PERFORMANCE OF ANNULAR FLOW STERILIZER IRRADIATED BY A GERMICIDAL LAMP

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Theoretical and experimental studies were conducted on the UV-inactivation characteristics of *Bacillus subtilis* spores in an annular-flow sterilizer irradiated by a germicidal lamp located at the center of the inner tube.

Experimental results for sterilizer performance in laminar spore-suspension flow were well simulated dynamically and statically by theoretical considerations which incorporated multi-targets with single-hit model for the UV-inactivation kinetic and the diffuse light model for the angle characteristic of lamp, along with due attention to the parabolic velocity distribution and the negligible diffusion of spores. Scale on the outside wall of the inner tube was checked by the use of a cylindrical chemical actinometer.

Calculated examples elucidate the reason why careful inspection is needed of UV-inactivation kinetics, angle characteristics of lamp, and fluid mixing to evaluate performance of actual flow UV sterilizers at high extent of inactivation up to the order of 99.999%.

Introduction

In a line of research on sterilizers irradiated by a germicidal lamp (UV sterilizers), the authors⁹⁾ previously reported the UV-inactivation kinetics of *Bacillus subtilis* spores, widely used for examining sterility, at high extent of sterilization (survival fraction down to 10^{-6}) with the use of batch UV sterilizers. They indicated the applicability of multi-targets with single-hit model with consideration of a small portion of UV-resistant spores for the reasonable formulation of the kinetics in such a highly sterilized region. It is essential to estimate the absolute UV intensity and the effect of flow mechanics on spore inactivation to establish the design procedure of UV sterilizers in flow systems.

The authors studied the inactivation characteristics of *Bacillus subtilis* spores in an annular UV sterilizer in laminar flow to evaluate the effects of angle characteristic of a light source, scale on the quartz-sterilizer wall, and residence time distribution of fluid elements in any type of flow UV sterilizer.

1. Experimental Apparatus and Procedure

The schematic diagram of an annular UV sterilizer is shown in Fig. 1. The body of the sterilizer, E, consisted of an inner quartz tube with outside diameter of 92.3 mm and inside diameter of 86.3 mm, and an outer Pyrex tube with inside diameter of 103.6 mm. The light source, F, was a germicidal lamp, GL-30

(Toshiba, 200V-30W, $26\phi \times 868$), with the assigned illuminating length of 200 mm achieved by covering rest of tube with sheets of black paper. The lamp was fixed at the center of the inner quartz tube. The test-section length of the sterilizer was made to be 700 mm by covering the upper and lower part of the inside wall of the inner tube with sheets of black paper. Holder D was provided to avoid segregation of flow. Tubes, connectives and cocks were made of Teflon to minimize adhesion of spores. The whole apparatus was set in a sterilized room (number of falling germs was

Fig. 1 Schematic diagram of annular UV-sterilizer

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less than 4–5 per hour).

The spore suspension of *Bacillus subtilis* NRRI with concentration of the order of 10^2 to 10^6 cm^{-3} , was first stored in reservoir A, then flowed through head tank C and entered holder D. Flowing up in an annulus, the suspension was irradiated in the test section by light source F, then overflowed to be measured volumetrically at each run. Flow rate was controlled by cock B'. The spore suspension, sampled at the exit, was quantitatively analyzed by counting the number of viable spores⁶⁾. To determine the dynamic characteristics of the sterilizer, sampling was sequentially conducted after the suspension flow-rate became stationary and shutter G, covering the illuminating part of a lamp, was removed. The details of cultivation of *Bacillus subtilis* NRRI and preparation of spore were described elsewhere⁶⁾. Internal light-filtering effect by spore suspension was checked by nephelometry and ascertained to be negligible under the experimental conditions studied.

2. Absolute UV Intensity in an Annulus

2.1 Estimation by diffuse light model

It is known that the radiant distribution characteristic is diffuse for the 254 nm resonance line of mercury vapor emitted from a germicidal lamp³⁾. If the lamp is assumed to be a line source, the relative UV intensity at an arbitrary point, $P(r, z)$, beneath the lamp of length l can be represented by Eq. (1)¹⁾ with the coordinates shown in Fig. 2:

$$I(r, z) \propto \frac{1}{r} \left\{ \frac{l-z}{\sqrt{(l-z)^2 + r^2}} + \frac{z}{\sqrt{z^2 + r^2}} \right\} \quad (1)$$

Once we determine the absolute UV intensity with the use of a small vessel of chemical actinometer at the position of (r_s, z_s) beneath the lamp of length l_s , we can calculate the absolute intensity at point P by Eq. (2):

$$I(r, z) = \frac{\frac{1}{r} \left\{ \frac{l-z}{\sqrt{(l-z)^2 + r^2}} + \frac{z}{\sqrt{z^2 + r^2}} \right\}}{\frac{1}{r_s} \left\{ \frac{l_s - z_s}{\sqrt{(l_s - z_s)^2 + r_s^2}} + \frac{z_s}{\sqrt{z_s^2 + r_s^2}} \right\}} \cdot I_s(r_s, z_s) \quad (2)$$

An example is as follows. For a watch-glass sterilizer placed 50 mm from a germicidal lamp with an illuminating length of 200 mm, the experimental results are shown in Fig. 3 for the inactivation of *Bacillus subtilis* spores, the same microorganism as that used for the previous study⁹⁾, along with the calculated line according to Eq. (3)⁹⁾:

$$f(\theta) = a_1 \cdot f_1(\theta) + a_2 \cdot f_2(\theta) \quad (3)$$

where

$$f_j(\theta) = 1 - [1 - \exp\{-(\tilde{\phi}\tilde{\alpha})_j \cdot I \cdot \theta\}]^m \quad (j=1, 2) \quad (4)$$

$$a_1 + a_2 = 1 \quad (5)$$

Chemical actinometer

Fig. 2 Arrangement of light source and chemical actinometer to determine absolute UV intensity at $P(r, z)$

Fig. 3 Comparison of observed and calculated inactivation characteristics of *Bacillus subtilis* spores in a batch UV sterilizer; $I_s(500, 434) = 8.75 \times 10^{-6} \text{ mol} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ ($l_s = 868$)

The absolute UV intensity, I , was calculated to be $1.20 \times 10^{-4} \text{ mol} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ on the basis of the reference intensity, I_s , of $8.75 \times 10^{-6} \text{ mol} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ at a point 500 mm beneath a germicidal lamp, GL-30, with an illuminating length of 868 mm. The fractions of UV-sensitive and UV-resistant spores, a_1 and a_2 , the inactivation cross-sections, and target number are represented by Eqs. (6), (7) and (8)⁹⁾.

$$a_1 = 0.95 \quad (\tilde{\phi}\tilde{\alpha})_1 = 1.02 \times 10^4 \text{ m}^2 \cdot \text{mol}^{-1} \quad (6)$$

$$a_2 = 0.05 \quad (\tilde{\phi}\tilde{\alpha})_2 = 0.46 \times 10^4 \text{ m}^2 \cdot \text{mol}^{-1} \quad (7)$$

$$m = 5 \quad (8)$$

Observed values shown in Fig. 3 are in good agreement with theoretical values in which the effect of reflection of light on the surface was neglected.

Fig. 4 Coordinates for estimation of absolute UV intensity at an arbitrary point, $P(r, z)$, in an annulus

The absolute UV intensity at an arbitrary point $P(r, z)$ in the annulus shown in Fig. 4 was estimated by Eq. (2) with correction of the scale on the sterilizer wall, as discussed in the following section, and neglecting reflection and absorption of light by the quartz glass.

2.2 Measurement of UV intensity with a cylindrical chemical actinometer and definition of cleanness factor

Incident intensity depression caused by the scale on the reactor wall is unavoidable for photoreactors in general⁷⁾. The authors determined the absolute UV intensity by inserting a cylindrical quartz-tube actinometer into the annulus after each run. The tube, with inside diameter of 3.3 mm and length of 200 mm, contained potassium trioxalatoferate (III) solution⁴⁾.

The cleanness factor was defined as a measure of the filter effect of scale on the outside wall of the inner quartz tube by Eq. (9):

$$\sigma = I'/I'_0 \quad (9)$$

where I' is absolute UV intensity in the annulus after a certain period of usage, and I'_0 is that at the initial stage. The authors evaluated I' by Eq. (10), assuming incident beams to be parallel¹⁾ owing to the same length of actinometric tube as that of the illuminating section of the lamp, and the small diameter of actinometric tube (3.3 mm) compared with the large diameter of the lamp (26 mm) and the short distance between them (49 mm), and considering the effect of thickness of the cylindrical reactor wall, i.e. the effect of refraction⁸⁾, to be negligible for calculating UV intensity.

$$I' = \frac{(\pi/4)d^2}{d \cdot \phi_a} \cdot \frac{dc_R}{d\theta} = \frac{\pi d}{4\phi_a} \cdot \frac{c_R}{\theta} \quad (10)$$

where c_R is ferrous ion concentration produced by irradiation for θ , d is inside diameter of cylindrical tube, and ϕ_a is quantum yield for ferrous ion production⁴⁾.

3. Theoretical Consideration of Annular UV Sterilizer Performance in Laminar Flow

The velocity profile is represented for the fully developed laminar flow in an annulus by Eq. (10)²⁾.

$$u(r) = 2\bar{u} \frac{\{1 - (r/r_o)^2 + (1 - \kappa^2) \ln(r/r_o) / \ln(1/\kappa)\}}{\{(1 - \kappa^4)/(1 - \kappa^2) - (1 - \kappa^2) / \ln(1/\kappa)\}} \quad (11)$$

When the shape of *Bacillus subtilis* spore is assumed to be spherical with the diameter of ca. 1 μm , its diffusion coefficient is estimated to be of the order of $10^{-12} \text{ m}^2 \cdot \text{s}^{-1}$; then the spore has the little probability of getting out of a certain fluid element. Consequently, it would be permissible to consider that each spore may move within a segregated fluid element. Under such a condition, the flow-averaged survival fraction for flowing suspension at the sterilizer exit, f_{av} , can be given by Eq. (12):

$$f_{av} = \int_{r_i}^{r_o} 2\pi r \cdot u(r) \cdot f(\theta, r) dr / \int_{r_i}^{r_o} 2\pi r \cdot u(r) dr \quad (12)$$

$f(\theta, r)$ is calculated by Eq. (13):

$$f(\theta, r) = a_1 \cdot f_1(\theta, r) + a_2 \cdot f_2(\theta, r) \quad (13)$$

where

$$f_j(\theta, r) = 1 - \left[1 - \exp \left\{ -(\tilde{\phi}\alpha)_j \cdot I_s \cdot \sigma \cdot \frac{\bar{I}(r)}{I_s} \cdot \tau(r) \right\} \right]^m \quad (j=1, 2) \quad (14)$$

$$a_1 + a_2 = 1 \quad (15)$$

$$\tau(r) = \begin{cases} \theta & (\theta < t(r)) \\ t(r) & (\theta \geq t(r)) \end{cases} \quad (16)$$

$$t(r) = l_R / u(r) \quad (17)$$

$\bar{I}(r)$ in Eq. (14), the longitudinally averaged UV intensity at radial point r , can be calculated by inserting Eq. (2) into Eq. (17).

$$\frac{\bar{I}(r)}{I_s} = \frac{1}{l_p} \int_{z_p}^{l_R + z_R} \frac{I(r, z)}{I_s} dz \quad (18)$$

where

$$l_p = \begin{cases} u(r) \cdot \theta & (\theta < t(r)) \\ l_R & (\theta \geq t(r)) \end{cases} \quad (19)$$

$$z_p = \begin{cases} z_R + l_R - u(r) \cdot \theta & (\theta < t(r)) \\ z_R & (\theta \geq t(r)) \end{cases} \quad (20)$$

z_R is z -coordinate of the lower end of the test section, as seen in Fig. 4.

4. Experimental Results and Discussion

4.1 Dynamic characteristics

Figure 5 shows an example of the sequential relation of observed flow-averaged survival fraction, f_{av} , at the sterilizer exit and elapsed time, θ , after the shutter was removed, for *Bacillus subtilis* spores with the inactivation kinetic that m equals 5; a_1 and $(\tilde{\phi}\alpha)_1$ are 1.0 and $0.79 \times 10^4 \text{ m}^2 \cdot \text{mol}^{-1}$, respectively, under the condition that the initial spore concentration was $4.3 \times 10^6 \text{ cm}^{-3}$ and mean residence time was 43.2 s. The reference UV intensity for the lamp, I_s , was $7.2 \times 10^{-6} \text{ mol} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ at the position of 500 mm

Fig. 5 Dynamic inactivation characteristic of *Bacillus subtilis* spores in an annular UV sterilizer in laminar flow

beneath the lamp with an illuminating length of 868 mm. The cleanness factor of the sterilizer, σ , was 1.0. With the information mentioned above, theoretical values were calculated by Eq. (12) and are represented in the figure by the solid line. The theoretical line simulates the experimental results well. The figure indicates the period of unsteady state for about 300 seconds, considerably longer than the mean residence time because of longer residence time for spores flowing near the wall. It can be noted that the induction period for laminar-flow UV sterilizer is quite different from that for the plug-flow sterilizer at the extent of sterilization of 99.9%, although the behavior of the laminar-flow photoreactor was reported to be rather similar to that of the plug-flow one at relatively low extent of conversion¹⁰.

4.2 Characteristics at steady state

Examples of sterilizer performance at steady state are plotted in Fig. 6 for *Bacillus subtilis* spores with the inactivation kinetic represented by Eqs. (6), (7) and (8) under the condition of initial spore concentration on the order of 10^8 to 10^9 cm⁻³. The reference UV intensity for the lamp applied here was 8.75×10^{-6} mol·m⁻²·s⁻¹ at a position of 500 mm beneath the lamp with an illuminating length of 868 mm. The cleanness factor of the sterilizer wall was 0.71. Theoretical performance was calculated at steady state ($\theta \gg t_m$) by Eq. (12), and is represented by a thick solid line. The theoretical line simulates the experimental results well up to the high extent of sterilization of 99.999%. The dotted line in Fig. 6 represents the calculated results, neglecting the fraction of UV-resistant spores. It can be seen that the deviation of the dotted line from experimental data increases as the extent of sterilization becomes higher.

Flow-averaged survival fraction of *Bacillus subtilis* spores was calculated on the assumptions that the angle characteristic of light source was taken to be specular¹ and that the other conditions were the same as those for the case of the thick solid line in Fig. 6. Calculated performance is shown as a solid line in

Fig. 6 Steady-state inactivation characteristics of *Bacillus subtilis* spores in annular flow UV sterilizers

Fig. 7 Comparison of observed inactivation characteristics of *Bacillus subtilis* spores in an annular UV sterilizer in laminar flow with calculated curve assuming angle characteristic of lamp to be specular

too high in the high inactivation region by the specular light model even if the effect is relatively small at low extent of sterilization.

4.3 Effect of fluid mixing on annular flow UV sterilizers

Theoretical performance was calculated for an annular plug-flow UV sterilizer of the same geometry as that used in the present study by Eq. (20):

$$f_{av}(t_m) = \int_0^\infty f\left(\theta, \frac{r_o + r_i}{2}\right) \cdot \delta(\theta - t_m) d\theta = f\left(t_m, \frac{r_o + r_i}{2}\right) \quad (20)^*$$

where $f(\theta, (r_o + r_i)/2)$ was calculated by Eq. (13); the radial distance of $(r_o + r_i)/2$ (the center of annulus) was chosen to evaluate the light intensity distribution in the annular plug-flow sterilizer. The result is shown in Fig. 6 by a thin solid line. A broken line in the figure also represents the theoretical characteristic, calculated by Eq. (21), for a macrofluid-type mixed-flow⁵⁾ UV sterilizer of the same geometry:

$$f_{av}(t_m) = \frac{1}{t_m} \int_0^\infty f\left(\theta, \frac{r_o + r_i}{2}\right) \cdot \exp(-\theta/t_m) d\theta \quad (21)$$

The broken line indicates the lowest efficiency in sterilizer performance: youngest fluid elements leaving the mixed-flow sterilizer affect more as the extent of inactivation gets higher. The characteristics for these two extreme flow conditions suggest that it would generally be difficult to obtain a high extent of sterilization in actual flow UV sterilizers having short-circuit channels; it is preferable to use plug-flow types for achieving high inactivation.

Conclusion

Dynamic and steady-state performance was determined for the laminar suspension flow of *Bacillus subtilis* NRRI spores in an annular sterilizer irradiated by a germicidal lamp located at the center of the inner quartz tube. Experimental results were simulated well by the improved one-hit-killing model for the inactivation kinetic and by the diffuse light model for the angle characteristic of lamp, along with consideration of the scale on the sterilizer wall and the velocity distribution.

Extent of sterilization would be overestimated in the high-sterility region if the angle characteristic of light source were taken to be specular. It is difficult to achieve a high extent of inactivation with the use of flow sterilizers having short-circuit channels. It is best to apply a plug-flow UV sterilizer, and also advisable to use a laminar-flow sterilizer at steady state in that the flow pattern is distinct.

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* We can also derive Eq. (20) from Eq. (12).

Nomenclature

a	= constant	[—]
c_R	= ferrous ion concentration	[mol · m ⁻³]
d	= inside diameter of cylindrical actinometric tube	[m]
f	= survival fraction of spores	[—]
I	= light intensity	[mol · m ⁻² · s ⁻¹]
l	= length of light source	[m]
l_p	= defined by Eq. (18)	[m]
l_R	= test-section length	[m]
m	= target number	[—]
r	= radial distance from center-line of light source	[m]
r_i	= outside radius of inner tube	[m]
r_o	= inside radius of outer tube	[m]
t	= l_R/u	[s]
t_m	= $l_R/\bar{u}$, mean residence time	[s]
u	= linear velocity in annulus calculated by Eq. (11)	[m · s ⁻¹]
$\bar{u}$	= mean linear velocity	[m · s ⁻¹]
z	= longitudinal distance from end of light source	[m]
z_p	= defined by Eq. (19)	[m]
z_R	= z-coordinate of lower end of test section	[m]
α	= cross-section of UV absorption	[m ²]
δ	= δ -function	[s ⁻¹]
θ	= time	[s]
κ	= r_i/r_o	[—]
σ	= cleanness factor of outside wall of inner quartz tube	[—]
τ	= defined by Eq. (15)	[s]
ϕ	= quantum yield for UV inactivation of spore	[mol ⁻¹]
ϕ_a	= quantum yield for actinometry	[—]
<Subscripts>		
av	= averaged value for flowing suspension	
j	= 1: UV-sensitive; = 2: UV-resistant	
o	= initial stage or inside of outer tube	
s	= reference point or reference length	
λ	= wavelength	
<Superscripts>		
$\sim$	= averaged value for wavelength	
—	= averaged value for sterilizer length	
$'$	= actinometric study	

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SHORT COMMUNICATIONS

DENSITIES, VISCOSITIES AND AMINE DIFFUSIVITIES OF AQUEOUS MIPA, DIPA, DGA AND EDA SOLUTIONS

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Aqueous solutions of amines are frequently used for the removal of acidic gases from gas mixtures. Therefore, a knowledge of the diffusivities of amines in aqueous solutions of various concentrations is important in the rational design of absorption equipment. In our previous work²⁾, the diffusivities of MEA (monoethanolamine), DEA (diethanolamine) and TEA (triethanolamine) in aqueous solutions were measured and these measured diffusivities were found to be considerably well correlated by the empirical relation, $D/D_w = (\mu/\mu_w)^{-2/3}$.

In the present study, the diffusivities of MIPA (monoisopropanolamine), DIPA (diisopropanolamine), DGA (diglycolamine) and EDA (ethylenediamine) in aqueous solutions, together with the densities and viscosities of these amine solutions, were measured and compared with the above empirical relation.

1. Experimental

The diffusivities of MIPA, DIPA, DGA and EDA in aqueous solutions were measured by the diaphragm cell technique described by Stokes⁵⁾. The experimental apparatus and procedure were essentially the same as those in our previous work²⁾.

The densities of the aqueous amine solutions were measured by means of a 25 cm³ calibrated pycnometer, and the viscosities of the solutions were determined with an Ostwald viscometer with efflux times of several hundred seconds.

All the measurements were carried out at 25°C and atmospheric pressure.

2. Results and Discussion

The measured values of the densities ρ and viscosities μ of the aqueous solutions of MIPA, DIPA, DGA and EDA are given in **Table 1** as functions of amine concentration C . The estimated accuracies in the density and viscosity measurements are 0.01 % and 0.2 %, respectively. The viscosity data of Sada *et al.*³⁾ obtained at 25°C for aqueous MIPA and DIPA solutions are in good agreement with the present

Table 1 Densities and viscosities of aqueous MIPA, DIPA, DGA and EDA solutions at 25°C and 1 atm

Amine	Conc. of amines C [kmol/m ³]	Density ρ [kg/m ³]	Viscosity μ [mPa·s]
MIPA	0.199	997	0.943
	0.495	997	1.021
	1.65	998	1.417
	2.58	999	1.905
	3.69	1002	2.774
DIPA	0.497	1001	1.180
	0.902	1004	1.521
	1.75	1012	2.752
	2.13	1016	3.682
	2.78	1022	6.281
	3.17	1024	8.872
	3.53	1028	12.40
DGA	0.485	1002	1.044
	0.985	1006	1.234
	2.06	1018	1.826
	2.86	1026	2.504
EDA	0.501	996	0.994
	1.00	995	1.107
	1.54	994	1.248
	2.50	993	1.581
	3.60	993	2.112

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