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Kinetics of bacteria inactivation employing UV radiation under clear water conditions

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Abstract

Microbiologically contaminated water, in this case artificially infected with *Escherichia coli* was treated with low wavelength (253.7 nm) radiation in a laboratory reactor where all the significant operating variables were carefully measured and controlled. A modification of the series-event model was used to interpret the experimental data which were collected employing four different levels of the incident radiation arriving at the reactor. The developed model is based on a rather complex dependence with respect to the *E. coli* concentration and to the radiation that is effectively absorbed by the bacteria which was precisely quantified. The mathematical description of the kinetics has three parameters: (i) the threshold limit of bacteria damage ($n = 2$), (ii) the kinetic constant [$k = 9.03 \pm 0.36\ s^{-1}(cm^3\ W^{-1})^m$] and (iii) the reaction order with respect to the bacteria photonic absorption rate ($m = 0.205 \pm 0.015$). About 99.99% plus inactivation was reached in all cases for rather short effective contact times and predictions from the model agree very well with experimental data in the whole range of investigated variables.

Keywords: Disinfection; UV radiation; *Escherichia coli*; Kinetic model

1. Introduction

Microbiological contamination is a widespread problem and water is one of the most important vehicles for disseminating this type of pollution, contributing to the dispersion of bacteria, yeasts, fungi, spores, etc. Part of this contamination is the product of an uncontrolled discharge of biological wastes or domestic sewage systems without the corresponding treatment.

Typically, these problems are very often solved with chlorine (or its derivatives) disinfection, an old, low cost water treatment technology that is very efficient and has an extensive use. Alongside these advantages, it is well-known the existing of an important drawback resulting from the toxicity of some of the chlorine disinfection by-products (DBPs) produced by the interaction of chlorine and chlorine derivatives with organic substances either naturally existing in water, or resulting from improperly treated industrial or sanitary wastes [1]. Some of these DBPs have been already included in the existing lists of substances having mutagenic or carcinogenic properties [2]. During the last years, organizations of different origin have insisted in the need for a gradual substitution of chlorine for water disinfection and a request for more research efforts aimed at developing efficient alternatives having reasonable costs [3].

Among the viable substitutive alternatives to chlorine utilization there are several technologies, some of them already well established such as ozonization [4] and others that are being either in the first stages of application, for example UV radiation [5] or a different group under development recognized with the generic denomination of Advanced Oxidation Technologies (AOT) such as photocatalysis or a variety of methods using hydrogen peroxide [6].

One of the first evidences of the appropriateness of UV for disinfecting purposes can be found in the quote by Nagy [7] that mercury vapor lamps were used to sterilize water as far back as 1909. The microbiological treatment of water in hydrotherapeutic pools was studied by Gilpin et al. [8] in an application where the use of chlorine is clearly not recommended. The effective use of UV radiation for water disinfection was also shown by Kawamura et al. [9].

Subsequently, different encouraging results have been reported employing UV radiation for water disinfection such as the inactivation of enteric adenoviruses, poliovirus and coliphages in a well mixed tank [10], the feasibility study carried out by the US Environmental Pollution Agency [11] or the work reported by the French Compagnie Generale des Eaux [12].

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Nomenclature

A	area (cm^2)
CFU	colony forming units
$C_{Ec,i}$	*E. coli* concentration of species with state of damage i ($CFU\,cm^{-3}$)
DR	damaging reaction
e^a	local volumetric rate of photons absorption (LVRPA), $W\,cm^{-3}$ (Einstein $cm^{-3}\,s^{-}1$)
G	incident radiation, $W\,cm^{-2}$ (Einstein $s^{-1}\,cm^{-2}$)
I	specific intensity, $W\,cm^{-2}\,sr^{-1}$ (Einstein $s^{-1}\,cm^{-2}\,sr^{-1}$)
k_i	kinetic constant corresponding to DR i, s^{-1} $(cm^3\,s\,Einstein^{-1})^m$ or s^{-1} $(cm^3\,W^{-1})^m$
L_R	reactor length (cm)
m	reaction order with respect to e^a
$M_{Ec,i}$	microorganism in the state of damage i
n	number of events or threshold limit of damage
$\underline{N}$	CFU flux ($CFU\,s^{-1}\,cm^{-2}$)
$R_{Ec,i}$	reaction rate corresponding to the bacteria with a state of damage i ($CFU\,cm^{-3}\,s^{-1}$)
R_G	growth rate ($CFU\,cm^{-3}\,s^{-1}$)
sr	steradiant
t	time (s)
V	volume (cm^3)
x	Cartesian coordinate along the reactor length (cm)
$\underline{x}$	position vector (cm)

Greek letters

α	specific Napierian absorption coefficient of a chemical species ($cm^2\,g^{-1}$ or $cm^2\,mol^{-1}$)
$\alpha_{Ec,i}$	specific Napierian absorption coefficient of *E. coli* ($cm^2\,CFU^{-1}$)
κ	Napierian absorption coefficient (cm^{-1})
λ	wavelength (nm)
Φ	overall quantum yield ($mol\,Einstein^{-1}$)
Ω	solid angle (sr)
$\underline{\Omega}$	unit vector in the direction of radiation propagation

Subscripts

Ec	relative to *E. coli*
Fe^{2+}	relative to ferrous ion
Fe^{3+}	relative to ferric ion
i	relative to the damaging state i or to the species with a damaging level i
m	relative to reacting medium
R	relative to reactor
t	relative to tubing
T	relative to total
Tk	relative to tank
W	relative to reactor wall
λ	relative to wavelength
0	denotes initial value

Special symbol

$\langle\rangle$	means reactor volume averaged value

Practical applications of the UV technology have been communicated by Blatchley [13] in conventional disinfection of secondary effluents [14] as well as specific treatments for water reuse [15] and a variety of processes used in several countries of Europe [16]. Research in UV disinfection kept increasing at a substantial rate in the last few years [17–25].

It is known that the effect produced by UV radiation on microorganisms is a significant change in the DNA structure (pyrimidine dimerization) that has lethal consequences [26,27]. The radiation absorption spectrum of DNA has an important peak at a wavelength close to 254 nm that is the one corresponding to the main emission of low pressure, mercury lamps known as germicidal lamps. These contributions constitute a clear evidence that bacteria inactivation could be treated as a special type of chemical reaction induced by light.

Although different kinetic descriptions developed for chemical water disinfection have been adapted for processes employing UV radiation, a distinct approach was proposed by Severin et al. [28,29] in a pioneering work introducing particular models when light irradiation is used. They set forth two alternative ways for interpreting the attack on microorganisms: the multitarget model and the series-event model. Based on experimental evidences they reported their own preference for the last one. This contribution was the starting point for the development of the mathematical model described in this paper. Its main features and the principal differences will be pointed out along the description that follows. In this work, kinetic data were obtained with experiments operated in a specially designed batch reactor using *Escherichia coli* as the testing bacteria. The main objective of this report is to communicate a kinetic model that is independent of the employed experimental device, i.e., with intrinsic kinetic parameters that should be apt for scaling-up purposes. Consequently, the chemical reaction engineering approach aimed at the precise mathematical modeling of the whole process is the central part of this contribution. When a reaction kinetics independent of the reactor configuration is known, scale-up methods for continuous homogeneous photochemical systems are already available in the literature [30]. Eventually, the hydrodynamic description of the fluid flow may be more complex, but either with computed fluid mechanic commercially available codes or introducing residence time distribution functions to properly account for the actual exposure times of the fluid particles to the applied radiation, the task can be solved.

2. The kinetic model

The developed model is a modification of the series-event one originally proposed by Severin in his Ph.D. thesis [31]. The representation is based on the idea that an event is assumed to be a unit of microorganism damage. Events occur in a stepwise fashion and each step is considered a separate event. The rate at which an organism passes from one event to the next is formulated in terms of a kinetic model that is independent of the event level occupied by the microorganism. As long as the microorganism is exposed to UV radiation it collects damage. However, it exist a threshold limit. Organisms which have

reached an event level greater than the threshold limit are inactivated and those which are below this level survive. For this reason, part of the information needed for the model is to know this threshold limit. The original kinetic model was formulated in terms of a mixed second order kinetics with respect to the local light intensity and the concentration of microorganisms existing in each event level. However, in the experiments performed in a batch reactor, assumption was made that the light intensity was constant and the hypothesis was incorporated in the kinetic model (uniform intensity). Experiments were also carried out in a completely mixed, annular, flow-through reactor. In this case, the local light intensity distribution was approximately described with the radial model of incidence [32] and variations in the radiation field were artificially introduced adding to the water with the organism culture a UV light attenuating agent (parahydroxibenzoic acid). Kinetic data obtained in the batch system were not suitable to predict the performance of the continuous reactor.

Several assumptions are made in the modified model:

(i) The radiation field inside the disinfection reactor is not uniform. It may be altered due to absorption produced by the water environment and the bacteria and, with the exception of very special geometries, the radiation intensity takes on its maximum value at the point of radiation entrance into the reactor (from now on called reactor window). The model should be able to work in clear as well as in dark or fouled waters as long as their optical properties are amenable of quantification.

(ii) The phenomenon responsible for the damage caused to the bacteria is the radiation effectively absorbed by the microorganism. This is a local value that depends on the radiation arriving at the reactor window, the optical characteristics of the water under treatment and the radiation absorption property and concentration of the existing bacteria. The solution of the radiative transfer equation (RTE) inside the reactor provides this information. Thus, the Local volumetric rate of photons absorption by the bacteria must be quantified at each position and time inside the reactor separately from the filtering effect produced by other radiation absorbing substances present in the water to be treated.

(iii) The rate constant for each event level of the series, from now on called a damaging reaction (DR), is the same. This is a simplification that can only be relaxed at the expense of a much more complex model and could turn to be an unnecessary complication. It is supported on the idea that the rate at which an organism passes from one event to the next is independent of the event level occupied by the microorganism.

(iv) This work was performed using germicidal lamps; thus, the model can be restricted to monochromatic radiation. Hence, wavelength will not be an additional variable. The use of polychromatic radiation necessitates a more laborious extension of the proposed model dividing the existing wavelength interval in a discrete number of hypothetic monochromatic reactors for which all the required optical properties will have to be known. Moreover, the eventual different biological effect of each wavelength must also be taken into account.

The model is thought of as a series of consecutive "damaging reactions" (DRs) each one of them producing a partial alteration on the structure of the different chemical blocks that construct the DNA and RNA sequence of the bacteria. Thus

$$M_{\mathrm{Ec},0} \xrightarrow{k_1} M_{\mathrm{Ec},1} \xrightarrow{k_2} \cdots M_{\mathrm{Ec},i} \xrightarrow{k_{i+1}} \cdots M_{\mathrm{Ec},n-1} \xrightarrow{k_n} M_{\mathrm{Ec},n}$$
$$\text{with } i = 0, 1, 2, \ldots, i, \ldots, n \text{ and } k_1 = k_2 = \cdots = k_n = k \tag{1}$$

In Eq. (1) k is the kinetic constant corresponding to the passage from one event level of damage $M_{\mathrm{Ec},i}$ to the next $M_{\mathrm{Ec},i+1}$, i.e., for the DR number $i+1$. The number of event levels (partial damages) that are needed to reach the threshold limit of inactivation is n. For the model, each $M_{\mathrm{Ec},i}$ can be considered a different species. Hence, for a series-event process with a threshold limit equal to n, the number of different species is $n+1$, including those that have not modified their initial condition: $M_{\mathrm{Ec},0}$. Therefore, we will be forced to solve $n+1$ differential equations representing the inventories of bacteria corresponding to each different species. In practice these balances will be made on the basis of colony forming units (CFU) per unit volume which are the units adopted to express the bacteria concentration.

It can be seen that since the total concentration of the existing species are linked by an algebraic expression, the last equation needed to know the number of inactivated bacteria can be obtained from

$$C_{\mathrm{Ec},n}(t) = C_{\mathrm{Ec},0}(t=0) - \sum_{i=0}^{n-1} C_{\mathrm{Ec},i}(t) \tag{2}$$

Then, the solution of just n differential equations will be needed. For a system such as the one represented by Eq. (1) the following set of equations are proposed for the different DRs:

$$\begin{aligned}
&\text{For } i = 0: \quad R_{\mathrm{Ec},i}(\underline{x},t) = -kC_{\mathrm{Ec},i}[e^a_{\mathrm{Ec},i}(\underline{x},t)]^m + R_{\mathrm{G}};\\
&\text{For } i = 1, 2, \ldots, n-1: \quad R_{\mathrm{Ec},i}(\underline{x},t) = kC_{\mathrm{Ec},i-1}[e^a_{\mathrm{Ec},i-1}(\underline{x},t)]^m - kC_{\mathrm{Ec},i}[e^a_{\mathrm{Ec},i}(\underline{x},t)]^m + R_{\mathrm{G}};\\
&\text{For } i = n: \quad R_{\mathrm{Ec},i}(\underline{x},t) = kC_{\mathrm{Ec},i-1}[e^a_{\mathrm{Ec},i-1}(\underline{x},t)]^m
\end{aligned} \tag{3}$$

$e^a_{\mathrm{Ec},i}$ is the local volumetric rate of photon absorption by *E. coli* with an event level of damage that can go from $i=0$ to $i=n-1$. Thus, it is clear that with respect to Severin proposal four significant differences have been introduced:

1. There is always a spatial distribution of DRs because e^a_{Ec} is a function of position regardless the type of employed reactor.
2. The model considers that the effective lethal radiation is that *absorbed* by the surviving bacteria and not just directly

related to the existing incident radiation at each point inside the reactor. Therefore, e^a_{Ec} is also a function of time.

3. The DR is not assumed to be of first order neither with respect to the radiation absorbed by the bacteria nor to the local value of the incident radiation. Thus, this reaction order (m) with respect to the energetic variable must be obtained from experiments.
4. The possibility of bacteria reproduction during the treatment (R_G) is admitted. This addition is introduced to allow for the possibility of processing water containing substances that could be a suitable culture for the surviving bacteria. R_G can be treated as zero order kinetics for bacteria subjected to severe stress [33] or the more common exponential growing that renders a first order kinetics [34]. It is clear that R_G is always zero for step $i = n$.

These kinetic equations must be introduced into the mass balances and the value of the LVRPA (e^a) must be obtained from the solution of the radiative transfer equation (RTE). Both must be derived according to the type of experimental equipment that will be used such that, with the proper interpretation of the experimental data, the results can be of general validity and independent of the shape, size or configuration of the reactor. This procedure permits to obtain *process rates* (that depend on the fundamental physico-chemical variables) from the *observed rates of change* (that are also influenced by other variables such as volumes, flowrates, state of mixing, shape of the reactor, etc.). This is the type of result that can be used to design practical reactors having other sizes, shapes or operating modes.

3. Experimental apparatus

The reactor was a cylinder made of glass closed with two flat, circular windows made of quartz of Suprasil quality ($V_R = 74.5\ cm^3$). The reactor length was 4.9 cm and the inner diameter was 4.4 cm as shown in Fig. 1(a). Irradiation is produced by two tubular lamps placed at the focal axis of their respective, custom made, parabolic reflectors made of aluminum with Alzac® treatment.

Two different types of radiation sources were used: (i) two Philips TUV lamps having an input power of 15 W each (nominal output power equal to 3.5 W = 7.42 μEinstein s^{-1}) and (ii) two Heraeus UV-C lamps operated with an input power of 40 W each (nominal output power equal to 16 W = 33.9 μEinstein s^{-1}). They are low pressure mercury vapor lamps (Germicidal type) with one single significant wavelength emission at 253.7 nm (Fig. 2). The qualitative performance (spectral distribution) of these lamps has been verified in previous work with emission models and radiation detectors. Instead, the actual value of the input power to the reactor windows will be measured with actinometer experiments. Working with almost monochromatic lamps it is indifferent to use W or Einstein s^{-1} to indicate the radiation source output power. The second unit may be more suitable for modeling reactions with polychromatic light because it accounts for the fact that the energy carried by a photon (one quantum) is a strong function of wavelength. The lamp operation was monitored with voltamperimeters. Each window permitted

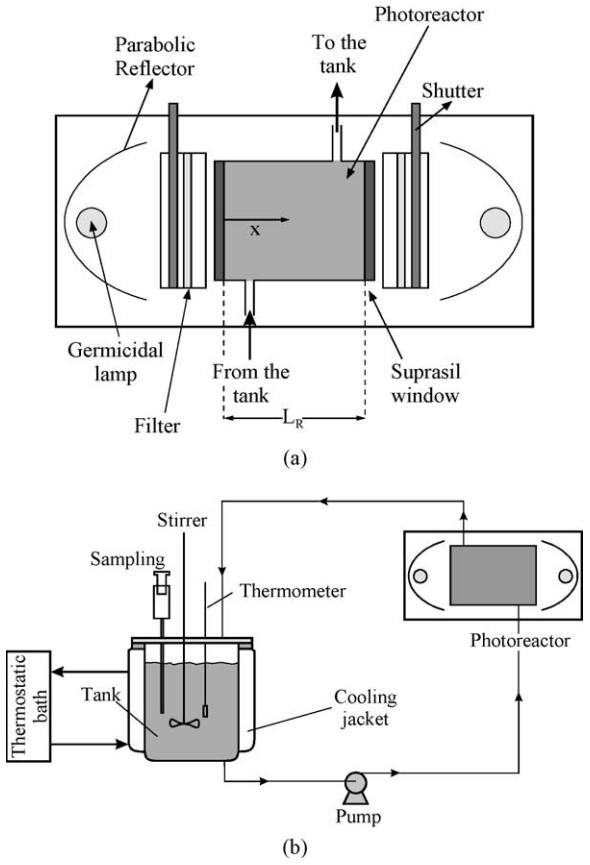

Fig. 1. (a) Schematic representation of the reactor; (b) flow-sheet of the experimental apparatus.

the interposing of: (1) one shutter to block the passage of light when desired (for example, to allow for the lamp to reach its steady state operation) and (2) neutral density filters to vary the irradiation rate from the lamps and reflectors allowing, with this device, two additional irradiation rates. Note that in this way four different levels of irradiation were obtained to permit the use of the irradiating condition as an additional independent variable and to be able to calculate the exponent m.

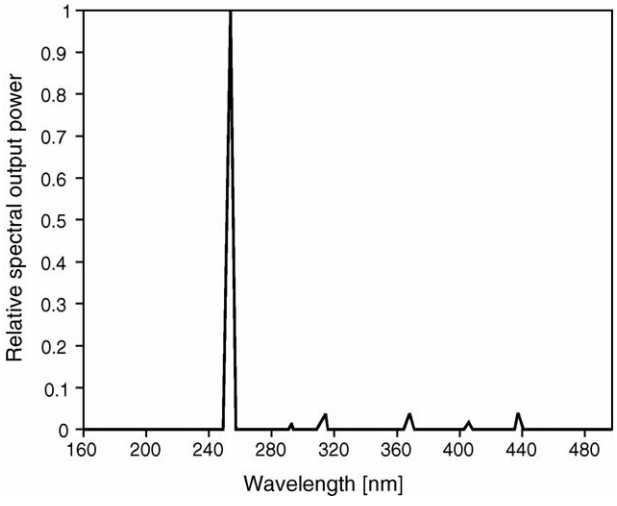

Fig. 2. Relative spectral output distribution of the employed lamps. The employed filters do not alter this relative distribution.

The liquid flow enters and leaves the reaction space through two exit ports placed at different longitudinal and angular positions along the reactor cylinder. The reactor was part of a recirculating system that included: (i) a well-stirred, large volume tank (1000 cm^3) with provisions for good mixing with mechanical stirring, sampling and temperature control and (ii) a recirculating pump (Masterflex Model 7553-76) as described in Fig. 1(b). Good mixing in the reactor was achieved, by means of an intense recirculation of the liquid. The tank was surrounded by a jacket that was connected to a water thermostatic bath (Haake) to keep the system temperature constant at 20 °C. Connections between the different components of the recycle were achieved with silicone tubing.

4. The CFU balance

The CFU balance can be derived employing the same tools that are used in the chemical reaction engineering literature for conventional mass balances. Starting from the general conservation equation written for an arbitrary event level of damage i:

$$\frac{\partial C_{\mathrm{Ec},i}(\underline{x},t)}{\partial t} + \underline{\nabla}\cdot \underline{N}_{\mathrm{Ec},i}(\underline{x},t) = R_{\mathrm{Ec},i}(\underline{x},t) \tag{4}$$

Under the following conditions: (1): isothermal operation, (2) very good mixing conditions, (3) the reactor volume (V_R) is much smaller than the tank and connecting lines volumes ($V_{Tk} + V_t = V_T - V_R$) which means that $V_R/V_T \ll 1$, (4) small changes in CFU concentration per pass in the reactor (derived from the high recirculating flowrate and the small reactor volume) it can be shown [30] that the time rate of change of the measured concentration in the tank results equal to

$$\left.\frac{\mathrm{d}C_{\mathrm{Ec},i}(t)}{\mathrm{d}t}\right|_{\mathrm{Tk}} = \frac{V_R}{V_T}\langle R_{\mathrm{Ec},i}(\underline{x},t)\rangle_{V_R} \tag{5}$$

where

$$\langle R_{\mathrm{Ec},i}(\underline{x},t)\rangle_{V_R} = \frac{1}{V_R}\int_{V_R} R_{\mathrm{Ec},i}(\underline{x},t)\,\mathrm{d}V \tag{6}$$

Eq. (6) represents an average of the reaction rate (taken over the reactor volume) corresponding to the bacteria (Ec) with an event level of damage i (species i), in order to account for a photon absorption rate that is a function of position inside the reactor and that the damaging reaction follows the same pattern. Thus, the final outcome will be represented by a volume average of different local inactivation rates. When the reactor is well mixed and the reactor cross-sectional area is constant, this averaging procedure can be taken over the reactor length L_R. The initial conditions are

$$t = 0: \quad i = 0, \quad C_{\mathrm{Ec},0}(t=0) = C^0_{\mathrm{Ec},0};$$
$$i = 1,\ldots,n, \quad C_{\mathrm{Ec},i}(t=0) = 0 \tag{7}$$

5. The radiation balance

Alfano et al. [35–37] have shown that, under some restricted geometric conditions that have been satisfied in our equipment, a combination of the parabolic reflector and the tubular lamp produces a radiation field that can be modeled as one-dimensional. Thus, for monochromatic radiation, illumination from one side and the employed device, the general RTE for a homogeneous medium in terms of the specific intensity:

$$\frac{\mathrm{d}I_{\lambda,\underline{\Omega}}(\underline{x},t)}{\mathrm{d}s} = -[\kappa_{\lambda,\mathrm{Ec}}(\underline{x},t) + \kappa_{\lambda,\mathrm{m}}(\underline{x},t)]I_{\lambda,\underline{\Omega}}(\underline{x},t) \tag{8}$$

can be reduced to the one-dimensional expression:

$$\frac{\mathrm{d}I_{\lambda,\underline{\Omega}}(x,t)}{\mathrm{d}x} = -[\kappa_{\lambda,\mathrm{Ec}}(x,t) + \kappa_{\lambda,\mathrm{m}}(x,t)]I_{\lambda,\underline{\Omega}}(x,t) \tag{9}$$

Considering that monochromatic radiation is employed, to simplify notation the subscript λ has been dropped. When irradiation is produced from both sides the result is

$$\frac{\mathrm{d}I_{\underline{\Omega}}(x,t)}{\mathrm{d}x} = -[\kappa_{\mathrm{Ec}}(x,t) + \kappa_{\mathrm{m}}(x,t)] \times [I_{\mathrm{I},\underline{\Omega}}(x,t) + I_{\mathrm{II},\underline{\Omega}}(L_R - x,t)] \tag{10}$$

The important photochemical property is the incident radiation; it is defined as follows:

$$G(\underline{x},t) = \int_{\Omega} I_{\underline{\Omega}}(\underline{x},t)\,\mathrm{d}\Omega \tag{11}$$

That is the collection of all intensities from all directions. For one-directional models it results equal to the one-dimensional intensity because only one direction of propagation is considered. Hence

$$\frac{\mathrm{dG}(x,t)}{\mathrm{d}x} = -[\kappa_{\mathrm{Ec}}(x,t) + \kappa_{\mathrm{m}}(x,t)][G_{\mathrm{I}}(x,t) + G_{\mathrm{II}}(L_R - x,t)] \tag{12}$$

With the boundary conditions:

$$x = 0: \quad G_{\mathrm{I}}(x,t) = G_{\mathrm{W,I}}(x,0);$$
$$x = L_R: \quad G_{\mathrm{II}}(x,t) = G_{\mathrm{W,II}}(x = L_R) \tag{13}$$

$G_{\mathrm{W,I}}(x=0)$ and $G_{\mathrm{W,I}}(x=L_R)$ are the incident radiation arriving at the windows of the reaction cell from the left and the right side respectively. Integrating Eq. (12) with boundary conditions given by Eq. (13):

$$G(x,t) = G_{\mathrm{W,I}}\exp -[[\kappa_{\mathrm{Ec}}(t) + \kappa_{\mathrm{m}}(t)]x] + G_{\mathrm{W,II}}\exp -[[\kappa_{\mathrm{Ec}}(t) + \kappa_{\mathrm{m}}(t)](L_R - x)] \tag{14}$$

In Eq. (14) κ_m is the absorption coefficient corresponding to the irradiated medium excluding absorption by surviving *E. coli* and κ_{Ec} is the surviving microorganism absorption coefficient. The last one comprises all living bacteria (without damage or with only partial damage). Thus, in symbolic form:

$$\kappa_{\mathrm{Ec}}(t) = \sum_{i=0}^{n-1}\kappa_{\mathrm{Ec},i}(t) = \sum_{i=0}^{n-1}\alpha_{\mathrm{Ec},i}C_{\mathrm{Ec},i}(t) \tag{15}$$

$\alpha_{Ec,i}$ corresponds to the specific absorption coefficient per unit CFU (Napierian absorptivity) of the *E. coli* having an event level of damage i. It is assumed that the specific absorption coefficients of all living "species" existing in the reacting system

are equal; i.e., from the radiation absorption point of view all "species" are undistinguishable. Moreover, eventually, if there were some slight differences among them, they cannot be measured. Then, one can write: $\alpha_{Ec,0} = \alpha_{Ec,1} = \cdots = \alpha_{Ec,i} = \alpha_{Ec}$. In this way a single value of the specific absorption coefficient is needed and can be obtained from spectrophotometric measurements and the application of the Beer's equation for low concentrations of solutes. Similar considerations are valid to calculate the absorption coefficient of the liquid solvent; i.e., the value of κ_m for the medium. κ_m is a very important property for dark or fouled waters because in this case it takes into account the existing significant changes in the radiation field of the reaction space.

In Eq. (14) the absorption coefficient has been assumed only a function of time because of the well-stirred conditions existing inside the reactor. Finally, the absorbed radiation by species i results equal to

$$\begin{aligned} e^a_{Ec,i}(x,t) &= \kappa_{Ec,i}(t) G(x,t) \\ &= \kappa_{Ec,i}(t)\{G_{W,I} \exp - [(\kappa_{Ec}(t) + \kappa_m)x] \\ &\quad + G_{W,II} \exp - [(\kappa_{Ec}(t) + \kappa_m)(L_R - x)]\} \end{aligned} \tag{16}$$

The absorption coefficient of the reacting medium has been assumed independent of time. The boundary condition can be obtained from radiation emission models [35–37]. For laboratory reactors it is more convenient to use actinometric measurements. Assuming that both lamps have equal emission characteristics, a single experiment for each irradiating condition will be sufficient because Eq. (16) reduces to

$$\begin{aligned} e^a_{Ec,i}(x,t) &= \kappa_{Ec,i}(t)\{G_W\{\exp - [(\kappa_{Ec}(t) + \kappa_m)x] \\ &\quad + \exp - [(\kappa_{Ec}(t) + \kappa_m)(L_R - x)]\}\} \end{aligned} \tag{17}$$

6. The RTE boundary condition

As indicated before, the boundary condition can be obtained from actinometric measurements. In this work, the potassium ferrioxalate method was employed [38] with the difference that our own method for the interpretation of the experimental data has been used. Essentially, a well-known reaction is used to know how many photons are entering into the reaction space. This is a reaction that, under well-specified experimental conditions, has a rate that is of first order with respect to the absorbed amount of photons; the proportionally constant is called the overall quantum yield (molecules of product formed by photon absorbed). The overall quantum yield is known as a function of wavelength for the employed concentration of the potassium ferrioxalate decomposition, allowing in this way to calculate, with the proper interpretation of the experimental results, the incident radiation at the reactor window. The reaction under consideration can be simply described by

$$2Fe^{3+} + C_2O_4{}^{2-} \xrightarrow{h\nu} 2Fe^{2+} + 2CO_2 \quad \text{with} \quad \Phi_{\lambda=253.7} \tag{18}$$

In the employed reactor, Eq. (5) results

$$\left.\frac{dC_{Fe^{2+}}(t)}{dt}\right|_{Tk} = \frac{V_R}{V_T}\langle R_{Fe^{2+}}(\underline{x},t)\rangle_{V_R} \tag{19}$$

With a reaction rate equal to

$$R_{Fe^{2+}}(x,t) = \Phi_{\lambda=253.7} e^a_{Fe^{3+},\lambda=253.7}(x,t) = \Phi e^a_{Fe^{3+}}(x,t) \tag{20}$$

The simplification in the notation is introduced because, with the employed radiation sources, only a single wavelength has to be considered. Φ is the overall quantum yield at $\lambda = 253.7$ nm, a constant value for a known range of ferric salts concentrations and independent of time and position. According to Eq. (17) the value of e^a is given by

$$\begin{aligned} e^a_{Fe^{3+}}(x,t) &= \kappa_{Fe^{3+}}(t)\{G_W\{\exp[-[\kappa_{Fe^{3+}}(t) + \kappa_{Fe^{2+}}(t)]x] \\ &\quad + \exp[-[\kappa_{Fe^{3+}}(t) + \kappa_{Fe^{2+}}(t)](L_R - x)]\}\} \end{aligned} \tag{21}$$

Thus, the problem reduces to extract from Eqs. (19)–(21) the value of G_W. The integration over the reactor volume [recall Eq. (6)] to get the average radiation absorption rate gives

$$\begin{aligned} \langle R_{Fe^{2+}}(x,t)\rangle &= \langle \Phi e^a_{Fe^{3+}}(x,t)\rangle = \Phi\langle e^a_{Fe^{3+}}(x,t)\rangle \\ &= \Phi\left\{\frac{2\kappa_{Fe^{3+}} G_W}{L_R[\kappa_{Fe^{3+}}(t) + \kappa_{Fe^{2+}}(t)]} \right. \\ &\quad \left. \times \{1 - \{\exp - [[\kappa_{Fe^{3+}}(t) + \kappa_{Fe^{2+}}(t)]L_R]\}\}\right\} \end{aligned} \tag{22}$$

The important point is that this equation can be applied to calculate the reaction rate when the reaction time approaches zero. Then, the concentration of Fe^{2+} is negligible ($\kappa_{Fe^{2+}} \cong 0$). Moreover, radiation absorption by Fe^{3+} is very high ($\kappa_{Fe^{3+}} \gg 1$) and the exponential term approaches zero. Hence, using the initial rate, the mass balance results

$$\left.\frac{dC_{Fe^{2+}}}{dt}\right|_{Tk}^{t\to 0} = \Phi\left(\frac{2V_R}{V_T L_R}\right) G_W \tag{23}$$

And the boundary condition is obtained from

$$G_W = \left(\frac{V_T}{2A_W\Phi}\right) \lim_{t\to 0}\left(\frac{C_{Fe^{2+}} - C^0_{Fe^{2+}}}{t - 0}\right) \tag{24}$$

The limit is very simple to calculate because for very low conversions of the ferric salt [the only condition where Eq. (23) is valid] the plot of $C_{Fe^{2+}}$ versus t is a straight line. The preparation of the reacting solution and the analysis of the ferrous salt were made according to the prescriptions described by Murov et al. [38]. Results for the two employed lamps, with and without filters are shown in Fig. 3. The employed concentration of $K_3Fe(C_2O_4)_3$ was 0.006 M and the value of Φ at 253.7 nm is 1.25 [39,40]. The values of the four different boundary conditions are reproduced in Table 1.

7. Final equations

To analyze the experimental results with *E. coli* we must apply Eq. (5) to each of the "species" coexisting in the reactor.

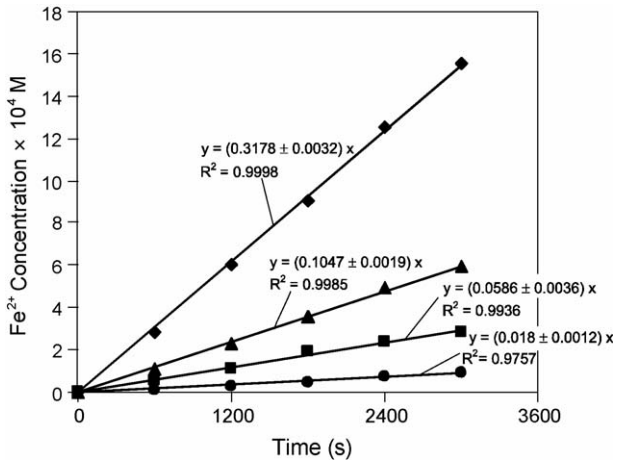

Fig. 3. Experimental results of actinometric measurements for four different conditions of irradiation of the reactor windows: (♦) Heraeus 40 W; (▲) Philips 15 W; (■) Heraeus whit filter; (●) Philips with filter.

Table 1
Incident radiation arriving at each of the reactor windows

Incident radiation (G_W)	Value ($\times 10^3$ μ Einstein cm^{-2} s^{-1})	Value (mW cm^{-2})
Heraeus, 40 W (100%)	14.95 ± 0.20	7.05 ± 0.09
Philips 15 W (100%)	5.85 ± 0.15	2.76 ± 0.07
Heraeus 40 with filter (17%)	2.55 ± 0.15	1.27 ± 0.08
Philips 15 W with filter (17%)	0.978 ± 0.06	0.45 ± 0.03

In each case, Eq. (6) must be used. Due to the well-stirred conditions, concentrations are uniform and can be taken outside the averaging integral. The only function that must be averaged over the reactor length $e^a_{Ec,i}(x,t)$. Unfortunately, this integration cannot be solved analytically for the arbitrary exponent m. Thus, all CFU balances must be solved numerically. In each case (i.e., every species) and for each time (because radiation absorption is a function of time) the following equation has to be calculated:

$$\langle [e^a_{Ec,i}(x,t)]^m \rangle_{L_R} = \frac{1}{L_R}\int_0^{L_R} [e^a_{Ec,i}(x,t)]^m \, dx \quad (25)$$

However, the only property that is a function of position is the incident radiation:

$$\frac{1}{L_R}\int_0^{L_R} [e^a_{Ec,i}(x,t)]^m \, dx = [(\alpha_{Ec})^m (C_{Ec,i}(t))^m]\frac{1}{L_R}\int_0^{L_R} [G(x,t)]^m \, dx = [(\alpha_{Ec})^m (C_{Ec,i}(t))^m]\langle [G(x,t)]^m \rangle_{L_R} \quad (26)$$

The equations to be solved representing the CFU balances have the following final form:

- For $i=0$:

$$\left.\frac{dC_{Ec,i}}{dt}\right|_{Tk} = -\frac{V_R}{V_T}\{kC_{Ec,i}(t)\{[(\alpha_{Ec})^m (C_{Ec,i}(t))^m] \times \langle [G(x,t)]^m \rangle_{L_R}\}\} + R_G \quad (27)$$

- For $i=1, 2, \ldots, n-1$:

$$\left.\frac{dC_{Ec,i}}{dt}\right|_{Tk} = \frac{V_R}{V_T}\{kC_{Ec,i-1}(t)\{[(\alpha_{Ec})^m (C_{Ec,i-1}(t))^m] \times \langle [G(x,t)]^m \rangle_{L_R}\} - kC_{Ec,i}(t)\{[(\alpha_{Ec})^m (C_{Ec,i}(t))^m] \times \langle [G(x,t)]^m \rangle_{L_R}\}\} + R_G \quad (28)$$

- For $i=n$:

$$\left.\frac{dC_{Ec,i}}{dt}\right|_{Tk} = \frac{V_R}{V_T}\{kC_{Ec,i-1}(t)\{[(\alpha_{Ec})^m (C_{Ec,i-1}(t))^m] \times \langle [G(x,t)]^m \rangle_{L_R}\}\} \quad (29)$$

$$t=0: \quad i=0, \quad C_{Ec,0}(t=0) = C^0_{Ec,0};$$
$$i=1,\ldots,n, \quad C_{Ec,i}(t=0) = 0$$

With the generic expression for the incident radiation given by

$$G(x,t) = G_W\{\exp -[(\kappa_{Ec}(t)+\kappa_m)x] + \exp -(\kappa_{Ec}(t)+\kappa_m)[(L_R - x)]\} \quad (30)$$

8. Experimental procedure

E. coli strain ATCC 8739 was used throughout this work. The purity of the strain was verified by conventional methods [41,42]. The culture was grown in two different types of broth: (i) a complex medium (nutritive broth) having as main component beef extract and (ii) a synthetic medium of well-known composition having as main component glucose. In the first case the broth composition was: triptone, 10 g L^{-1}; beef extract, 5 g L^{-1}; NaCl, 5 g L^{-1}. In the second case the broth was prepared according to the prescription described by Bailey and Ollis [34]. The working solution was prepared from a culture that had reached the stationary phase of growing and afterwards was brought to a 1/1000 dilution with physiological saline to simulate clear, transparent waters.

The specific absorption coefficients (Napierian absorptivities) of the two different culture media and *E. coli* were measured in a UV–vis Lamda 40 Perkin-Elmer Spectrophotometer at 253.7 nm. The results are shown in Table 2. Absorbance measurements for *E. coli* as a function of CFU concentrations are shown in Fig. 4.

Initial *E. coli* CFU concentrations ranged from 10^4 to 10^6 CFU cm^{-3}.

Table 2
Optical properties of the reacting medium components

Specific absorption coefficients	Units	Value at 253.7 nm
Nutritive broth	cm^2 g^{-1}	1284
Glucose broth	cm^2 g^{-1}	144
E. coli	cm^2 CFU^{-1}	1.38×10^{-9}

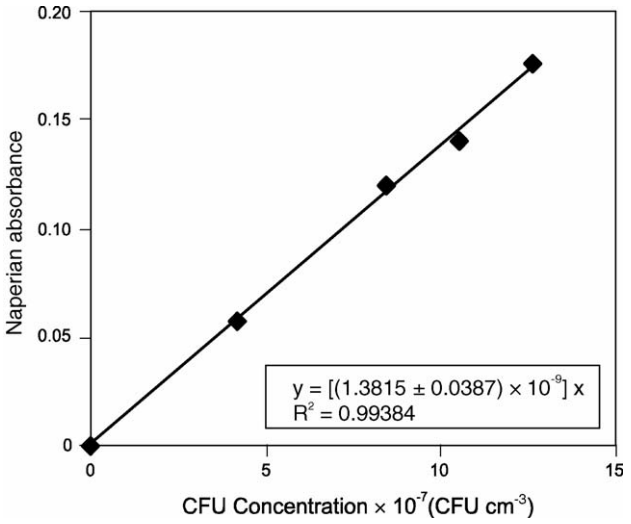

Fig. 4. Experimental data of *E. coli* Napierian absorbance.

The lamps were turned on, allowing for 30 min to stabilize their operation (during this time the shutters at the reactor windows were on). The working solution was added to the reactor. Immediately after, recirculation was established until the temperature gave a constant reading. The sample at $t = 0$ was taken at the same time that the lamp shutters were taken off. Afterwards, samples were taken at different time intervals for several measurements. After every run the whole equipment was carefully disinfected with sodium hypochlorite dilute solution, followed by dilute ethanol and several washing operations with distilled water. For every experimental condition, runs were duplicated.

Samples were taken initially every 10 s and afterwards every 60 s. A normal run lasted for 1800 s. It must be noted that due to the type of equipment used in this work (a recycle with a large volume tank) this time does not represent the one corresponding to the irradiation of the total reaction volume. Thus, the actual exposure to radiation must take into account the ratio given by V_R/V_T. Dilution of the samples to obtain the optimum concentration for the CFU counting method was made with sterile peptone water solution.

Each sample was subjected to the following triplicate measurements: absorbance a 253.7 nm and CFU counting using specific Pretrifilm™ plates (3 M Microbiology Products) for *E. coli* and coliform bacteria. This method has been recognized by the American Public Health Association in Standard Methods for the Examination of Dairy Products [41] and the AOAC (Association of Official Analytical Chemists International) in Official Methods of Analysis [43] as equivalent to the conventional plate method for this type of microorganisms. The plates were incubated, after spreading with the appropriate volume of the sample, during 24 h at 37 °C.

9. Results and discussion

The first series of runs were made with the lamps turned off. No growing of bacteria was observed during 120 min (this time is much longer than the one corresponding to a normal run with UV irradiation). These experiments permitted to conclude that under the described experimental conditions $R_G = 0$. Hence, when the medium is very dilute and essentially the water is free from nutritive substances, the possibility of bacteria growing is absent.

Afterwards, runs were made with the four different levels of irradiation (the four irradiation boundary conditions listed in Table 1).

Fig. 5(a) and (b) shows a typical experimental result for two levels of irradiation. The apparently long time required for inactivation of the existing bacteria is due to the existence of the tank with a ratio of $V_R/V_T = 0.07$ that permitted longer and more precise sampling time intervals, i.e., as said before, the effective irradiation time was much shorter. It can be seen that more than 99.99% change in bacteria concentrations was always achieved. Thus, it was confirmed that with low wavelength radiation ($\lambda = 253.7$ nm) and short effective contact times, under clear water conditions, bacteria inactivation can always be achieved.

Since $R_G = 0$ and α_{EC} and α_m were experimentally measured, once G_W is known according to Eq. (24) and the actinometric measurements, from Eqs. (7), (25)–(30) we have just three unknowns: n, m and k.

The obtained experimental information was compared with predictions resulting by solving numerically Eqs. (7), (25)–(30) and feeding the CFU concentration results and the experimental data to a nonlinear multiparameter regression estimation program. The values of n, m and k were obtained using all runs; i.e., with different initial bacteria concentrations, prepared with the two different cultures and inactivation runs employing four

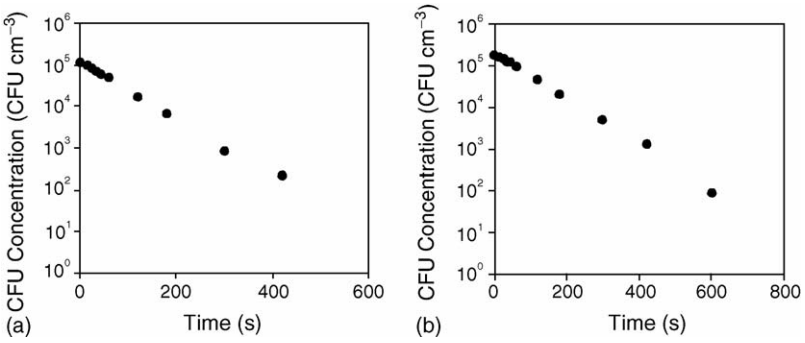

Fig. 5. Typical experimental data of bacteria inactivation corresponding to two levels of irradiation: (a) lamp of 40 W; (b) lamp of 15 W with filter.

levels of irradiation. Only integer numbers were allowed for the value of n.

The results with a 95% confidence interval are: $n = 2$; $m = 0.205 \pm 0.015$; $k = 9.0279 \pm 0.3579\ s^{-1}\ (cm^3\ W^{-1})^m$ or $k = 1.3142 \times 10^2 \pm 5.21\ s^{-1}\ (cm^3\ s\ Einstein^{-1})^m$.

The standard deviation for k is 3.96% and that for m is 7.32%. These results mean that the inactivation reaction has a very short induction time and, on the average, only two event levels of bacteria damage produced by the photons are enough.

These results indicate that the kinetics of *E. coli* inactivation is a function of the bacteria concentration and the rate of radiation absorption by the bacteria. The observed dependences are:

- Direct first order with respect to the bacteria concentration plus an additional, indirect, more complex influence with respect to the same variable, through the term describing the radiation absorption by the bacteria.
- Of order 0.205 with respect to the local volumetric rate of photon absorption by the bacteria.

Fig. 6 shows typical experimental results and predictions from the model (solid lines) for the four levels of irradiation. It is important to realize that with more than one order of magnitude change in the irradiating conditions, the reaction rates are only slightly different. This lack of direct proportionality of the absorbed radiation (that is translated to the incident radiation at the reactor window) with the rate of inactivation has important economical consequences.

It should be noted that this is the first time that a kinetic model introduces the radiation absorption rate by the bacteria as the independent variable [Eq. (17)]. It has been usually reported that the inactivation rate is of first order with respect to the local value of the incident radiation [see Eqs. (11)–(14)]. Moreover, more crude approximations, through the proposal of a practical "dose" concept [44–46] sometimes relate the inactivation rate with a direct proportionality to the product of the radiation flux arriving at the reactor window or, with a somewhat better approach, the average of the radiation intensities existing in the reactor, times the corresponding average exposure time (in flow reactors, the ratio of the reactor volume to the flowrate). It must be noted, however, that more elaborate and precise models for dose calculations in flow reactors have been proposed [47,48] that are defined in terms of the actual, nonuniform local values of the radiation intensities existing in the reactor and their correspondence with the residence time distribution of the elements of the fluid inside the reactor considering that the exposure time cannot be described by a single value. However, in all these cases, even when more rigorous calculations are performed, the value that is always employed is the local radiation intensity and not the photon absorption rate by the organism. Moreover, the first order dependence with respect to the local intensity is never questioned.

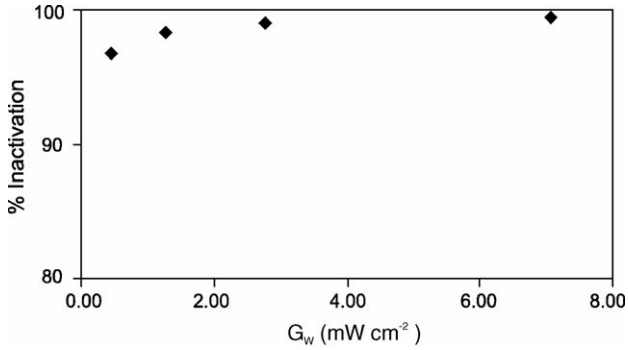

Fig. 7. Effect of the incident radiation at the reactor window on the bacteria inactivation. Reaction time: 300 s (effective exposure time: 21 s).

Results in the present model show that the inactivation rate has a very mild dependence with respect to the radiation absorption rate actually experimented by the microorganism (order 0.2). This extremely moderate effect is shown in Fig. 7 and suggests that the actual exposure time could be much more important than the level of the irradiation rate. The biological origin of this very distinct result is outside the scope of the present report, but is certainly related to very complex

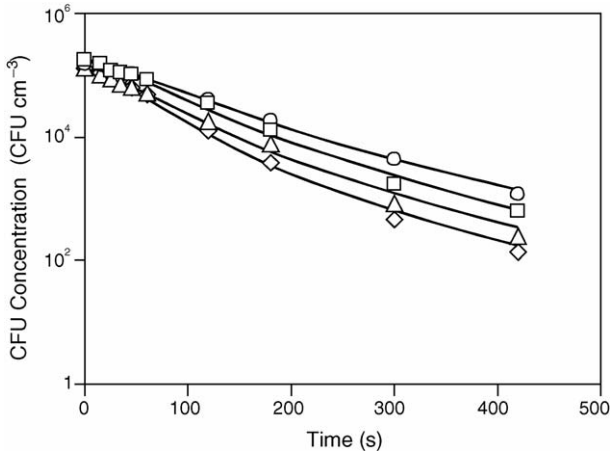

Fig. 6. Bacteria inactivation. Comparison of model predictions and experimental data. Solid lines: model predictions. (◯) Philips lamp with filter; (□) Heraeus lamp with filter; (△) Philips lamp; (◇) Heraeus lamp.

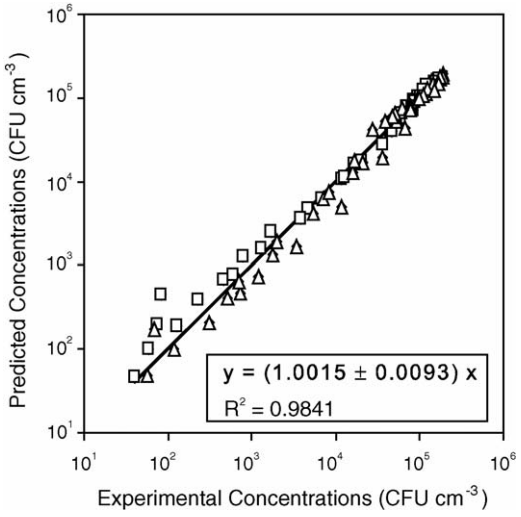

Fig. 8. Bacteria inactivation. Compendium of all experimental data comparing model predictions with experiments. (□) Bacteria grown with the glucose medium; (△) bacteria grown with nutritive broth.

elementary processes that take place in the interaction of the absorbed photons with the chemical constituents of the bacteria.

The threshold value equal to two indicates a very short plateau at the beginning of the reaction or, equivalently, a reduced resistance of the bacteria to become almost immediately inactivated.

Fig. 8 includes all runs and is a comparison of experimental data versus predictions from the model obtained using the calculated kinetic parameters. It can be seen that the model agrees very well the experimental results and seems to be a reliable representation of the inactivation process. According to the method employed to derive this result, this kinetic model should be apt for scaling-up purposes which is the ultimate objective of the present project.

Acknowledgments

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