently; therefore, individual viral stocks (MS-2 and Qβ) were not mixed and each was evaluated without the presence of the other. A standard collimated beam protocol, as described in USEPA (2006), was followed; bacteriophage samples (10mL) were placed under the LED collimated beam and exposed to UV light to achieve various UV dosages. Control samples were collected prior to exposure to UV LEDs. A 2250 mL solution of saline-calcium spiked with the appropriate bacteriophage (MS-2 or Qβ; $10^8$ pfu/mL) was used for the flow-through reactor experimental tests. The solution volume provided time for steady-state conditions to be achieved in the reactor, allowed for 3 times the HRT (hydraulic retention time) to pass through the reactor prior to sample collection, and was consistent with the testing volume used in the actinometry tests. Operating conditions included 109 mL/min flow rate at two different UV transmittance (UVT) values; 92% and 80%. Each test included 30 active LEDs (260nm) at a total current and voltage of 0.6A and 9.7V, respectively (as suggested by the manufacturer). The sample was pumped through the reactor using a peristaltic pump (Ocaton PC 2700 Master flex L/S Drives, Cole-Parmer, Court Vernon Hills, Illinois) for 15 minutes prior to collecting the first of three samples. The remaining two samples were collected in 2.0 minute increments following the first sample collection. A control sample for each test was collected and stored in a sterile dark tube prior to exposure to the UV-LEDs. Enumeration of MS-2 and Qβ followed the assay method used by Bohrerova et al., 2006.

Biodosimetry validation was completed using the response kinetics of bacteriophages MS-2 and Qβ. Numerical modeling of MS-2 and Qβ was solved using a steady state convective diffusion equation. MS-2 and Qβ both show log-linear dose response curves (as found from collimated beam testing); therefore, the kinetics most accurately follows the Chick-Watson equation (Arafen et al., 2011).

2.4. Reactor construction
The reactor design and construction was based on the initial modeling effort done by Arafen et al., (2011). The reader is guided to the literature for details on the construction of the unit. A picture of the shell of the reactor has been provided in Figure 1. Figure 2 displays a graphical representation of the ideal fluid trajectory, as well as the location of the mid-baffle wall and mid-baffle lip.
2.5. Numerical simulations

A finite element based CFD modeling software, COMSOL Multiphysics (COMSOL Inc., Burlington MA), was used to solve conservation of mass and momentum equations to predict the flow field and pressure distribution inside the specified domain. This software was used for geometry generation, meshing, physics settings, solving, and post processing. An iterative matrix solver (GMRES) was used to solve the governing equations of fluid dynamics, and convergence was considered to be achieved when relative errors of the numerical solutions were below 1E-06.

The entire flow domain was discretized by 436,123 tetrahedral elements to calculate flow rates of 109 mL/min and 190 mL/min. A 22.56 cm long pipe was modeled at the inlet to obtain a fully developed flow profile prior to entrance into the main reactor body. The same length of pipe was also provided at outlet, according to the suggestion of Durst et al., (2005). A constant velocity (uniformly distributed over the inlet area), was maintained at the inlet and zero pressure boundary condition was imposed at the outlet. Other closed boundaries were modeled as no slip wall. Positive step and negative step tracer tests were simulated by using the unsteady convective diffusion equation. Light model equation was used to calculate the fluence rate distribution inside the computational domain (Liu et al., 2004; Bowker et al., 2010). All of the LEDs were assumed to have flat window (FW) configurations, as established by the manufacturer (SET, Inc., 2008). Viewing angle $\alpha = 60^\circ$ was imposed in this study based on the manufacturer specifications.
3. RESULTS AND DISCUSSION

3.1. Hydraulic results
A more in-depth hydraulic analysis was completed in a previous study (Arafin et al., 2011), to which the reader is directed for further insight. In summary, using a continuous input of NaCl tracer, the residence time density function was generated at both the 109 mL/min and 190 mL/min flow rates. The amount of dispersion present during both operating conditions was calculated for each flow rate. Under plug flow conditions the dispersion number should approach zero, while under low dispersion and high dispersion conditions these values will gravitate toward 0.002 and 0.2, respectively (Levenspiel, 1972). Analysis showed little dispersion (dispersion numbers were 0.0062 and 0.014 for the 109 mL/min and 190 mL/min flow rates, respectively) and thus near plug flow conditions at both operating conditions. Mixing increased as flow rate increased.

For hydraulic validation, the cumulative residence time density (RTD) functions for the numerical prediction and 95% confidence intervals from experimental testing were compared at both flow rates. Relatively good agreement was established at the higher flow rate compared to the low flow rate. The model more accurately predicts the dispersion that occurs during the 190 mL/min flow condition, predicting a dispersion number of 0.024 (experimental analysis found a dispersion number of 0.014). However, under the low flow condition the model does not as accurately capture the dispersion, as the dispersion number is predicted to be 0.025 (experimental analysis found a dispersion number of 0.0062). This represents a situation where the model is over-predicting the macro-scale dispersion within the reactor at the low flow condition. Using the Lagrangian particle tracking method in COMSOL, it was found that the model predicts a significant amount of fluid mixing near the mid-baffle lip during the 109 mL/min flow condition. This is likely the region where the model over-predicts dispersion at the 109 mL/min operating condition.

3.2. Fluence rate results: Chemical actinometry
The chemical kinetics of the actinometry solution was determined using collimated beam experiments as discussed earlier in the methods section (Section 2.2). Figure 3a and Figure 3b depict the formation of triiodide in the reactor during the 109 mL/min operating condition, with 30 and 25 LEDs activated, respectively. Each figure displays the high concentration of triiodide formed in the fluid layer adjacent to the UV LEDs. The highly concentrated triiodide layer is convected and dispersed in accordance to the hydraulics of the reactor. The light intensity is unable to penetrate deeply through the reactor as a result of the triiodide formation and low transmissivity conditions of the iodide/iodate solution. This low transmissivity solution results in most of the triiodide production in the fluid layer adjacent to the UV LEDs. An analogous triiodide concentration profile (not shown here) was found for the 190 mL/min operating conditions; however, as expected, the formation of triiodide was less efficient at the higher flow rate.
Experimental and numerical evaluation of a UV-LED point of use device

Figure 3. Triiodide formation under 109 mL/min flow conditions a) 30 activated LEDs (260nm) b) 25 activated LEDs (260nm)

The experimental and numerical effluent triiodide molar concentrations determined at the outlet of the reactor are shown in Error! Reference source not found.. Although the model agrees well with the experimental results and shows the decrease in effluent triiodide concentration with increasing flow rate, the model seems to under predict the experimental triiodide concentrations at the low flow rate while over predict the results at the higher flow rate. The inaccuracies at the low flow condition likely stem from the model over-prediction of dispersion within the reactor. This inappropriate, over-characterization of dispersion decreases the efficiency of the reactor and results in an under-prediction of triiodide formation at the low flow rate.

Figure 4. Triiodide formation (M) under various operating conditions

Experimental and numerical evaluation of a UV-LED point of use device
3.3. Microbial Inactivation: Biodosimetry
The microbial response kinetics of MS-2 and Qβ were determined for 254nm low pressure (LP) lamps and 260nm LEDs (Error! Reference source not found.). The kinetics for the LP dose-response curve for MS-2 displays a slight tailing region at doses greater than 30 mJ cm$^{-2}$. This result is consistent with the study completed by Bowker et al., (2010). However, the MS-2 did not experience this tailing phenomenon when exposed to 260nm LEDs; rather, there was a log-linear relationship between fluence values ranging from 0-60 mJ cm$^{-2}$. The results also show an overall greater log inactivation was achieved for both Qβ and MS-2 at a LED wavelength of 260nm in comparison to LP lights (254nm). The higher overall log inactivation is a result of microorganisms having a peak DNA absorbance to light near 260nm (USEPA, 2006).

![Collimated Beam Dose Response Curve MS2 and Qβ](Image)

**Figure 5. Microbial response kinetics to 254nm low pressure and 260nm LEDs**

The continuous flow reactor was tested at two operating conditions; 109 mL/min flow rate at 92% UVT and 109 mL/min flow rate at 80% UVT. Both conditions were operated with all 30 LEDs (260nm) activated. Lab testing found that MS-2 was the more resistant bacteriophage, achieving only 0.63 log inactivation (92% UVT), while Qβ displayed more sensitivity to the LEDs, achieving more than 1.5 times greater log inactivation at the same UVT. Qβ was expected to result in higher log inactivation in comparison to MS-2 based on the more UV sensitive response kinetics for Qβ. Error! Reference source not found. compares the numerical and experimental results from the biodosimetry testing. The model shows good agreement between numerical predictions and experimental results for both phages at both operating conditions.

<table>
<thead>
<tr>
<th>Bacteriophage</th>
<th>Flow Rate (mL/min)</th>
<th>UVT (%)</th>
<th>Log Inactivation</th>
<th>Percent Difference (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Qβ</td>
<td>109</td>
<td>91.97</td>
<td>1.01</td>
<td>0.97</td>
</tr>
<tr>
<td>Qβ</td>
<td>109</td>
<td>81.10</td>
<td>0.77</td>
<td>0.82</td>
</tr>
<tr>
<td>MS-2</td>
<td>109</td>
<td>92.47</td>
<td>0.63</td>
<td>0.65</td>
</tr>
<tr>
<td>MS-2</td>
<td>109</td>
<td>80.50</td>
<td>0.48</td>
<td>0.54</td>
</tr>
</tbody>
</table>

**Figure 6. Comparison of experimental results and numerical simulations for log inactivation of bacteriophage Qβ and MS-2 at 109 mL/min flow rate (92% and 80% UVT).**
The relatively low log inactivation found in experimental testing may be a result of the limited power output of the LEDs. UV-LED technology in the germicidal range is currently at the embryonic stage of research; typically operating with 0.5-2% input power efficiencies and only able to generate approximately 1mW of power (Bowker et al, 2012). The LEDs used in this study averaged 0.5mW of output power. Such low output powers can result in a fraction of the light that is unable to penetrate large distances through the testing solution; this effect is compounded as UVT decreases. For those microorganisms that travel in areas where light is unable to penetrate, they will pass through the reactor unexposed to a significant amount of UV light. In addition, for those microorganisms that travel on pathways close to the LEDs, they must be exposed to light for large time periods in order to be inactivated.

4. CONCLUSIONS

In this study, CFD was used to evaluate the process of a continuous flow UV LED reactor. COMSOL Multiphysics was used to predict and validate the hydraulic characteristics, light distribution, and microbial log inactivation within the disinfection reactor. Analysis of residence time distribution data showed that the behavior of the reactor more resembles an ideal plug flow reactor. Although there was some mixing identified inside the reactor, the effect was not severe.

Experimental and numerical validation was also completed comparing the effluent triiodide concentration with 25 lights and 30 lights on, and at two flow rates: 109 mL/min and 190 mL/min. The model seemed to under predict the experimental triiodide concentrations at the low flow rate while over predict the results at the higher flow rate. A possible reason for the deviation with increasing flow rate is due to the difference in the model characterization of the dispersion that has occurred in the reactor, particularly at the 109 mL/min flow rate.

Biodosimetry testing included examining the kinetics of two non-pathogenic challenge microorganisms, Qβ and MS-2. The kinetics show that these bacteriophages are more sensitive to 260nm LEDs compared to the low pressure mercury-based 254nm lamps. The increased UV sensitivity is likely due to the greater DNA absorbance for both of the bacteriophages to 260nm light. The continuous flow experimentation resulted in higher Qβ log inactivation than the more UV-resistant phage, MS-2, and both saw decreasing log inactivation as UVT decreased. The model showed good agreement in predicting the log inactivation of both bacteriophages at a 109 mL/min flow rate and at two different UVTs (92% and 80%).

As technology and LED efficiency improves, the price of UV-C LEDs is expected to decline. The increased efficiency is likely to result in larger output power and decrease the required exposure time for inactivation of pathogenic microorganisms. The advancement of germicidal LED technology will make such point-of-use water disinfection units more economically viable, safer to handle, easier to dispose and longer lasting than mercury-based disinfection devices.

ACKNOWLEDGMENT

This research was funded by the National Science Foundation; project BES 0932116. We would like to thank Mr. Mohammad Arafin for his work in validating the hydraulic portion of this research.

References


