ABSTRACT

Lagrangian actinometry using dyed microspheres has been developed as a new method for validation of ultraviolet (UV) reactor systems. When properly applied, Lagrangian actinometry allows measurement of the UV dose distribution delivered by a UV reactor for a given operating condition. This method has been successfully applied to a series of UV disinfection systems, which were validated by biodosimetry as well. Excellent agreement was evident between the results from Lagrangian actinometry and biodosimetry, as well as those from corresponding numerical simulations. The ability of Lagrangian actinometry to physically measure UV dose distribution is extremely beneficial to the UV disinfection industry because it eliminates much of the uncertainty associated with conventional reactor validation approaches which depend solely on the application of biodosimetry. The combined application of Lagrangian actinometry, biodosimetry, and numerical simulation is therefore being proposed as a superior UV validation strategy to provide a more in-depth description of reactor performance than any of these methods individually, or in other combinations.

KEYWORDS: Disinfection, UV dose distribution, Lagrangian actinometry, dyed microspheres, ultraviolet, UV, validation.

INTRODUCTION

In drinking water and water reuse applications, ultraviolet (UV) disinfection systems must be validated based on physical measurements to ensure adequate performance relative to a treatment objective. Several protocols have been developed to describe appropriate methods for reactor validation; in all cases, the default method for reactor validation is biodosimetry (ÖNORM 2001, 2003; DVGW 2003; USEPA/UVDGM 2006; NWRI/AWWARF 2003). The primary advantage of biodosimetry is widespread familiarity with the method among the community of engineers and scientists involved in water treatment applications. However, biodosimetry assays all suffer from the inability to provide a measure of the UV dose distribution delivered by a UV reactor system. Because of this, the results of biodosimetry cannot be used to develop quantitative predictions of the inactivation response of any organism other than the challenge organism, unless the organism of interest has identical UV dose-response behavior to that of the challenge organism. As such, generous factors of safety are usually applied to the results of biodosimetric testing.

Previously, the only methods available to estimate UV dose distributions involved integrated numerical simulations of fluid mechanical behavior [usually based on Computational Fluid Dynamics (CFD)] and the UV intensity field; collectively, these models are now referred to as CFD-I simulations. However, the use of numerical models is complicated by uncertainty in the values of some critical input parameters and the complex nature of the models themselves. In addition, the results of numerical simulations must be validated against measured system behavior. Therefore, the development of an experiment-based method for measurement of the UV dose distribution delivered by a UV disinfection reactor represents a potentially important advance.

To address this need, a new method of UV reactor validation was developed based on the application of dyed microspheres (DMS). This method, termed “Lagrangian actinometry”, allows measurement of the UV dose distribution delivered by a UV reactor for a given set of operating conditions. Currently, Lagrangian actinometry has been successfully applied to different types of UV systems under a range of operation conditions. UV reactor testing by Lagrangian actinometry has been shown to provide accurate measurements of UV dose distributions delivered by UV disinfection systems and to provide excellent agreement with results from biodosimetry as well as numerical simulations.
Dyed Microspheres

Dyed microspheres involved in Lagrangian actinometry are prepared through conjugation of a biotinylated compound to streptavidin-coated polystyrene microspheres (Polysciences, Inc., Warrington, PA) as described previously (Blatchley et al. 2006). The compound, \((E)-5-[2-(methoxycarbonyl)-ethenyl]cytidine\) (hereafter referred to as \(S\)), yields a single photoproduct \(3\-\beta-D-\text{ribofuranosyl}-2,7\-\text{dioxopyrido}[2,3-d]pyrimidine\) (hereafter referred to as \(P\)) when subjected to germicidal UV radiation (Bergstrom et al. 1982). This reaction has been shown to have a high quantum yield across the germicidal UV spectrum (Shen et al. 2005); therefore, the \(S \rightarrow P\) system is an effective actinometer for germicidal UV radiation. In addition, the stable photoproduct \(P\) is brightly fluorescent with an excitation maximum at 330 nm and an emission maximum at 385 nm, whereas the starting material (\(S\)) is not. This allows the photoconversion of \(S\) to \(P\) to be quantitatively measured as an increase in fluorescence intensity (FI), which in turn allows measurement of UV dose delivery to an individual microsphere.

Dyed microspheres used in Lagrangian actinometry have a specific gravity of 1.05 and a mean diameter of 5.6 μm, which are similar to the physical properties of some waterborne microorganisms (e.g., protozoan (oo)cysts). Therefore, the trajectories of DMS are assumed to be similar to those of microorganisms when traveling through UV disinfection systems. Moreover, the action spectrum of DMS is also similar to that of several relevant waterborne microorganisms (and DNA) (see Shen et al. 2007). It has been hypothesized that such similarity should allow the exposure of DMS to polychromatic radiation to mimic the responses of challenge microbes when subjected to polychromatic UV radiation, and the results of DMS applications to polychromatic reactors support this hypothesis (Shen et al. 2007). Therefore, the application of a large population of DMS to a UV system, with appropriate sample collection downstream of the irradiated zone, allows measurement of the UV dose distribution. Figure 1 provides a conceptual representation of Lagrangian actinometry through the use of dyed microspheres.

The fluorescence intensity (FI) of dyed microspheres is quantified through flow cytometry. Thousands of DMS are analyzed individually in a matter of seconds, and the collected information will yield a distribution of FI. The FI distribution may be presented in the form of a histogram (see Figure 2).

UV Dose Distribution Estimation by Lagrangian Actinometry

Lagrangian actinometry requires definition of the UV dose-response (DR) behavior of DMS. DR testing of DMS is quantified by typical collimated beam experiments (USEPA/UVDGM 2006) using a low-pressure collimated beam apparatus with monochromatic output at wavelength of 254 nm. Results from the collimated-beam experiments (Figure 2) provide detailed information regarding the FI distribution behavior of the DMS over the range of relevant UV doses for the reactor system of interest. Non-linear regression is applied to the DR data to allow interpolation of DMS DR behavior at UV doses that fall between those used in the actual DR experiment.

DMS are also imposed on a continuous-flow reactor and collected downstream of the irradiated zone of the reactor that is being validated. The flow-cytometric analysis of a DMS testing sample will also generate a FI distribution (see Figure 3). Numerical deconvolution of these data and the DR results allows estimation of the UV dose distribution delivered by a reactor for the operating conditions corresponding to the test (see example in Figure 4).
APPLICATION OF LAGRANGIAN ACTINOMETRY TO UV DISINFECTION SYSTEMS

Lagrangian actinometry has been applied to several UV disinfection systems, ranging in size from bench-scale, single-lamp reactors to field-scale systems with hundreds of lamps. Lagrangian actinometry has been used to provide UV dose distribution estimates for UV reactors based on low-pressure (LP), LP high-output (LPHO) lamps, medium-pressure (MP), and excimer lamps. Operating conditions for these tests have covered wide ranges of flow rate, water transmittance (UVT), and system power levels. DMS tests have been conducted on UV reactors used for drinking water and water reuse applications. The results from these applications have demonstrated the validity and accuracy of Lagrangian actinometry. The method has also been demonstrated to provide highly repeatable results based on measurements of replicate samples collected from various reactors.

Perhaps the most important aspect of the results of Lagrangian actinometry experiments conducted to date is the fact that when combined with measured UV dose-response behavior for challenge organisms used in parallel or simultaneous biodosimetry experiments, the UV dose distribution estimates developed by Lagrangian actinometry yield excellent agreement with measured inactivation responses from the biodosimetry experiments. Moreover, these results have been in agreement with biodosimetry results even when more than one challenge organism was tested in a reactor for a given set of operating conditions. Figure 5 illustrates the results of such a comparison for a large-scale LPHO reactor tested for the NYC Catskill/Delaware UV disinfection facility; the predictions of challenge organism inactivation based on the UV dose distribution estimates from the DMS test were in excellent agreement with measured inactivation of coliphage MS-2 and Qβ for all operating conditions tested.

When available, UV dose distribution estimates by Lagrangian actinometry were also compared with the results from numerical simulations (CFD-I) using the same operating conditions as the simulation inputs. Again, good agreement was demonstrated among these methods of reactor characterization, as shown in Figure 4.

What can Lagrangian Actinometry Bring to Us?

As described previously, all existing validation protocols for UV reactors are based on biodosimetry. Because biodosimetry cannot yield a UV dose distribution measurement, validation protocols based on biodosimetry must employ uncertainty factors to define reactor operating conditions that will safely meet treatment objectives. These safety factors are intended to account for uncertainty in UV dose-distribution delivery by the reactor, as well as uncertainty associated with the use of microorganisms in a validation protocol.
In contrast with biodosimetry, Lagrangian actinometry yields an estimate of the UV dose distribution. In principle, the results of microspheres tests can be used to yield accurate predictions of the performance of a UV reactor for any microorganism, as long as reliable information regarding the UV dose-response behavior for the organism(s) of interest is available. To illustrate this point, the results of biodosimetry and Lagrangian actinometry conducted on the same ITT/Wedeco reactor were used to define the RED bias factor ($B_{\text{RED}}$) according to protocols defined in the UV Disinfection Guidance Manual (USEPA/UVDGM 2006). The UVDGM incorporates the “RED bias” ($B_{\text{RED}}$) as an uncertainty factor in calculating the credited RED or $\log_{10}$ inactivation of a target organism for a specific validated UV unit. This factor is a direct multiplier when sizing a UV system or in determining the operating status of a UV system.

Based on MS2 biodosimetry, $B_{\text{RED}}$ for the system is approximately 1.7. An argument can be made that $B_{\text{RED}}$ could be assigned a value of unity for the microspheres assay, given that the UV dose distribution is known by direct measurement. However, for purposes of this presentation, a conservative approach has been maintained. Following the protocols outlined in the UVDGM, a $B_{\text{RED}}$ value of 1.1 was determined from the microspheres results. According to Table 1, a 35% reduction in electrical service requirement and an annual electricity cost saving of nearly $1,000,000 (based on a rate of $0.12/kWh) can be achieved for a facility with operating flow of 1.5 BGD (235,000 m$^3$/h) by applying the lower $B_{\text{RED}}$ derived from the microspheres results. Even considering a small facility with an average operating flow of 10 MGD (1,577 m$^3$/h), such annual energy cost saving could still approximate $6,000. Note that this analysis considers only the reduction in energy costs, and ignores other potential O&M cost reductions such as lamp replacement and labor, which could equal or exceed the energy savings associated with the reduced system operations. Additionally, the ability to reduce the size of the system by up to one-third would have obvious implications relative to capital costs. Therefore, Lagrangian actinometry is beneficial not only economically, but also environmentally in terms of energy conservation.

### SUMMARY

Lagrangian actinometry using DMS represents the only available method to measure the UV dose distribution delivered by a UV reactor and reduce the uncertainty associated with predictions of process performance. Therefore, DMS tests represent a potential method for reducing the costs of UV disinfection by reducing the safety factors required for the current validation protocols. In addition, Lagrangian actinometry is also the only method whereby CFD-I numerical models can be validated at the level of the UV dose distribution. The method can be an invaluable tool in the optimization of an installed commercial system and in the development of new reactor designs.

A logical approach to reactor validation can involve the combined application of Lagrangian actinometry, biodosimetry, and numerical simulations. Specifically, Lagrangian actinometry and biodosimetry could be examined over the range of operating conditions for which the reactor is designed. The results of these tests can then be compared with each other for purposes of validation of the reactor, as well as the test methods. In turn, these results can also be used for validation of a numerical model. If properly validated over the range or relevant operating conditions, it should then be possible to develop accurate predictions of reactor performance at operating conditions that fall within the range of conditions over which the model was validated by comparing with results of Lagrangian actinometry and biodosimetry. Ultimately, one can suggest that the actinometric approach, when combined with well-developed collimated beam UV dose-response information for specific microbes, can satisfy in itself the validation requirements for new systems. CFD-I models that employ DMS UV dose-distribution data for calibration and verification will be enhanced, and gain greater confidence in their predictive use for reactor optimization and the commissioning of installed systems.

### Table 1. Examples of the impact of the DMS method for measurement of UV dose delivery with respect to electrical service demand and electrical power costs with LPHO UV systems.

<table>
<thead>
<tr>
<th>System</th>
<th>Average Service Flow (MGD)</th>
<th>Surrogate for Validation</th>
<th>Required Power Service (kW)</th>
<th>Annual Power Cost ($ US)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Large Facility</td>
<td>1500</td>
<td>MS-2</td>
<td>2800</td>
<td>2,950,000</td>
</tr>
<tr>
<td></td>
<td></td>
<td>DMS</td>
<td>1800</td>
<td>1,908,000</td>
</tr>
<tr>
<td>Small Facility</td>
<td>10</td>
<td>MS-2</td>
<td>18.7</td>
<td>19,000</td>
</tr>
<tr>
<td></td>
<td></td>
<td>DMS</td>
<td>12.1</td>
<td>13,000</td>
</tr>
</tbody>
</table>
ACKNOWLEDGEMENTS
The authors wish to acknowledge the significant support provided by the New York State Energy Research and Development Authority (NYSERDA) and the American Water Works Association Research Foundation (AWWARF).