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# An experimental investigation and modelling of fluid flow in solar photocatalytic reactor for contaminated water treatment

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## Abstract

Different types of solar photocatalytic reactors exists in the literature for water treatment, Thin-film fixed-bed reactor (TFFBR) is one of them which is used in this study because of its high surface area to volume ratio for water treatment. TFFBR is an inclined plate coated with P25 $TiO_2$ DEGUSSA over which contaminated water flows during treatment. The TFFBR also contains a pump, by which the water flow rate can be controlled. The main advantages of this TFFBR are its high efficiency, simple construction and low investment costs. Modelling of the fluid flow is important and crucial for the efficient design of the device.

A computational model of fluid flow in TFFBR to treat aquaculture pond water contaminated with an aquaculture organism *A. hydrophila* ATCC 35654 is presented in this paper. A pilot-scale thin-film-fixed-bed solar photocatalytic reactor is used for experimental measurement of disinfection of *A. hydrophila* ATCC 35654 as a function of flow rates and solar intensity. Modelling and simulation is done in MATLAB software for both aerobic and anaerobic conditions. Computational results are discussed, compared and verified against experimentally measured results. Furthermore, a fluid flow model of microbial disinfection against the cumulative energy of each flow rate is presented, discussed and recommended. Clearly, this study attempted to develop a fluid flow model and to optimize a pilot-scale TFFBR for the effective use of solar energy to treat water contaminated with pathogenic microbes.

## Introduction

Infectious diseases are the main constrain for operation and expansion of aquaculture industry. A wide range of pathogenic microbes are broadly responsible for this [1] and disinfection is an effective treatment to eliminate these viruses, bacteria, fungi and protozoan parasites [2]. However, to treat these pathogens, water disinfection is the scientific and technical challenge to overcome[3]. Chemical treatments (chlorination, ozone treatment or antibiotic application) are the most commonly used techniques for water disinfection [4]. Unfortunately all these generate toxic by-products which not only affect fish product but also cause health risks to human population [5, 6]. Therfore, the application of photocatalytic water disinfection process has gained significant attention due to its effectiveness in disinfecting organic contaminants in wastewater. Titanium dioxide ($TiO_2$) is one of the most widely used, stable and active photocatalysts that has established its application for water purification purposes by solar photocatalytic disinfection process.

Different types of solar photocatalytic reactors have been developed for water treatment for over the last 20 years [7]. They are parabolic trough reactor (PTR), Double skin sheet reactor (DSSR), Compound parabolic collecting (CPC) reactor and Thin-film fixed-bed reactor (TFFBR). TFFBR is a sloping plate coated with P25 $TiO_2$ DEGUSSA over which flows the contaminated water during use. The TFFBR also contains a pump, by which the water flow rate can be controlled. The main advantages of this TFFBR are (i) its high optical efficiency, (ii) its simple construction method and (iii) the low investment costs involved in development. Further advantages are that oxygen transfers effectively into the water film and there is no need for $TiO_2$ separation from the treated water.

In order to improve the water disinfection technology, many mathematical empirical models were developed in these years [8]. Mathematical models are normally used as tools to interpret the interactions between the set up of experimental designs and practical procedures [9]. Alvarez et al. stated that these models are an essential component of hazard analysis and critical control point systems or help the equipment manufacturers to predict the safety and shelf-life of foods at the design state [9]. Tallentire *et al*., (1971) and Dwyer et al., (1985) formulated a mathematical model which predicts the efficiency of radiation sterilised medical materials by calculating the probability of contamination occurrence [10, 11]. This model was evaluated with a range of radiation doses on microorganism to obtain the inactivation kinetics which was also considered in microbiological quality control testing. Most research published on mathematical models that were developed for microbial inactivation during disinfection was obtained from reactor where water is in static condition [26]. One recent report based on pilot plant system has been developed CFD (computational fluid dynamics) modelling for water disinfection through CPC pilot-plant reactor (Misstear and Gill, 2012). They simulated the percentage of tracked particles (a number of successful interactions between the bacterial cells and $TiO_2$ immobilised catalyst surface) that stroked each inserts (cylindrical, conical and consecutive frusta and a spring), at a fixed rate, as an indicator of efficiency of disinfection. However, no practical laboratory experiments were evaluated in this report to prove its efficiency practically.

The reaction on TFFBR reactor is different from CPC pilot plant and the system where aqueous flow is in static condition. Therefore, the kinetic model related to TFFBR system is expected to be more robust than other empirical models. Not many studies have dealt with kinetic models which are related to $TiO_2$ coated single-pass TFFBR reactor. BekbÖlet et al. investigated the percentage of degradation of landfill leachates' organic pollutants by using TFFBR under UV light with different pH values [12]. They used single pass experiments with TFFBR to study the effect of initial TOC (total organic carbon) concentration under different conditions. For Single pass experiment with TFFBR, they calculated the degradation rate of initial TOC by using the following mathematical equation.

$$\Delta n/\Delta t = (C^0 - C)*v = \Delta C*v \quad (1)$$

Where, C = initial substrate concentration (mole/l), $C^0$= substrate concentration (mole/l), $\Delta C$ = substrate concentration change (mole/l), v = Flow rate (L/h).

The present study focused on the development of a photocatalytic reactor for the disinfection of water contaminated with *A. hydrophila* ATCC 35654 with 5 different flow rates experiments, in aquaculture systems. The result reported here that solar photocatalysis can offer a functional means of inactivation of *A.hydrophila*, which proves the effectiveness of the application of solar photocatalysis in aquaculture systems.

**Methods**

Reactor

A pilot-scale thin-film fixed-bed reactor (TFFBR) system has been developed, based on two previous research studies [7, 12]. The overall experiment was set-up as a single-pass experiment and the reactor consisted of a water reservoir (representing an aquaculture pond in the model system), an air-controlled pump, a solar collector (glass plate) with immobilised photocatalyst, P25 $TiO_2$ Degussa and a collector vessel for the treated water (Fig 1). As in previous studies of chemical degradation [7, 12] and recent studies of microbial inactivation [3, 13], the reactor angle was maintained at $20^o$ throughout, and the light intensity was measured from the same angle as that of the reactor. The reactor angle was maintained at North facing throughout the experiments to get the best possible effect of natural sunlight in the southern hemisphere. The reactor was set to face north, to maximise direct sunlight. The illuminated surface area was 0.468 $m^2$ with 1.17m in depth and 0.4 m in width; the irradiated volume was 200 mL in 2.5 min (irradiance time). The thinness of the film of water across the photoreactor plate is at <0.3 mm. The density of the $TiO_2$ photocatalyst 20.50 g $m^{-2}$ and the photocatalyst layer was not covered during the experiments.

The $TiO_2$ P25 Degussa photocatalyst was coated on four pieces of 3.3 mm thick Borofloat 33 glass plates (Schott, Australia). Plates were degreased using a reagent grade Piranha solution (3:1 sulphuric acid and 30% hydrogen peroxide). Then slurry of $TiO_2$ was prepared with methanol and the glass was coated by spraying. Then it was baked at 450°C for an hour or two to anneal the $TiO_2$ to the glass.

Source of water

Experiments were performed by using natural spring water (Satur8 Pty, Ltd, Australia).

Bacterial Culture and lab enumeration

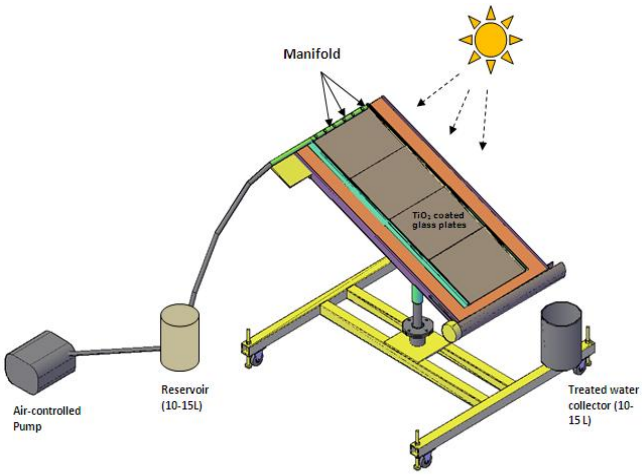
*Aeromonas hydrophila* ATCC 35654 was purchased from Oxoid. Bacterial culture maintenance, preparation and lab enumeration was detailed in Khan et al. [14]. Aerobic counts were enumerated to get healthy cell counts and ROS-neutralised counts were enumerated for healthy and injured cell together where reactive oxygen species were neutralised by adding peroxide scavenger into the growth medum. When $TiO_2$ (powder or film form) is in an aqueous medium and irradiated with near UV $\lambda < 385$ nm, $^{\bullet}OH$ radicals are generates which are highly toxic or microorganisms and lead to cell death. High level of produced ROS has been described as lethal for cell integrity. These cells cannot grow properly under aerobic condition. So, ROS-neutralised condition is required to get total inactivation.

Experimental conditions

This study considered five flow rate conditions (1.8 L $h^{-1}$, 4.8 L $h^{-1}$, 8.4 L $h^{-1}$, 16.8 L $h^{-1}$ and 32.4 L $h^{-1}$) and high sunlight conditions 1000-1100 W $m^{-2}$, as experimental conditions. Experiments were performed in a day with above experimental conditions and repeated in 2 different days. Therefore, for each flow rate, there were 3 identical sets of samples collected for further lab enumeration. Overall six counts against each flow rate were obtained and the average was considered.

Modelling and programming

For the processing of the experimental data and the prediction of observed inactivation curves, Matlab$^R$ (The MathWorks Inc., Natick, USA) programming was used in this study. The BekbÖlet et al., 1996 [12] model was slightly modified for better use of the equation according to the experimental results and it was implemented to compare the efficiency rate which is given below.




(b)

**Figure 1: (a) schematic diagram and (b) photograph of the thin-film fixed-bed reactor (TFFBR) used in this study.**

Microbial death (inactivation) rate (d) can be defined by,

$$d = (C\text{-}C^0) \times v \times RT \quad (2)$$

Where, C = initial cell concentration (CFU/ mL), $C^0$ = cell concentration after treatment (CFU/mL), v = Flow rate (L/h) and RT= measured residence time per flow rate.

Residence time is the proportion of the total volume of water sample on the reactor plate against per flow rate. In a single-pass continuous flow system, the residence time (sec) is calculated by,

$$RT=Q/v \quad (3)$$

Where, RT = residence time, Q is the total volume of water sample on the reactor (mL), v=Flow rate $Lh^{-1}$. In this study, both Q and v are obtained by experimental measurement.

Cumulative energy value is used to estimate the accumulated solar energy in the photoreactor per unit of volume of treated water for a certain period of time during the experiment [15]. Cumulate energy (kJ) was calculated by,

$$CE = [(SI \times RT)/1000] \times 0.468 \quad (4)$$

Where, CE= cumulative energy (kJ), SI= Average Sunlight intensity ($W\ m^{-2}$), RT= residence time in sec, and 0.468 $m^2$is the illuminated surface area of the reactor (1.17m in depth and 0.4m in width).

**Analytical method and experimental error analysis**

In this study, the least square method was used to fitting all the experimental results. The best fit of this method would minimize the sum of squared residuals, which is given by Equation [16]

$$SS(\exp) = \sum_{i=1}^{n} (\delta N_{i,obs})^2 \quad (5)$$

Where $\delta N_{i,obs}$ is the estimate of the uncertainties in $N_{i,obs}$. The sum squares of the experimental error depend on the number of observed points. The error at each measurement point was calculated using the rules for combining errors. If the independent variables given by $X = \{x_1, x_2, \ldots, x_n\}$ are combined to give Y by the relation,

$$Y = y(x_1, x_2, \ldots, x_n) \quad (6)$$

Then the error combining rule is given by,

$$\Delta y^2 = \sum_{i=1}^{n} \left( \frac{\delta y}{\delta x_i} \Delta x_i \right)^2 \quad (7)$$

All the modelling, calculations, and data processing are programmed by MATLAB. The polyfit method in MATLAB is used to find the coefficients of a polynomial of degree n (in this study, N=1 or 2) that fits the data Y best in a least-squares sense. A vector of length n+1 containing the polynomial coefficients in descending powers is obtained by this method.

**Results and discussion**

Flow rate vs Microbial inactivation

Figure 2 shows the inactivation counts for *A.hydrophila* ATCC 35654 with error bars in sterile spring water run through the TFFBR at 5 different flow rates (1.8 L $h^{-1}$, 4.8 L $h^{-1}$, 8.4 L $h^{-1}$ 16.8 L $h^{-1}$ and 32.4 L $h^{-1}$) under high total sunlight conditions. Thus, each experiment provides two sets of inactivation data, (i) an aerobic result with healthy cells only and (ii) a ROS-neutralised result with healthy and injured cells together. The lowest flow rate (1.8 L $h^{-1}$) showed higher inactivation of approximately, $1.28 \times 10^5$ CFU $mL^{-1}$ as expected, where the initial count was $1.3 \times 10^5$ CFU $mL^{-1}$ and a final count was approximately, $3.9 \times 10^3$ CFU $mL^{-1}$ under both aerobic and ROS-neutralised condition (Table 2). Similarly, at 16.8 L $h^{-1}$ the inactivation was $1.1 \times 10^5$ CFU $mL^{-1}$ which was less than that of at 1.8 L $h^{-1}$, 4.8 L $h^{-1}$, 8.4 L $h^{-1}$ under both growth conditions. The highest flow rate (32.4 L $h^{-1}$) showed the lowest inactivation count of $1.5 \times 10^4$ CFU $mL^{-1}$, with similar number of initial count and a final count of approximately, 1. 3 x $10^5$ CFU $mL^{-1}$ under both aerobic and ROS-neutralised condition (Table 2). Therefore, under high sunlight conditions, there was minimal cell injury observed under ROS-neutralised condition.

The relationship between inactivation count (CFU/mL) and flow rate (L/h) is analysed by bi-quadratic curve according to the principle equation given below:

$$IC = a"v^2 + b"v + c" \quad (8)$$

Where, IC=inactivation number, $a"$, $b"$ and $c"$ are the coefficients of bi-quadratic curve.

Therefore, a non-linear regression analysis was conducted by fitting bi-quadratic curve to the experimental data using "MATLAB CODE". The best fit bi-quadratic curve equations for aerobic and ROS-neutralised counts are given in Table 1. Both equations derived the data from aerobic and ROS-neutralised condition counts gave $R^2$ value of 0.9936 and 0.9960, respectively, at 95% confidence limit. As the $R^2$ value was close to +1, the hypothesis "the inactivation of *A.hydrophila* increases when the flow rate decreases" - is significant. Overall, there was a major effect on *A.hydrophila* inactivation, irrespective of whether the TFFBR sample was counted under aerobic or ROS-neutralised conditions.

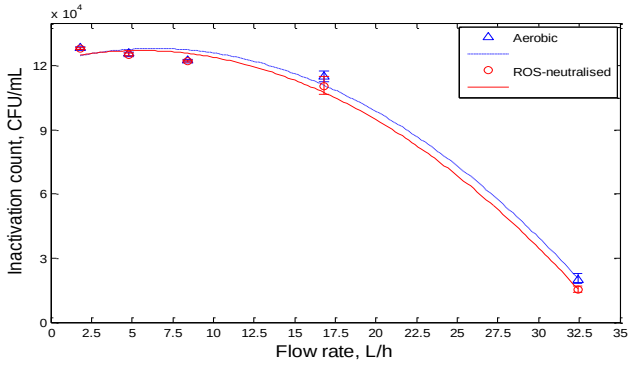



**Figure 2: Effect of TFFBR on inactivation of *A. hydrophila* (ATCC 35654) under high sunlight condition (1000-1100) W $m^{-2}$ at 5 different flow rate condition (1.8 L $h^{-1}$, 4.8 L $h^{-1}$, 8.4 L $h^{-1}$ 16.8 L $h^{-1}$ and 32.4 L $h^{-1}$).** Enumeration was aimed at under standard aerobic condition (blue dash line) and under ROS-neutralised condition (red line). Error bar represents 95% confidence limits (n=6).

| Enumeration condition | Bi-quadratic curve equation | $R^2$ values |
|---|---|---|
| Aerobic | Y= -158.52x2+2023.03x+121645.8 | 0.9936 |
| ROS-neutralised | Y= -153.67x2+1684.6x+122541.88 | 0.9966 |

**Table 1: Bi-quadratic curve equations for aerobic and ROS-neutralised counts calculated against five flow rates (1.8 L $h^{-1}$, 4.8 L $h^{-1}$, 8.4 L $h^{-1}$, 16.8 L $h^{-1}$ and 32.4 L $h^{-1}$).**

Figure 3 demonstrates the death rate of *A. hydrophila* against different flow rates by calculating through Bekbolet et al. [12] model modified equation 2 which shows an exponential decrease of microbial inactivation with the increasing flow rates conditions. Table 2 shows the calculated death rate (d) of *A. hydrophila* counted under ROS-neutralised condition using Bekbolet et al. [12] model modified equation 2. Marugan et al. reported that photocatalytic oxidation of methylene blue cannot always be extrapolated to microbial photocatalytic disinfection [17]. They established that microbial disinfection was sensitive towards water compositions and different types of immobilised $TiO_2$ reactors provided evidence of opposite degradation behaviours of methylene blue. This variation could be due to difference in chemical and microbial oxidation system. Van Grieken et al. used a fixed bed reactor to compare the disinfection pattern of *E. coli* with methyl blue degradation [18]. In their experiment, $TiO_2$ was immobilised onto glass Raschig rings into an annular reactor. Their finding was that methylene blue degradation was not correlated with the activity for *E. coli*

inactivation. They suggested that as the *E. coli* size was several magnitudes larger than the $TiO_2$ particle, $TiO_2$ diffused inside the microbial porous structure and the contact between them was then only restricted to the external $TiO_2$ surface. In contrast, Lim et al. used a different (honey comb shaped, immobilised $TiO_2$) photocatalytic reactor for ground water applications [19]. They used methylene blue to characterise the performance of the reactor by removing it at single pass experiment under different flow rates. They showed that methylene blue removal efficiency decreased linearly with increasing flow rates. Similarly, Sordo et al showed similar pattern between the methylene blue decolourisation and *E.coli* photocatalytic disinfection [13]. However, the Figure 2 (experimental measurement graph) and the Figure 3 (death rate-Bekbolet modified model equation graph) clearly shows similar pattern of decrease in microbial inactivation as shown in for TOC degradation by using similar TFFBR in a single-pass, under high sunlight condition.

TFFBR was comparatively less evidenced to for observing microbial disinfection. Therefore, the current study is the first study to investigate the efficiency of TFFBR by comparing that with [12] theory through disinfecting *A. hydrophila*. It showed that *A. hydrophila* inactivation followed similar trend of chemical degradation, with this single pass TFFBR reactor.

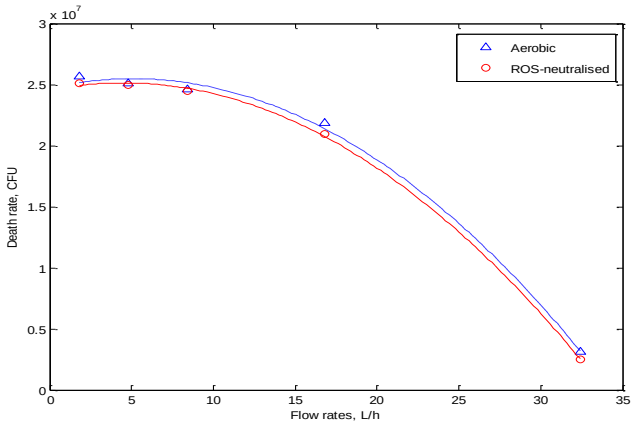



**Figure 3**: **Death rate of *A. hydrophila* (ATCC 35654) against five different flow rates, 1.8 L $h^{-1}$, 4.8 L $h^{-1}$, 8.4 L $h^{-1}$ 16.8 L $h^{-1}$ and 32.4 L $h^{-1}$.** Enumeration was aimed at under standard aerobic condition (red line) and under ROS-neutralised condition (blue dash line).

| Flow rates ($Lh^{-1}$) | Avg Initial counts (CFU $mL^{-1}$) | Avg After treatment counts (CFU $mL^{-1}$) | Avg Inactivation (CFU $mL^{-1}$) | RT (Sec) | Death rate (d) | CE (KJ) |
|---|---|---|---|---|---|---|
| 1.8 | $1.3 \times 10^5$ | $3.9 \times 10^3$ | $1.28 \times 10^5$ | 400 | $2.5 \times 10^7$ | 187.2 |
| 4.8 | $1.3 \times 10^5$ | $6.9 \times 10^3$ | $1.24 \times 10^5$ | 150 | $2.4 \times 10^7$ | 70.2 |
| 8.4 | $1.3 \times 10^5$ | $9.9 \times 10^3$ | $1.21 \times 10^5$ | 86 | $2.3 \times 10^7$ | 40.25 |
| 16.8 | $1.3 \times 10^5$ | $2.2 \times 10^4$ | $1.10 \times 10^5$ | 41 | $2\ 0 \times 10^7$ | 19.19 |
| 32.4 | $1.3 \times 10^5$ | $1.1 \times 10^5$ | $1.5 \times 10^4$ | 18 | $3.2 \times 10^6$ | 8.42 |

-To calculate death rate (d) at 1.8 L $h^{-1}$, Flow rates was considered in mL $h^{-1}$ as 1.8 L $h^{-1}$ = 1800 mL $h^{-1}$ and RT was considered in h as 400 sec = 400/3600 h. Similar calculation was considered for each flow rate experiments.
-As all the experiments were performed under high sunlight condition (≥ 1000 W $m^{-2}$), for better cumulative energy calculation for every experiment the sunlight intensity was considered as 1000 W $m^{-2}$.
-RT =Residence time and CE = cumulative energy

**Table 2: death rate and inactivation of *A. hydrophila* under ROS-neutralised condition against different flow rate condition and different cumulative energy (KJ).**

Cumulative energy vs Microbial inactivation

As detailed in the introduction that injured cells can only be cultured and counted under conditions where reactive oxygen species are neutralised. Therefore, ROS-neutralised condition provided both healthy and injured cells with a possible complete level of microbial inactivation. Hence, here only inactivation of *A. hydrophila* under only ROS-neutralised condition is considered.

In order to evaluate the influence of high sunlight intensities and the effect of flow rates, Cumulative energy (kJ) on the reactor plate was calculated and tabulated in Table 2. Table 2 clearly shows that the measured residence time (sec) was different for each flow rate conditions. As all the experiments were performed under high sunlight condition (≥ 1000 W $m^{-2}$), here for calculation the sunlight intensity was considered as 1000 W $m^{-2}$. The lowest flow rate of 1.8 L $h^{-1}$ was found to be the most effective for inactivation of *A. hydrophila* ATCC 35654 because of highest residence time of 400 sec compared to the other flow rates. The residence time was 150 sec at 4.8 L $h^{-2}$ experiment. When the total sunlight intensity was at 1000 W $m^{-2}$, the cumulative energy was found to be 187 kJ whereas; the cumulative energy was 70 kJ at 4.8 L $h^{-1}$ which plays a major role for *A. hydrophila* inactivation. Similarly, at 16.8 L $h^{-1}$ the cumulative energy was 19 kJ whereas at 32.4 L $h^{-1}$ it was only 8kJ. Therefore, the highest cumulative energy at 1.8 L $h^{-1}$ revealed the highest inactivation ($1.28 \times 10^5$ CFU $mL^{-1}$) compared to the other flow rate experiments. Therefore, a non-linear regression analysis was conducted by plotting the average ROS-neutralised inactivation counts for *A.hydrophila* against cumulative energy/flow rate to express a bi-quadratic equation (Figure 4).

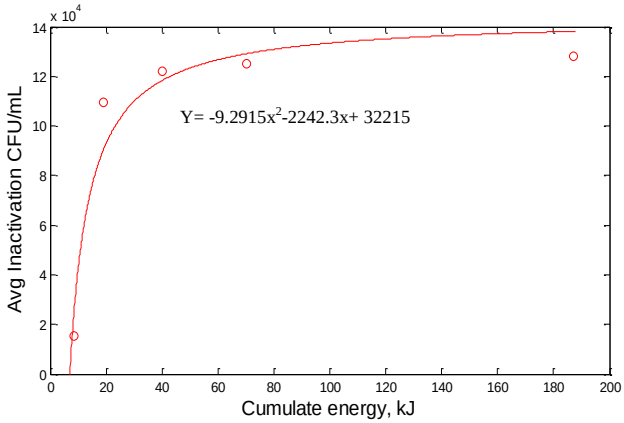



**Figure 4: Effect of $TiO_2$ photocatalyst on *A. hydrophila* (ATCC 35654) inactivation against different cumulative energy under high sunlight condition (1000-1100) W $m^{-2}$ at 5 different flow rates (1.8 L $h^{-1}$, 4.8 L $h^{-1}$, 8.4 L $h^{-1}$ 16.8 L $h^{-1}$ and 32.4 L $h^{-1}$).** Enumeration was aimed at under ROS-neutralised condition.

## Conclusions

This study investigated the efficiency of TFFBR disinfecting *A. hydrophila* using TFFBR. It showed for the first time that inactivation of *A. hydrophila* followed the similar pattern of chemical degradation, with this single pass TFFBR reactor. This study has developed an empirical model equation for microbial inactivation against the flow rate and cumulative energy calculated on the plate whether the experiments were performed either with 5 flow rates or with single flow rate.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