Korean J. Chem. Eng., 36(11), 1785-1790 (2019)
DOI: 10.1007/s11814-019-0353-4

pISSN: 0256-1115
eISSN: 1975-7220

Inactivation of *Escherichia coli* and MS2 coliphage via singlet oxygen generated by homogeneous photosensitization

Taewan Kim*, Hyung-Eun Kim, Jiyoon Cho***, Hak-Hyeon Kim***, Jiwon Seo***, Junghun Lee***, Joon-Young Choi****, and Changha Lee***,†**

*School of Urban and Environmental Engineering, Ulsan National Institute of Science and Technology (UNIST), 50 UNIST-gil, Ulsan 44919, Korea
**Center for Water Resource Cycle Research, KIST School, Korea Institute of Science and Technology (KIST), 5 Hwarang-ro 14-gil, Seongbuk-gu, Seoul 02792, Korea
***School of Chemical and Biological Engineering, and Institute of Chemical Process (ICP), Seoul National University, 1 Gwanak-ro, Gwanak-gu, Seoul 08826, Korea
****Hyorim Industries Inc., 96-8, Yatap-ro, Bundang-gu, Seongnam-si, Gyeonggi-do 13517, Korea
(*Recevied 10 June 2019 • accepted 5 August 2019*)

Abstract−The inactivation kinetics of *E. coli* and MS2 coliphage by singlet oxygen ($^1O_2$) were investigated in a homogeneous photosensitization system using Rose Bengal (RB) and visible light illumination (the Vis/RB system). The inactivation of *E. coli* and MS2 in the Vis/RB system was monitored over time with variations of several parameters such as pH, light intensity, concentration of RB, and the presence of dissolved oxygen. In addition, the concentration of $^1O_2$ generated by the Vis/RB system was quantified using furfuryl alcohol under each microbial inactivation conditions. Based on the obtained results, the degree of microbial inactivation was quantitatively correlated with $^1O_2$ exposure using the (delayed) Chick-Watson model. The *Ct* (concentration-time product) values of $^1O_2$ required for 2 log microbial inactivation were found to be 1.3×10^{-4} mg·min/L for *E. coli* and 1.9×10^{-5} mg·min/L for MS2, respectively. The inactivation of *E. coli* exhibited an initial lag phase until 0.5×10^{-4} mg·min/L of *Ct*.

Keywords: *E. coli*, MS2 Coliphage, Singlet Oxygen, Disinfection, Inactivation Kinetics

INTRODUCTION

Microbial inactivation using reactive oxygen species (ROS) generated by photochemical reactions such as hydroxyl radical (•OH) and singlet oxygen ($^1O_2$) has been studied from the perspective of understanding natural disinfection processes driven by sunlight [1-15] as well as developing alternative disinfection technologies for water and wastewater treatment [1-6,10-15]. Different photochemical systems, including UV/H_2O_2, UV/TiO_2, UV/C_{60}, Vis/NOM, Vis/MoS_2, and Vis/edible dyes, have been tested for the inactivation of *E. coli*, MS2, and *B. subtilis* [1-6,11-15]. These studies also investigated the factors affecting the efficacy of microbial inactivation such as light intensity, pH, and temperature. It is known that ROS exert oxidative damages to cell membranes and other intracellular components [16-19].

As it is a more selective oxidant than •OH, $^1O_2$ can be more effective in inactivating microorganisms by avoiding undesired consumption through reactions with water constituents [20]. $^1O_2$ is readily generated by photosensitizers with visible light illumination in the presence of oxygen [21,22]. Microbial inactivation by $^1O_2$ has been examined using different visible light-photosensitizers such as organic dyes, metal complexes, and fullerenes [4,7-13,15]. These $^1O_2$-generating systems using visible light-photosensitizers have often been suggested to be applied as solar disinfection technologies for drinking water treatment in underdeveloped countries [15,23].

While there have been previous reports on microbial inactivation by $^1O_2$, only a few studies have quantified the microbicidal efficacy of $^1O_2$ based on the kinetics of microbial inactivation as a function of the $^1O_2$ exposure. In one example, Cho et al. [4] demonstrated the photosensitized inactivation of MS2 coliphage by functionalized fullerenes by quantitatively exploring the kinetics of MS2 inactivation by $^1O_2$. A recent study [15] also quantified the inactivation efficacy of MS2 by $^1O_2$ in a dye-photosensitization system using erythrosine. However, the microbicidal efficacies of $^1O_2$ on MS2 reported by the two studies were slightly different. In addition, the inactivation kinetics of *E. coli* by $^1O_2$ have yet to be quantitatively explored based on the measured $^1O_2$ exposure.

Therefore, our objective was to quantify the microbicidal efficacies of $^1O_2$ for the inactivation of *E. coli* and MS2. For this purpose, a homogeneous visible light-photosensitization system using Rose Bengal (RB) (Vis/RB system) was employed to generate $^1O_2$. The inactivation experiments of *E. coli* and MS2 were performed under different conditions (varying pH, light intensity, the concentration of RB, and the presence of dissolved oxygen) while monitoring the concentration of $^1O_2$ using a probe compound, furfuryl alcohol (FFA). The (delayed) Chick-Watson model was used to plot the log inactivation of *E. coli* and MS2 as a function of $^1O_2$ exposure.

†To whom correspondence should be addressed.
E-mail: leechangha@snu.ac.kr

MATERIALS AND METHODS

1. Reagents

All chemicals used in this study were of reagent grade and used without further purification. Agar, nutrient broth, tryptone, and yeast extract for *E. coli* and MS2 coliphage cultivation were purchased from Becton Dickinson Co. RB, FFA, D-(+)-Glucose, thiamine, calcium chloride, sodium chloride, sodium phosphate monobasic, sodium phosphate dibasic, L-histidine, and perchloric acid were obtained from Sigma-Aldrich Co. All solutions were prepared using deionized Milli-Q water (>18 MΩ·cm, Millipore Co.) and stored at 4 °C. All glassware was sterilized by autoclaving at 121 °C for 15 min prior to use.

2. Culture and Analysis of Microorganisms

E. coli (ATCC 8739) was inoculated in 30 mL of Difco nutrient broth and grown at 37 °C for 18-24 h. Next, the cells were harvested by 15 min centrifugation at 3,000 g and washed at least three times with 30 mL of phosphate buffered solution (PBS, 10 mM, pH 7.2). A stock suspension of *E. coli* was prepared by resuspending cells in 20 mL PBS (~10^9 CFU/mL), then stored at 4 °C until use. The spread plate method (using nutrient agar plates) was used to determine the cell populations of *E. coli* in the samples (undiluted and serially diluted). The plates after spreading were grown for 18-24 h at 37 °C, and the numbers of cell colonies were counted [24,25].

MS2 coliphage (ATCC 15597-B1) was grown with *E. coli* C3000 (ATCC 15597) as a host. The host *E. coli* was inoculated in tryptone broth containing 10 g/L tryptone, 1 g/L yeast extract, 8 g/L NaCl, 2 mM $CaCl_2$, 1 g/L glucose, and 0.01 g/L thiamine, and grown at 37 °C for 18-24 h under vigorous shaking. MS2 was grown in the host *E. coli* stock solution for 18-24 h at 37 °C without shaking. Next, the mixture of host *E. coli* and MS2 was centrifuged at 3,000 g for 15 min, and the supernatant was filtered with a 0.22 μm PTFE syringe filter in order to remove the cell debris of *E. coli*. The stock solution of MS2 produced contains ~10^{10} PFU/mL viral particles. The plaque assay method was then used to determine the populations of MS2 in samples (undiluted and serially diluted) [26]. Triplicate plates were used for the analyses of *E. coli* and MS2.

3. Photochemical Experiments

All of the experiments were performed in a quartz reactor with 50 mL of reaction solution under vigorous stirring at room temperature (24±1 °C). The reaction solution contained either RB and microorganisms (~10^7 CFU or PFU/mL) or other reagents such as FFA (100 μM) in order to measure $^1O_2$ and histidine for $^1O_2$ scavenging. The initial pH of the reaction solution was adjusted using phosphate buffer (10 mM); no significant variation in pH values were observed during the reaction. The reaction was initiated by illuminating light from a 150 W xenon arc lamp (LS 150, Abet Technologies Inc.) equipped with an AM 1.5 G filter and a 400 nm long pass filter. Aliquots of the samples (1 mL) were withdrawn from the reaction solution at predetermined time intervals and then analyzed to count the numbers of viable cells (or viral particles) following serial dilution with PBS. For the experiments with FFA, the residual concentrations of FFA in the samples were analyzed. All of the experiments were replicated at least three times; averages and standard deviations (error bars) are presented.

For the experiments requiring anoxic conditions, the reactor with the light source was placed inside an N_2 chamber, and the reaction solution was continuously sparged with ultrapure N_2 gas at a flow rate of 0.5 mL/min. The intensity of the incident light was varied between 50, 100, and 200 mW/cm^2 by adjusting the distance between the reactor and the light source.

4. Quantification of $^1O_2$ Concentration

The steady-state concentration of $^1O_2$ ($[^1O_2]_{ss}$) was calculated using the decomposition kinetics of the probe compound (FFA). The decomposition of FFA during the reaction exhibited pseudo-first order kinetics, expressed as follows in Eq. (1).

$$-\frac{d[FFA]}{dt} = k_{obs}[FFA] = k[^1O_2]_{ss}[FFA] \quad (1)$$

where k_{obs} is the observed pseudo-first order rate constant for the FFA decomposition and k is the second-order rate constant for the reaction of FFA with $^1O_2$ ($k=1.2\times10^8$ M^{-1} s^{-1}) [27]. The concentration of FFA during the reaction was monitored over time, and the k_{obs} value was obtained from the slope of the linear plot of $\ln([FFA]_0/[FFA])$ versus time. $[^1O_2]_{ss}$ was then calculated using Eq. (2).

$$[^1O_2]_{ss} = k_{obs}/k \quad (2)$$

The concentration of FFA was measured using high performance liquid chromatography (HPLC, Ultimate™ 3000, Dionex Co.) with UV absorbance detection at 219 nm. Separation was conducted on a ZORBAX Eclipse XDB-C18 column (5 μm, 150 mm× 4.6 mm, Agilent Co.). An 80 : 20 mixture of phosphoric acid (0.1%v/v) solution:acetonitrile was used as mobile phase at a flow rate of 1 mL/min.

5. Kinetic Models for Microbial Inactivation

Kinetic models were used to interpret the inactivation kinetics of *E. coli* and MS2 by $^1O_2$. The Chick-Watson model (Eq. (3)) [28,29] was applied to the inactivation of MS2.

$$-\mathrm{Log}(N/N_0) \propto Ct \quad (3)$$

where N and N_0 are the numbers of viable cells (or viral particles) at times t and 0, respectively, *C* is the concentration of disinfectant ($[^1O_2]_{ss}$ in this study), and *t* is the reaction time.

Unlike the inactivation of MS2, the inactivation curves of *E. coli* showed an initial lag phase. Therefore, the delayed Chick-Watson model (Eq. (4)) [30] was applied to the inactivation of *E. coli*.

$$-\mathrm{Log}(N/N_0)=0 \text{ (for } Ct<b), -\mathrm{Log}(N/N_0) \propto Ct \text{ (for } Ct \geq b) \quad (4)$$

where b is the lag coefficient.

RESULTS AND DISCUSSION

1. Role of $^1O_2$ in Microbial Inactivation by the Vis/RB System

The Vis/RB system was examined for the inactivation of *E. coli* and MS2 (Figs. 1(a) and 1(b), respectively), as well as for the decomposition of FFA (Fig. 1(c)). Control experiments with either light illumination or RB alone did not inactivate microorganisms. Under those conditions, the decomposition of FFA was also negligible. In the absence of oxygen (anoxic conditions), the microbial inactivation by the Vis/RB system was minor; *E. coli* and MS2 were inactivated by 0.7 log in 150 min and 0.6 log in 30 min, respectively.

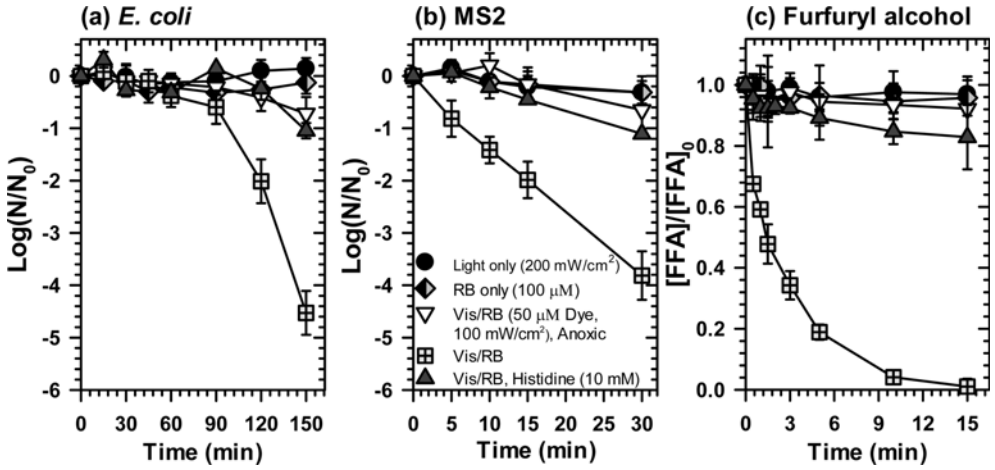

Fig. 1. Inactivation of *E. coli* (a) and MS2 coliphage (b) and decomposition of FFA under different experimental conditions ($[E.\ coli]_0$= ~10^7 CFU/mL for (a); $[MS2]_0$=~10^7 PFU/mL for (b); $[FFA]_0$=100 μM for (c); $[Histidine]_0$=20 mM; pH=7.1; unless specified otherwise, light intensity=100 mW/cm^2, [RB]=50 μM).

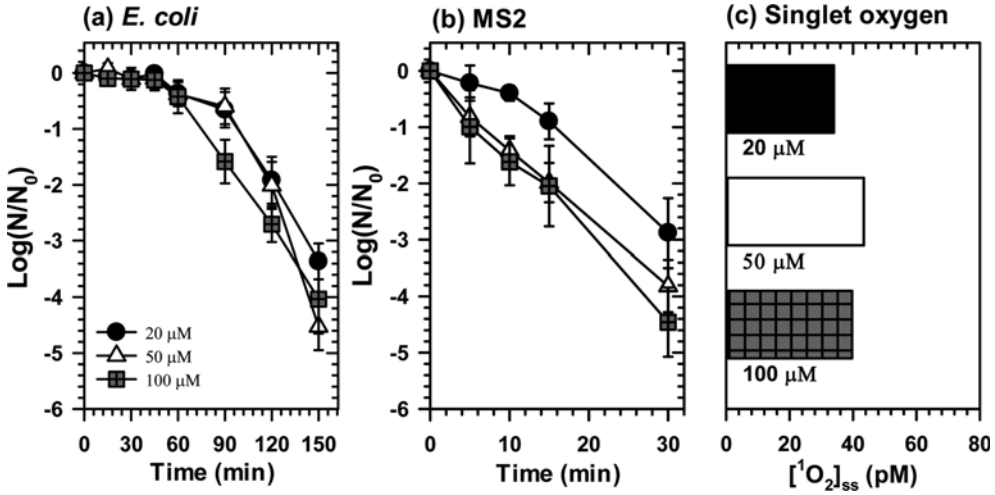

Fig. 2. Inactivation of *E. coli* (a) and MS2 coliphage (b), and the steady-state concentration of $^1O_2$ in the Vis/RB system at different concentrations of RB ($[E.\ coli]_0$=~10^7 CFU/mL for (a); $[MS2]_0$=~10^7 PFU/mL for (b); $[FFA]_0$=100 μM for (c); pH=7.1; light intensity=100 mW/cm^2).

In addition, the decomposition of FFA was negligible in the Vis/RB system under anoxic conditions, indicating that $^1O_2$ was not generated. The minor microbial inactivation by the Vis/RB system under anoxic conditions appears to have been caused by the interaction of photoexcited RB with cells (or viral particles).

The Vis/RB system in the presence of oxygen showed significantly inactivated *E. coli* and MS2 (Figs. 1(a) and 1(b)) with a noticeable decomposition of FFA (Fig. 1(c)), indicating that the Vis/RB system generates microbicidal $^1O_2$ by photosensitization. The Vis/RB system showed greater inactivation of MS2 than *E. coli*; *E. coli* and MS2 were inactivated by 4.5 log in 150 min and 3.8 log in 30 min, respectively. The inactivation curve of *E. coli* exhibited an initial lag phase. FFA was completely decomposed in 15 min, following pseudo-first order kinetics. According to the observed decay rate constant of FFA and using Eqs. (1) and (2), the steady-state concentration of $^1O_2$ was calculated to be 43 pM. When an excess amount of L-histidine (20 mM) as an $^1O_2$ scavenger was added into the system, the decomposition of FFA and microbial inactivation were greatly inhibited, confirming that $^1O_2$ is the responsible microbicidal species.

2. Effects of RB Concentration and Light Intensity

The inactivation of *E. coli* and MS2 by the Vis/RB system was examined with varying concentrations of RB ranging from 20 to 100 μM (Figs. 2(a) and 2(b)). Overall, the microbial inactivation was not significantly affected by the concentration of RB; only a slight enhancement was observed at higher RB concentrations. The $[^1O_2]_{ss}$ values calculated from the kinetics of FFA decomposition did not show substantial variations at different concentrations of RB (Fig. 2(c)). The minor influence of RB concentration indicates that the light absorption by RB is nearly saturated at these ranges of concentrations.

The effect of light intensity on the inactivation of *E. coli* and MS2 by the Vis/RB system was examined as well (Figs. 3(a) and 3(b), respectively). The results indicated that the microbial inactivation was accelerated with increasing incident light intensity. The inactivation efficacy of *E. coli* was enhanced from 2.1 log inactiva-

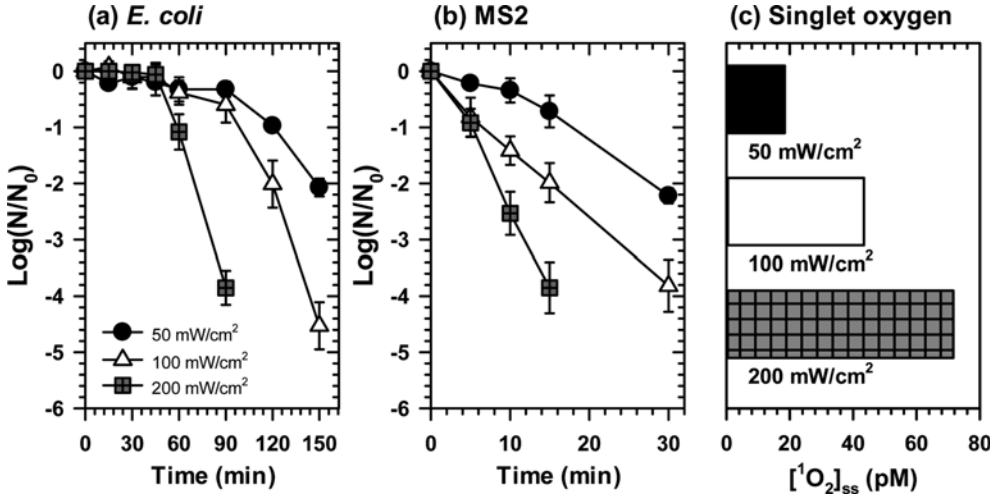

Fig. 3. Inactivation of *E. coli* (a) and MS2 coliphage (b), and the steady-state concentration of $^1O_2$ in the Vis/RB system at different light intensities ($[E. coli]_0$=~10^7 CFU/mL for (a); $[MS2]_0$=~10^7 PFU/mL for (b); $[FFA]_0$=100 μM for (c); pH=7.1; [RB]=50 μM).

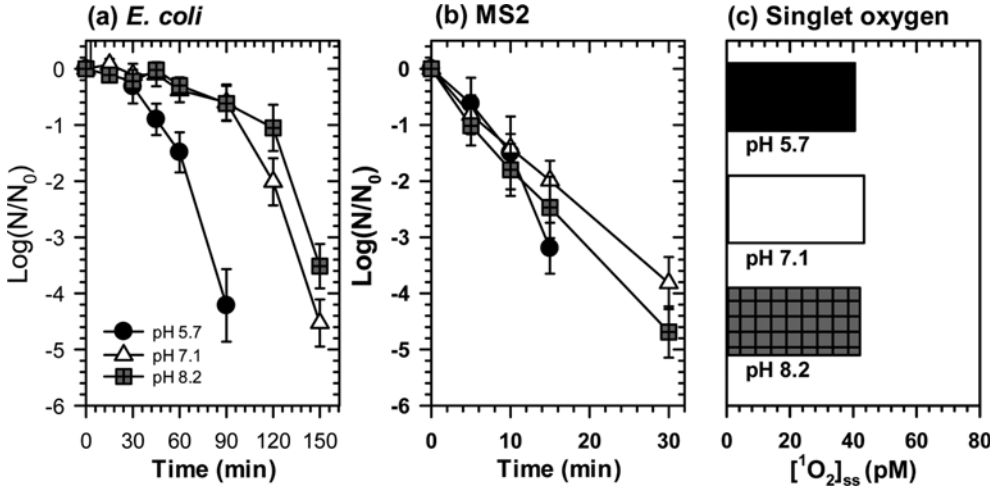

Fig. 4. Inactivation of *E. coli* (a) and MS2 coliphage (b), and the steady-state concentration of $^1O_2$ in the Vis/RB system at different pH values ($[E. coli]_0$=~10^7 CFU/mL for (a); $[MS2]_0$=~10^7 PFU/mL for (b); $[FFA]_0$=100 μM for (c); light intensity=100 mW/cm^2; [RB]=50 μM).

tion in 150 min at 50 mW/cm^2 radiance to 3.9 log inactivation in 90 min at 200 mW/cm^2 radiance. The inactivation efficacy of MS2 was enhanced from 2.2 log inactivation in 30 min at 50 mW/cm^2 radiance to 3.9 log inactivation in 15 min at 200 mW/cm^2 radiance. The $[^1O_2]_{ss}$ value also increased with increasing light intensity (Fig. 3(c)), showing almost a linear increase with the light intensity. The increased generation of microbicidal $^1O_2$ at higher light intensity is responsible for the enhanced microbial inactivation.

3. Effect of pH

The inactivation of *E. coli* and MS2 by the Vis/RB system was examined at different pH values ranging from 5.7 to 8.2 (Figs. 4(a) and 4(b)). The inactivation efficacy of *E. coli* was considerably higher at pH 5.7 than that at pH 7.1 and 8.2 (Fig. 4(a)). However, the inactivation efficacies of MS2 did not show large differences depending on pH (Fig. 4(b)). The $[^1O_2]_{ss}$ values almost remained constant, regardless of pH (Fig. 4(c)), indicating that the greater inactivation of *E. coli* at pH 5.7 was not caused by the higher yield of $^1O_2$. Similar results were obtained by a previous study [13] showing that the inactivation rate of *E. coli* at pH 4.5 was approximately eight-fold higher than that at pH 7.0 in the RB-photosensitized system at 25 °C, while the generation of $^1O_2$ was not pH-dependent. No clear explanation is currently available for the enhanced inactivation of *E. coli* and MS2 by the Vis/RB system at lower pH values. However, the change in pH may disturb the transport system of the cell membranes, facilitating the penetration of RB or $^1O_2$ into the cell inside [13,31].

4. Kinetic Modeling of Microbial Inactivation

Using the data on microbial inactivation and $[^1O_2]_{ss}$ in Figs. 2-4, the degrees of log inactivation of *E. coli* and MS2 were plotted as functions of *Ct* (concentration-time product, i.e., $[^1O_2]_{ss}t$) (Figs. 5(a) and 5(b), respectively); all of the data points in Figs. 2-4 were used in the plot, except for those of the *E. coli* inactivation at pH 5.7, which showed a substantial deviation from the $[^1O_2]_{ss}$ trend.

The *Ct* plot of *E. coli* inactivation exhibited a clearer shoulder

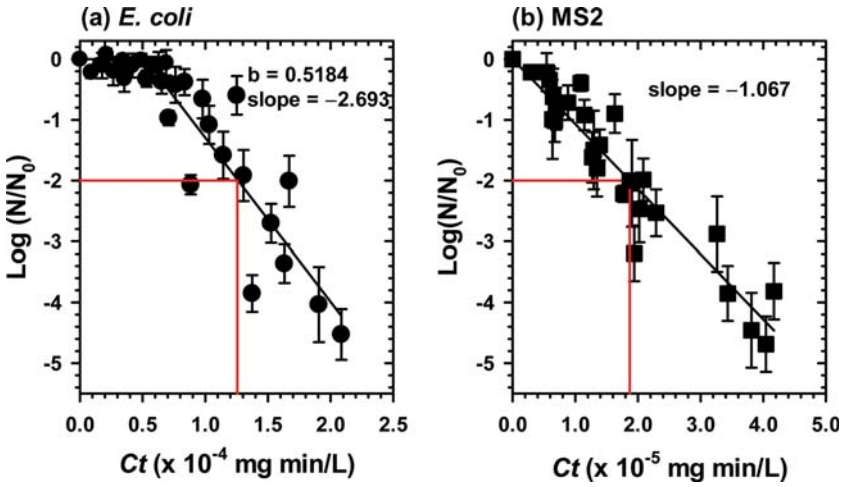

Fig. 5. Inactivation of *E. coli* (a) and MS2 (b) as a function of *Ct*. Symbols and lines represent the experimental data points and regression curves according to the (delayed) Chick-Watson model, respectively.

Table 1. *Ct* values required to achieve 2 log microbial inactivation (pH 7.1, buffered condition)

	Disinfectant		*Ct* value [mg min/L]	Reference
E. coli	Chlorine		1.7×10^{-1}	[28,32]
	Chlorine dioxide		$6.0\text{-}8.0\times10^{-2}$	[32,33]
	Ozone		$4.0\text{-}4.2\times10^{-3}$	[29,32]
	Hydroxyl radical	UV/TiO_2	8.0×10^{-6}	[2]
		UV/H_2O_2	4.5×10^{-9}	[5]
	Singlet oxygen		1.3×10^{-4}	This study
MS2 coliphage	Chlorine		1.4×10^{-1}	[34]
	Chlorine dioxide		1.7×10^{-1}	[34]
	Hydroxyl radical	UV/TiO_2	1.1×10^{-5}	[4]
	Singlet oxygen	Vis/C_{60}	1.1×10^{-5}	[4]
		Vis/erythrosine	5.0×10^{-6}	[15]
		Vis/RB	1.9×10^{-5}	This study

(the initial lag phase) (Fig. 5(a)), which is consistent with previous observations [2]. The lag phase in the inactivation curve of *E. coli* has often been observed for other oxidizing disinfectants such as $^{\bullet}OH$, free chlorine, and chlorine dioxide [2,5,28,29]. The cell membranes of *E. coli* can serve as barriers protecting the intracellular components from attacks by oxidants. Gram-negative bacteria such as *E. coli* (which have an outer cell membrane) showed larger lag phases than Gram-positive bacteria in the inactivation curves by $^1O_2$ [13]. The delayed Chick-Watson model (Eq. (4)) was applied to fit the kinetics of *E. coli* inactivation. The lag coefficient (b) and inactivation rate in the subsequent linear phase (the slope of the linear regression curve) were determined to be 5.2×10^{-4} mg·min/L and 2.7×10^4 log inactivation/(mg·min/L), respectively (Fig. 5(a)). The *Ct* value of $^1O_2$ required for the 2 log inactivation of *E. coli* was found to be 1.3×10^{-4} mg·min/L.

The inactivation of MS2 showed no lag phase (Fig. 5(b)), which may be attributable to the simpler structure of MS viral particles as compared to *E. coli* cells; MS2 has no barriers to shield from attacks by oxidants. The Chick-Watson model (Eq. (3)) was used to interpret the kinetics of MS2 inactivation. The inactivation rate of MS2 (the slope of the linear regression curve) was determined to be 1.1×10^5 log inactivation/(mg·min/L). The *Ct* value of $^1O_2$ required for 2 log inactivation of MS2 was 1.9×10^{-5} mg·min/L.

Table 1 summarizes the *Ct* values of oxidizing disinfectants required to achieve the 2 log inactivation of *E. coli* and MS2. The microbicidal efficacies of disinfectants as gauged from the *Ct* values (with lower *Ct* values indicating higher microbicidal efficacies) follow the order of chlorine≈chlorine dioxide<ozone<$^1O_2$<$^{\bullet}OH$, which is generally consistent with the order of oxidizing reactivity. The *Ct* value of $^1O_2$ for the inactivation of MS2 measured in this study (1.9×10^{-5} mg·min/L) was higher than that reported by previous studies (1.1×10^{-5} mg·min/L and 5.0×10^{-6} mg·min/L) [4,15] (also refer to Table 1); the different photosensitization systems used may have led to this discrepancy. A more drastic discrepancy in the disinfection kinetics has been reported for the inactivation of *E. coli* by $^{\bullet}OH$; the two *Ct* values reported in the literature were different by almost three orders of magnitude (8.0×10^{-6} mg·min/L for UV/TiO_2 and 4.5×10^{-9} mg·min/L for UV/H_2O_2, Table 1).

CONCLUSIONS

This study examined the inactivation kinetics of *E. coli* and MS2 coliphage by the Vis/RB system in which $^1O_2$ is generated as a microbicidal species via homogeneous photosensitization. Certain reaction parameters, such as pH, light intensity, concentration of RB, and the presence of dissolved oxygen, were found to affect the inactivation efficacy of *E. coli* and MS2 coliphage by the Vis/RB system. There was no significant microbial inactivation observed in the absence of oxygen or in the presence of an excess $^1O_2$ scavenger, indicating that $^1O_2$ is the responsible microbicide. The concentration of RB had a minor effect on microbial inactivation. Increasing the incident light intensity almost linearly increased the inactivation efficacies of *E. coli* and MS2. The effect of pH was minor for the inactivation of MS2, whereas the inactivation of *E. coli* was accelerated at lower pH values. By using the steady-state concentration of $^1O_2$ as measured by a probe compound, the microbial inactivation was found to be correlated with *Ct*; an initial lag phase could be observed in the inactivation curve of *E. coli*. The (delayed) Chick-Watson model was applied for the *Ct* plots. The *Ct* values of $^1O_2$ required for 2 log inactivation were found to be 1.3×10^{-4} mg·min/L and 1.9×10^{-5} mg·min/L for *E. coli* and MS2, respectively. These *Ct* values were higher than those of $^{\bullet}$OH but lower than those of free chlorine, chlorine dioxide, and ozone.

ACKNOWLEDGEMENTS

This work was supported by the Korea Ministry of Environment as an "Advanced Industrial Technology Development Project" (2017000140005) and as "Industrial Facilities & Infrastructure Research Program" (88107), and by NRF-2017-Global Ph.D. Fellowship Program.

REFERENCES

1. T. Kohn and K. L. Nelson, *Environ. Sci. Technol.*, **41**, 192 (2007).
2. M. Cho, H. Chung, W. Choi and J. Yoon, *Water Res.*, **38**, 1069 (2004).
3. Y. Horie, D. A. David, M. Taya and S. Tone, *Ind. Eng. Chem. Res.*, **35**, 3920 (1996).
4. M. Cho, J. Lee, Y. Mackeyev, L. J. Wilson, P. J. J. Alvarez, J. B. Hughes and J.-H. Kim, *Environ. Sci. Technol.*, **44**, 6685 (2010).
5. H. Mamane, H. Shemer and K. G. Linden, *J. Hazard. Mater.*, **146**, 479 (2007).
6. C. Liu, D. Kong, P. -C. Hsu, H. Yuan, H.-W. Lee, Y. Liu, H. Wang, S. Wang, K. Yan, D. Lin, P. A. Maraccini, K. M. Parker, A. B. Boehm and Y. Cui, *Nat. Nanotechnol.*, **11**, 1098 (2016).
7. T. A. Dahl, W. R. Midden and P. E. Hartman, *Photochem. Photobiol.*, **48**, 345 (1987).
8. T. A. Dahl, W. R. Midden and P. E. Hartman, *Photochem. Photobiol.*, **48**, 605 (1989).
9. T. A. Dahl, W. R. Midden and D. C. Necker, *J. Bacteriol.*, **171**, 2188 (1988).
10. S. A. Bezman, P. A. Burtis, T. P. J. Izod and M. A. Thayer, *Photochem. Photobiol.*, **28**, 325 (1978).
11. E. M. Hotze, A. R. Badireddy, S. Chellam and M. R. Weisner, *Environ. Sci. Technol.*, **43**, 6639 (2009).
12. K. Müller-Breitkreutz, H. Mohr, K. Brivida and H. Seis, J. *Photoch. Photobio. B-Biol.*, **30**, 63 (1995).
13. M. Schäfer, C. Schmitz, R. Facius, G. Horneck, B. Milow, K.-H. Funken and J. Ortner, *Photochem. Photobiol.*, **71**, 514 (2000).
14. A. I. Silverman, B. M. Peterson, A. B. Boehm, K. McNeill and K. L. Nelson, *Environ. Sci. Technol.*, **47**, 1870 (2013).
15. E. Ryberg, C. Chu and J.-H. Kim, *Environ. Sci. Technol.*, **52**, 13361 (2018).
16. M. Cho, H. Chung, W. Choi and J. Yoon, *Appl. Environ. Microbiol.*, **71**, 270 (2005).
17. H. A. Foster, I. B. Ditta, S. Varghese and A. Steele, *Appl. Microbiol. Biotechnol.*, **90**, 1847 (2011).
18. M. Castro-Alférez, M. I. Polo-López and P. Fernández-Ibáñez, *Sci. Rep.*, **6**, 38145 (2016).
19. A. Amrullah, N. Paksung and Y. Matsumura, *Korean J. Chem. Eng.*, **36**, 433 (2019).
20. J. Brame, M. Long, Q. Li and P. Alvarez, *Water Res.*, **60**, 259 (2014).
21. C. S. Foote, *Science*, **162**, 963 (1968).
22. C. S. Foote, *Acc. Chem. Res.*, **1**, 104 (1968).
23. M. E. Jiménez-Hernández, F. Manjón, D. García-Fresnadillo and G. Orellana, *Sol. Energy*, **80**, 1382 (2006).
24. J. D. Buck and R. C. Cleverdon, *Limnol. Oceanogr.*, **5**, 78 (1960).
25. S. Y. Park and C. G. Kim, *Environ. Eng. Res.*, **23**, 282 (2018).
26. B. B. Wentworth and L. French, *Exp. Biol. Med.*, **135**, 253 (1970).
27. F. E. Scully, Jr. and J. Hoigné, *Chemosphere*, **16**, 681 (1987).
28. Y. Kouame and C. N. Haas, *Water Res.*, **25**, 1027 (1991).
29. N. K. Hunt and B. J. Mariñas, *Water Res.*, **31**, 1355 (1997).
30. J. L. Rennecker, B. J. Mariñas, J. H. Owens and E. W. Rice, *Water Res.*, **33**, 2481 (1999).
31. D. J. Müller and A. Engel, *J. Mol. Biol.*, **285**, 1347 (1999).
32. M. Cho, J. Kim, J. Y. Kim, J. Yoon and J.-H. Kim, *Water Res.*, **44**, 3410 (2010).
33. M. Cho, Y. Lee, H. Chung and J. Yoon, *Appl. Environ. Microbiol.*, **70**, 1129 (2004).
34. M. Cho, Doctoral dissertation, Seoul National University, Seoul, Korea (2005).