

ภาคผนวก ฉ
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BACTERIA DISINFECTION BY SOLAR ENERGY AND PHOTOCATALYSIS

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ABSTRACT

The comparative experimental study on disinfection of *Escherichia coli* (E.coli) in water using solarization in associated with a compound parabolic collector (CPC) and TiO₂-based photocatalysis process with and without light were performed. A set of experimental work was carried out to examine the effect of solar intensity on the inactivation performance. As a result, E.coli population declined faster and to greater extent under solarized photocatalytic process than in the solarization or in TiO₂ mixture only. However, E.coli has been decreased by photocatalysis disinfection up to 2-log unit in 60 min at a low-intensity radiation. In high-intensity irradiant environment, the photocatalysis deactivated bacterial up to 5-log unit of concentration in 60 min whereas the best for solar disinfection was 3-log in 60 min. Moreover, the experiment with catalyst in the dark condition could decrease E. coli up to only 2-log

INTRODUCTION

There have been numerous research works on renewable energy for sustaining our future energy including solar, wind, biomass and geothermal. Among those, solar energy has gained much attention as it can be utilized for many applications at no cost of fuel. Furthermore, solar energy can be applied for bacteria disinfection via the processes called "Solarization" and "Photocatalysis" [1]. Photocatalysis for bacterial inactivation in drinking water using commercial Titanium-dioxide (TiO₂) has been evaluated by means of solar simulator and the effectiveness of solar disinfection against E. coli compared to solar photocatalysis disinfection was reported [2]. The recent studies on water disinfection by photocatalysis mostly focused on using TiO₂ and solar radiation and examining key parameters that effect system performances [3-4]. Moreover, a compound parabolic concentrator has been applied for water disinfection with solar radiation as well as various sources of light showing a threshold of catalyst concentration on disinfection efficacy [5]. The efficiency of photocatalysis to inactivate bacterial is derived by light intensity, light intermittent, water temperature, catalyst concentration and solar radiation [6].

This work aimed to evaluate the effectiveness of the simple photocatalysis disinfection system using suspended TiO₂ in E. coli contaminated water compared with a typical solar disinfection method-solarization and non solar exposure one. To further study, the effect of solar intensity on the bacteria inactivation was also analyzed.

MATERIALS AND METHODS

Reactor setup

In this study, Pyrex test tubes were adopted as a reactor. To enhance the solar radiation intensity on the reactors, non-tracking Compound Parabolic Concentrator (CPC), which was originally developed for soil solarization application, has been employed for wastewater disinfection purpose during this research. The CPC has a flat receiver as shown in Fig. 1 for which all parameters can be calculated according to the following equations [7] where H is the height of CPC, a is the receiver, θ_c is the Acceptance half-angle, f is the focus of parabola

$$H = \frac{a}{2} \left(\frac{1}{\tan \theta_c} + \frac{1}{\tan \theta_c \sin \theta_c} \right) \quad (1)$$

$$f = \frac{a}{2} (1 + \sin \theta_c) \quad (2)$$

$$x^2 = 4fy$$



Fig. 1 Flat-Plate Compound Parabolic Concentrator.

The flat-plate compound parabolic collector (CPC) was designed to focus solar radiation beam onto the reactors mounted on platform tiled 14° local latitude and held by aluminum frames. The pyranometer was used to measure solar radiation for analyzing influence of intensity level of solar radiation on solarized and photocatalysis disinfection systems. Titanium-dioxide was used as a photocatalyst. The experiments were carried out in three conditions: (1) solar disinfection without catalyst, (2) photocatalysis disinfection with suspended catalyst in tubes and (3) the dark condition in which there was suspended catalyst in tube but covered with a black plastic. For one batch of the experiment, it

consisted of 4 tubes for solar disinfection, 4 tubes for photocatalysis disinfection and 4 tubes for the dark condition totaling 12 tubes on the CPC. All tubes were experimented as the same time. All the experiments were carried out at King Mongkut's University of Technology Thonburi, Bangkok (KMUTT) (13° 45' N and 100° 31' E).

Bacteria strain

Escherichia coli were used as contaminating bacteria in household wastewater. *E. coli* K-12 was inoculated at 37 °C with constant revolution at 100 rpm for 24 hr before each experiment. All experiments used initial bacterial concentration of 4.7×10^7 CFU/mL and inoculated *E. coli* in tubes directly. The process has been done in the Biochemical Laboratory at School of Bioresources and Technology, KMUTT.

Catalyst preparation

In this study, UNILAB Titanium dioxide (TiO₂) by AjexFinechem is used as a photocatalyst by dispersed into the wastewater samples at a concentration of 100 mg/L for each sample. All catalyst and test tubes were sterile in autoclave every time before experiments.

Experimental parameters

In the experiments, the parameters that may have effect on water disinfection such as high-intensity radiation, low-intensity radiation and catalyst in the dark were evaluated

Methodology of experiments

For all experiments, the *E. coli* suspension were prepared in tube with an initial concentration as 4.7×10^7 CFU/mL. TiO₂ was then dispersed in the reactor at 100 mg/L dose for photocatalysis for all 4 tubes and sampling every 15 min for high-intensity radiation and 30 min for low-intensity radiation.

All experiments were performed in exactly the same photoreactor for all 3 conditions: photocatalysis disinfection, solar disinfection and the dark condition with the catalyst. The photocatalysis disinfection was carried out in the presence of sunlight with the catalyst of 100 mg/L while solar disinfection experiment was the same as photocatalysis disinfection but no catalyst in tube and in the dark experiment, tube with suspended TiO₂ of 100 mg/L and covered by black plastic to conceal solar radiation.

RESULT AND DISCUSSION

In Fig.(2), at low intensity of radiation with the average solar irradiance of 73.33 W/m², there is no significant decrease in *E. coli* concentration in both the solarization without catalyst and in the dark condition while photocatalysis disinfection could inactive bacterial concentration in 2-log unit in 90 min. This can be explained that at low solar radiation the water temperature is not high enough to harmful the bacteria. Similarly, without the light, TiO₂ cannot react to produce OH to deactivate the bacteria as well. Regardless to low intensity the photocatalytic process is capable to generate OH for bacteria deactivation.

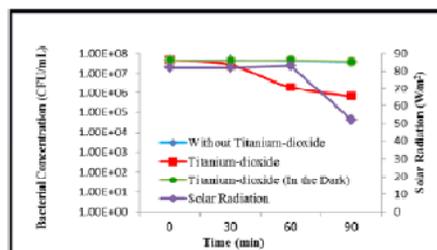


Fig. 2 Water disinfection by solar radiation and photocatalysis with low-intensity radiation.

For the high intensity of radiation case shown in Fig. (3), the solar radiation was measured at 892 W/m². Bacterial concentration was decreased 2-log by catalyst in the dark condition. Moreover, solar disinfection decreased bacterial concentration almost 3-log while 5-log decreasing in bacterial concentration was observed in photocatalysis disinfection experiment. Moreover, both curves show the effect for water disinfection from TiO₂ and solar radiation. The experimental result demonstrates that at the high radiation intensity the photocatalysis disinfection is more effective than solar disinfection.

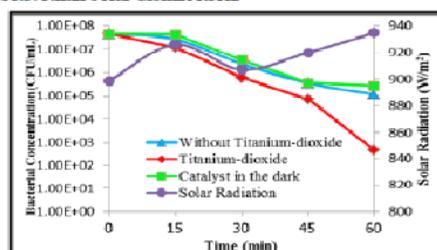


Fig. 3 Water disinfection by solar energy and photocatalysis with high-intensity radiation

Comparison of the effect of solar intensity on the solar disinfection is shown in Fig.4. At the low intensity, bacterial disinfection is shown in Fig.4. At the low intensity, bacterial concentration is still the same as initial concentration while decreasing up to 3-log at high-intensity radiation. This can be concluded that solar disinfection is more affected by the increase in the light intensity.

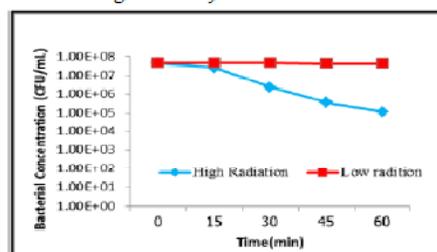


Fig. 4 Water disinfection by solar energy

To further investigate the effect of light intensity, results of photocatalysis disinfection with high-intensity radiation and low-intensity radiation are now compared in Fig. 5. As a result, bacterial concentration in case of TiO₂ exposure to high irradiance decreases up to 5-log whereas the one under low-intensity reaches at 2-log.

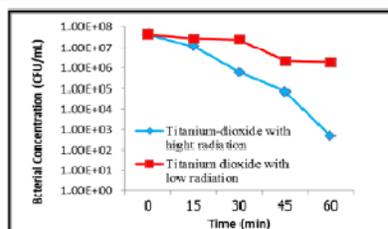


Fig. 5 Water disinfection by photocatalysis

Duffie, J.A., *Solar Engineering of Thermal Processes*, 2nd edition, John Wiley & Sons Inc, New York.(1991)

CONCLUSION

The compound parabolic concentrator has been demonstrated to improve water disinfection efficacy over solar disinfection and photocatalysis disinfection with TiO_2 during 0-60 min.

For low-intensity radiation photocatalysis could fairly inactivate E.coli whereas catalyst-only in the absence of light could not decrease bacterial concentration. Moreover, Bacterial concentration has not been decreased by solar disinfection with low-intensity radiation and catalyst has been proven to affect bacterial concentration for low-intensity radiation.

For high-intensity radiation, solar disinfection and catalyst-only in the dark condition has significant in decreasing bacterial concentration and hence the intensity of solar radiation affects the disinfection. However, photocatalysis disinfection is more effective in bacterial inactivation than solar disinfection and the catalyst-only in absence of light.

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THE EFFECT OF BACTERIAL DISINFECTION BY SOLAR ILLUMINATION AND PHOTOCATALYTIC DISINFECTION

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ABSTRACT

The disinfection of *Escherichia coli* (*E.coli*) in water using solar and photocatalytic processes associated with a compound parabolic concentrator (CPC) have been studied. Moreover, Titanium-dioxide (TiO_2) has been applied to the solar photocatalysis process. The set of experiment is conducted to examine the effects of solar intensity, illuminated area and water temperature on the bacteria inactivation rate. As a result, bacteria concentration declined after 90 min of illuminated at water temperature between 37-44 °C for all cases and no effect on bacteria inactivation is observed. Moreover, for all cases of solar disinfection, *E.coli* concentration decreases to 2 logs unit in 90 min whereas the bacteria concentration decreases up to 4 logs unit for the same durations in the photocatalytic disinfection.

INTRODUCTION

In recent years, there have been numerous researches on renewable energy. Among those,

solar energy has been utilized for many applications as it has no cost of fuel. Specifically, in many studies, solar energy has been applied for bacteria disinfection, so called "solarization" and "photocatalytic disinfection". Drinking water has been found to be a cause of diarrhea and it becomes a serious problem. Therefore, solar energy has been introduced to bacteria disinfection process [1]. Moreover, TiO_2 , as a photocatalyst, has also been applied to such a process to enhance the bacteria disinfection efficiency of [2, 3], by means of hydroxyl radical ($\bullet\text{OH}$) produced by UV radiation in a wavelength of nearly 390 nm. [4]

The compound parabolic concentrator has been adopted to increase solar intensity on reactor surfaces for solar water disinfection and photocatalytic disinfection [5]. Moreover, a combination of CPC and TiO_2 has been applied to enhance the solar photocatalysis process for bacteria inactivation [6]. The effective of solar disinfection and photocatalytic disinfection with



TiO₂ was also evaluated using *E. coli* as an indicator for bacteria inactivation. The effecting parameters including the effect of irradiated surface, water temperature, cumulative solar energy and concentration of TiO₂ were analyzed.

THE COMPOUND PARABOLIC CONCENTRATION SET UP

The compound parabolic concentrator (CPC)

$$H = \frac{a}{2} \left(\frac{1}{\tan \theta_c} + \frac{1}{\tan \theta_c \sin \theta_c} \right)$$

$$f = \frac{a}{2} (1 + \sin \theta_c)$$

$$x^2 = 4fy$$

made by stainless plate to reflect global solar irradiation, which is used to enhance the intensity of global solar irradiation on the reactor surfaces. The CPC has been designed to have a flat receiver as shown in Fig. 1. The parameters of CPC can be calculated using the following equation [7].

where H is the height of CPC. a is the receiver, θ_c is the acceptance half-angle and f is the focus of parabola.

With the equations [1-3], the calculation gives that the height of CPC is 1.234 m, the aperture area of CPC is 0.696 m and the focus is 0.092 m. The θ_c is set at 21° and the flat receiver is 12.5 cm.



Fig.1 Compound parabolic concentrator.

The compound parabolic concentrator (CPC) is set up to focus light intensity of solar irradiation onto reactor surfaces mounted on a platform tiled 14° facing south. This angle proved that the best angle to receive solar radiation in Thailand. Moreover, a pyranometer has been used to measure solar radiation to analyze water disinfection and photocatalytic disinfection performances.

THE EXPERIMENT SET UP

The schematic diagram of the experiment set-up including reactors, compound parabolic concentrator, reservoir, sampling valve, regulator valve, circulating pump and flow meter is shown in Fig. 2. Four reactors, 7cmX5cmX20cm each, connected in series are located on the CPC. The water flows directly through a sampling valve and a regulator valve by a circulating pump and then through the reactors that are connected to a reservoir and directly returns to reservoir again. All the experiments are carried out at King Mongkut's University of Technology Thonburi, Bangkok (13° 45' N 100° 31' E)

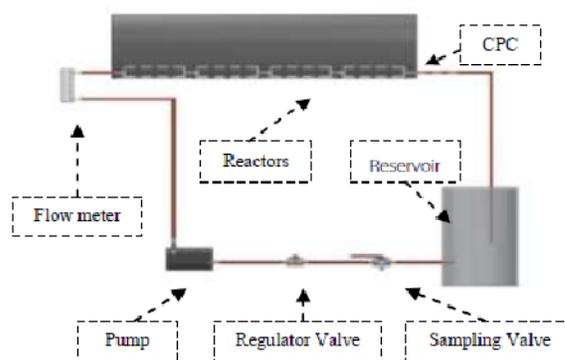


Fig. 2 Diagram of experiment



EXPERIMENT PROCEDURE

The bacteria inactivation effect is determined by measuring *E.coli* concentration under natural sunlight on the CPC for every 15 min. The suspension of bacteria culture about 400 mL is added into the reservoir with 6 L of de-ionized water inside and the initial reaction cell counts 10^7 CFU/mL. The operation is designed to quantify bacteria concentration against the time for 90 min. This experiment has been tested in August.

BACTERIA CONCENTRAION ANALYSIS

Escherichia coli (*E.coli*) are used as a contaminant in de-ionized water. Moreover, bacteria cells are diluted and plated on LB-agar. The plates are incubated at 37 °C and a colony count is then taken after 24 hr. After that, the colonies are counted and the initial of bacteria cell are calculated. All experiments have been performed in the Biochemical Laboratory at School of Bioresources and Technology, KMUTT.

RESULTS AND DISCUSSION

The experimental results reported in this section are divided into four parts. First is to evaluating the effect of global solar intensities on the bacteria disinfection processes in the experiments with four different reactor configurations (1, 2, 3 and 4 reactors per set).

With the same configuration, then, the effect of cumulative solar energy is analyzed. Following is the effect of water temperature. Finally, the performances of the only solar disinfection process and photocatalyst disinfection process are compared.

1. THE EFFECT OF SOLAR IRRADITION ON BACTERIA CONCENTRATION.

Fig (3) shows the normalized bacteria concentration under solar irradiation with

different illuminated areas as a function of time. The effect of solar irradiation to bacteria disinfection with different amount photoreactors is then analyzed.

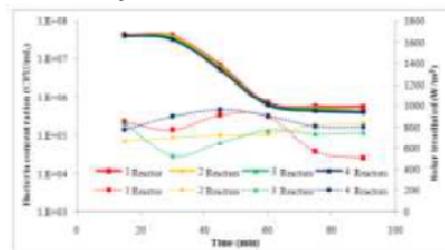


Fig. 3. The effect solar irradiance (W/m^2) against time on bacteria concentration (log (CFU/mL))

As seen, the photoreactor surfaces are illuminated by instable solar irradiation. However, all four cases of experiments show that bacteria concentration is decreased almost the same. It can be observed that, high light intensities of solar irradiation have no effect on bacteria inactivation. Moreover, the results have demonstrated that the more reactors, the lower efficiency. This is consistent with the work reported in the literature [2]. On the other hand, the low light intensities of solar radiation have an effect on bacteria disinfection. This also agrees with the previous research result shown that the bacteria disinfection is no more affected when increasing reactor surfaces [8]. With the result, it can be further applied to the system design to save cost of reactor used in the system.

2. THE EFFECT OF CUMULATIVE SOLAR ENERGY ON BACTERIA INACTIVATION

To further investigate the cause of bacterial inactivation, the cumulative solar energy is plotted against time with bacteria concentration for four reactor surfaces as depicted in Fig. 4.

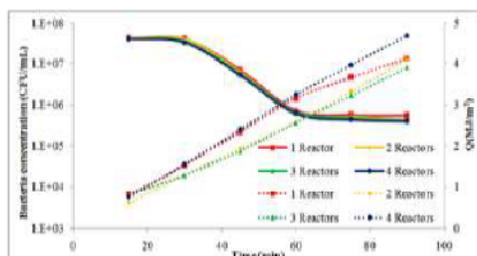


Fig. 4. The effect of cumulative solar energy (MJ/m^2) against experimental time on bacteria concentration ($\log(\text{CFU}/\text{mL})$)

There is no different in the decrease of bacteria concentration even though the cumulative solar energy is increased with the reactor surfaces. In fact, it is clearly seen that the more reactor area, the more absorbed photon from solar irradiation. After 90 min of illuminated time, the cumulative solar energy on the reactor with maximum surface area (4 reactors connected in series) is about $47 \text{ MJ}/\text{m}^2$ and that on the minimum surface reactor (one reactor) is about $4.0 \text{ MJ}/\text{m}^2$. However, there is no significantly different of bacteria inactivation rates. Therefore, the largest irradiated area reactor cannot decrease bacterial concentration better than the smaller area and the smallest irradiation area shows the highest efficiency for bacteria disinfection. It can be noted that an amount of energy is not a key to success in the bacteria inactivation process. This may deduce that the time of contaminated water exposure to sunlight has less significant in the bacteria disinfection process as well.

3. THE EFFECT OF WATER TEMPERATURE ON BACTERIA CONCENTRATION

In this step, the water temperature in each condition of experiments is measured to examine the effect of water temperature on bacteria concentration.

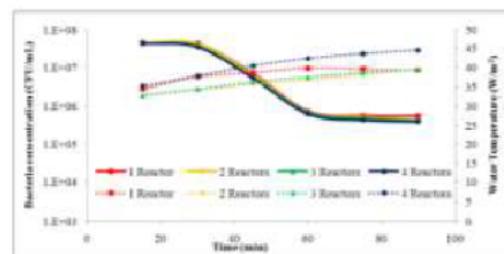


Fig.5 The effect of water temperature ($^{\circ}\text{C}$) against experimental time on bacteria concentration ($\log(\text{CFU}/\text{mL})$)

Fig. 5 shows the water temperatures in the test reactors. For the 4-reactor set, the temperature has increased up to 44°C because it has the largest area to absorb the incident solar irradiation. Moreover, the water temperatures of the other reactors are in a range of $32^{\circ}\text{C} - 40^{\circ}\text{C}$. However, none of them shows a significant different result in a decrease of bacteria concentration. It is noted that, the water temperature is not over 45°C may not has major influence to bacteria disinfection.

4. PERFORMANCE COMPARISON OF SOLAR DISINFECTION WITH AND WITHOUT TiO_2

To examine the effect of TiO_2 on bacterial inactivation process, a $1 \text{ g}/\text{L}$ TiO_2 is added into reservoir to catalyze the solar photocatalysis process. In this experiment, only a four-reactor set is employed. The result is shown in Fig. 6.

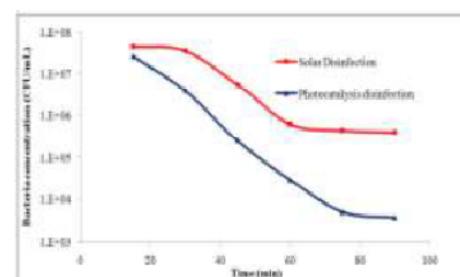


Fig. 6. The effect of solar disinfection and photocatalytic disinfection ($\log(\text{CFU}/\text{mL})$) against experimental time (min).



In Fig. 6, the photocatalytic disinfection process with suspended TiO_2 has more effect to bacterial inactivation than the only solar. Moreover, the photocatalytic disinfection could inactive bacteria concentration in 4 logs in 90 min while the solar disinfection could do only 2 logs in the same duration. The result can be explained that TiO_2 could react with the photons to product OH^\bullet radicals which enhance the deactivation process so that bacteria concentration decrease rate is higher than the only solar disinfection process. Moreover, the experimental result has demonstrated that the photocatalysis process is more effective than the only solar disinfection process.

CONCLUSION

This work has demonstrated that the solar intensities, energy and the temperature of contaminated water with different amount of reactor have no significant effect on bacteria disinfection. This could be attributed to the flow of photons is not high enough to attack to bacteria cells. However, the photocatalytic disinfection is proven to have higher efficiency than the only solar disinfection process due to the OH^\bullet radical generated by TiO_2 at the certain wavelength of solar irradiation to attack the bacteria cells. Moreover, the water temperature is increased by solar light intensities not over 45 °C for all cases. The experiments reveal that the temperature less than 45 °C is not influent to bacteria inactivation in both solar energy and photocatalytic disinfection processes.

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Disinfection of water containing *Escherichia coli* by use of a compound parabolic concentrator: effect of global solar radiation and reactor surface treatment

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Abstract Effects of photocatalytic and solar disinfection on *Escherichia coli* K12 (TISTR 780) are examined. Experiments were conducted with titanium dioxide (TiO₂) as catalyst, coated on plates by thermal spraying, with global solar illumination. Inactivation of *E. coli* as a function of time was conducted to find the rate of inactivation of the bacteria. The results revealed that the solar disinfection in the absence of the catalyst plates did not significantly affect the rate of inactivation of the bacteria when increasing the reactor surface area of solar illumination. Black reactor surfaces were slightly more affected when increasing the area of the reactor surface. However, the catalyst reactor surface had the greatest effect on inactivation of the bacteria when the reactor surface area was increased. Water temperature less than 45 °C did not affect the decrease in the rate of inactivation of the bacteria for either solar or photocatalytic disinfection.

Keywords Solar disinfection · Photocatalytic disinfection · Compound parabolic concentrator (CPC)

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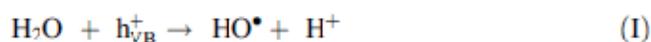
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Introduction

Chemical and biological contamination of drinking water has recently become a critical problem for people in many countries. These problems must be eliminated; for example, bacteria must be removed by disinfection before human consumption of water. The methods used for disinfection of water include heating, chlorination, and ozonation but these have a negative effect on people. Chlorination, for example, leaves an unpleasant taste in drinking water because of the formation of trihalomethanes (THMs). Ozonation and heating are expensive methods of disinfection of water [1].

Solar radiation is used for disinfection of water, because its pasteurization effect successfully inactivates microorganisms [2–5]. Recent papers discuss UV disinfection by considering the UV dose in kJ/m^2 or as W/m^2 [6]. In some experiments solar energy used for disinfection has been measured in kJ/l . [7, 8]. The semiconductor TiO_2 has been added as a photocatalyst to enhance inactivation of bacteria by solar radiation [9–12]. Photocatalytic disinfection for inactivation of bacteria in drinking water, by use of commercial titanium dioxide (TiO_2), has been used to evaluate the effectiveness of solar disinfection against *E. coli* compared with solar photocatalytic disinfection [13–15]. TiO_2 is used in solar photocatalysis because it generates the reactive hydroxyl radical OH^\bullet on its surface [16, 17]. TiO_2 has been used for photocatalytic disinfection in water treatment [18–21] and has also proved effective against organic contaminants [22–24]. In recent years, there has been interest in using compound parabolic concentrators (CPC) with TiO_2 because of its efficiency of collection of both diffuse and direct sunlight [25], and solar photocatalytic disinfection by the CPC method has been reported to achieve inactivation of bacteria [26–28]. Solar photocatalysis has been helpful in improving and protecting health in some countries.

Nano and conventional TiO_2 powder have been used to check the properties of coating by thermal spraying, a new and rapid means of coating plates [29]. This method was used in our work on inactivation of bacteria. The advanced oxidation process (AOP) is another new technique in which the hydroxyl radical (OH^\bullet) is produced when the semiconductor is excited by UV radiation of wavelength near 390 nm [30]. The semiconductor produces electrons and holes ($e_{\text{CB}}^-/h_{\text{VB}}^+$) which separate into the conduction and valence bands [31]. This reaction in TiO_2 photocatalysis involves water and dissolved oxygen, as summarized in Eqs. (I) and (II):



This research was conducted to evaluate and analyze the effect of use of TiO_2 and CPC on photocatalytic disinfection of water by use of solar energy, using *E. coli* as indicator organism. Experiments were conducted to determine the effects of different conditions, for example area of solar irradiation, water temperature, and area of fixed catalyst. Three types of reactor surface were used:

- untreated reactor surface;
- black reactor surface; and
- catalyst-coated reactor surface.

Experimental

Preparation of the black reactor and the photocatalytic reactor

Glass plates 7 cm × 20 cm × 6 mm and 7 cm × 5 cm × 6 mm were used to construct reactors for disinfection of water, as shown in Fig. 1a; the dimensions of each reactor were 7 cm × 5 cm × 20 cm. The bottom plate was coated with acrylic matt black spray (TOA). The effectiveness of the TiO₂ photocatalyst (Amperit) was also evaluated in a reactor. The TiO₂ was coated on a 7 cm × 20 cm × 6 mm stainless-steel plate by high-current, high-temperature thermal spraying (Fig 1b). Before coating, the plate was cleaned by sandblasting. The coating was 3 nm thick. These plates were placed in the bottom of the reactors (Fig. 1c).

Reactor setup

All experiments were conducted at King Mongkut's University of Technology Thonburi, Bangkok (13° 45'N 100° 31'E). Experiments were conducted on three types of reactor surface. This research was conducted to evaluate and analyze the effect of use of TiO₂ and CPC on photocatalytic disinfection of water by use of solar energy, using *E. coli* as indicator organism. Experiments were conducted to determine the effects of different conditions, for example area of solar irradiation, water temperature, and area of fixed catalyst. Three types of reactor surface were used:

- untreated reactor surface;
- black reactor surface; and
- catalyst-coated reactor surface.

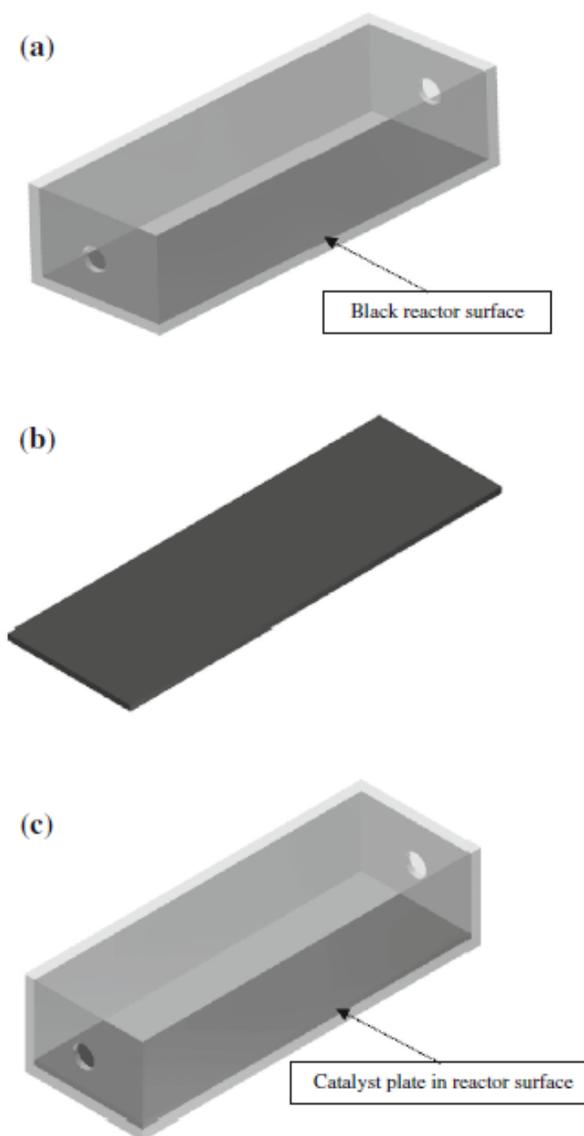
Four reactors were connected in series in the CPC. The volume of water used for each experiment was six liters and water flow from a reservoir went directly through a sampling valve, regulator valve, pump, and flow meter in series with the reactors, and was then returned to the reservoir (Fig. 2). The experimental conditions are listed in Table 1.

The reactors were positioned in a CPC aligned in a north–south direction on a platform tilted at an angle of 14°. The reflector contain a flat receiver, as shown in Fig. 3a, and was designed to focus solar radiation on to the reactor, as shown in Fig. 3b.

All variables were calculated as reported elsewhere [32].

$$H = \frac{a}{2} \left(\frac{1}{\tan \theta_c} + \frac{1}{\tan \theta_c \sin \theta_c} \right) \quad (1)$$

Fig. 1 **a** The black reactor surface. **b** Catalyst (TiO_2) coated on a plate by thermal spraying. **c** catalyst plate in the reactor



$$f = \frac{a}{2}(1 + \sin \theta_c) \quad (2)$$

$$x^2 = 4fy \quad (3)$$

where H is the height of the CPC, a is the receiver, θ_c is the acceptance half-angle, and f is the focus of the parabola. This experiment was conducted in Bangkok, so θ_c was 21° and the width of the flat receiver was 12.5 cm. Therefore, as calculated from Eqs. (1)–(3), the height of the CPC was 0.617 m and the focus of the parabola was 0.084 m. The CPC was designed to reduce cost and increase the intensity of the solar irradiation. The dimensions of the CPC were 61.7 cm \times 12.5 cm \times 100 cm.

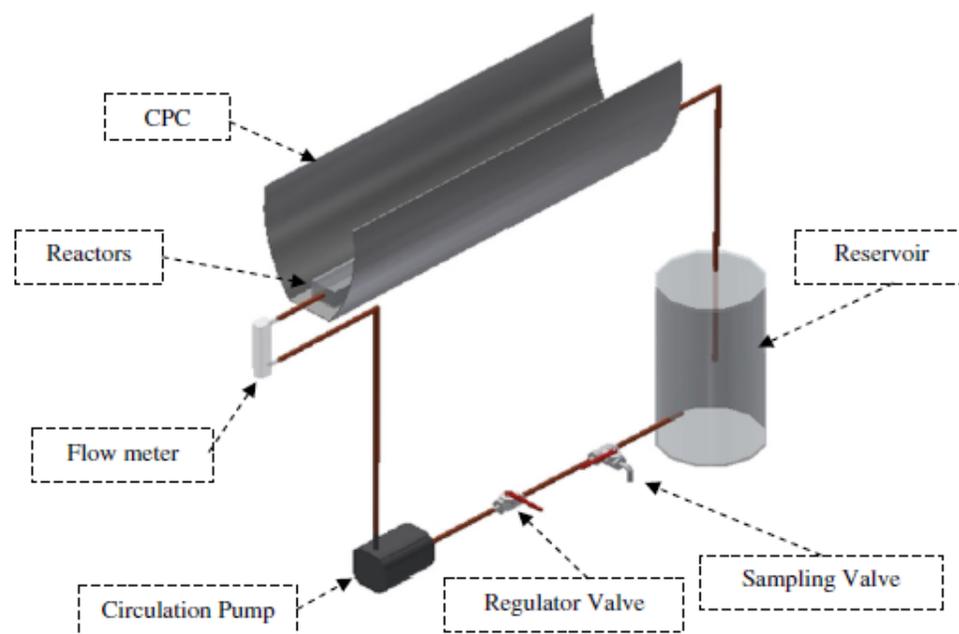


Fig. 2 Diagram of the experiment

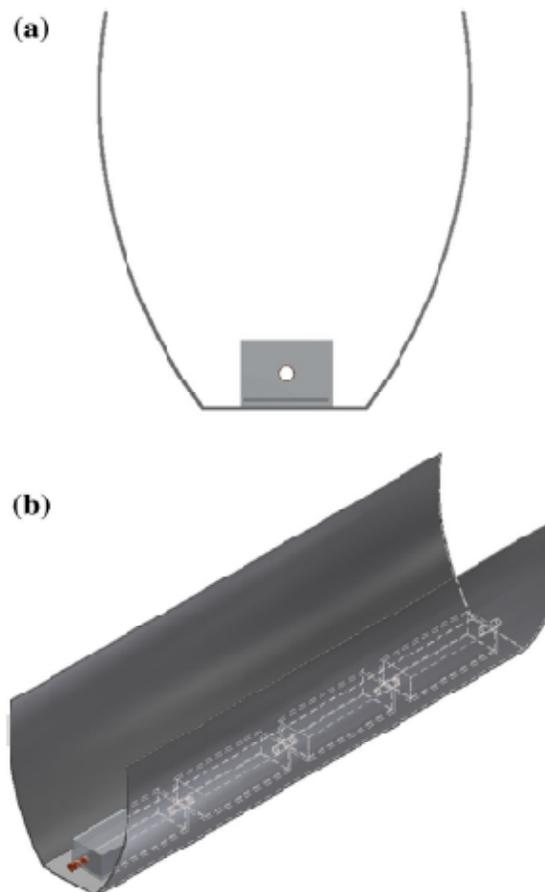
Table 1 Experimental conditions

Illuminated surface	0.014 m ²	0.028 m ²	0.042 m ²	0.056 m ²
Number of reactors	1	2	3	4
Total volume	6 L			
Flow rate	2 L/min			
Treatment time	90 min			

Reactor surfaces

The effect of sunlight on inactivation of bacteria was tested for different reactor surface areas, 0.014, 0.028, 0.042, and 0.056 m², to analyze the effectiveness of inactivation of bacteria. Experiments were performed in a four-reactor configuration, shown in Fig. 4, in which each reactor had the same surface area. The reactors were illuminated by solar radiation using different reactor surface areas. The first experiments were performed without TiO₂ to analyze the inactivation of bacteria by natural sunlight. The second experiments were performed without TiO₂ but with a black reactor surface area to absorb photons during solar irradiation. The third experiments were performed with TiO₂ to determine the effect of photocatalytic disinfection. All experiments were performed under the same conditions. Experiments were conducted to determine the effect of solar light intensity.

Fig. 3 **a** Front of the CPC and reactor, **b** four reactors connected in series



Microorganisms, culture media, and culture conditions

The *E. coli* bacterial strain K12 (TISTR 780) was obtained from the Thailand Institute of Scientific and Technological Research. An initial volume of 5 mL overnight culture of *E. coli* was re-suspended in 100 mL Luria Broth (LB) liquid medium and further incubated at 37 °C with shaking at 100 rpm until the stationary state was reached (OD 0.85–0.95 at 650 nm or 10^8 – 10^9 colony-forming units (CFU)/mL). Bacterial cells were harvested by centrifugation at $7000 \times g$ for 15 min and the pellets were washed twice in tryptose solution to complete removal of culture medium. Each bacterial cell pellet was resuspended in 100 mL tryptose solution and 5 mL of this was used to inoculate photocatalytic reactors previously filled with 100 mL isotonic solution (NaCl 8 g/L, KCl 0.8 g/L). The initial concentration of bacterial cells ranged from 10^7 to 10^8 CFU/mL.

Experimental procedures

Photocatalytic activity was evaluated from bactericidal effect against *E. coli* under the solar reflector. The bactericidal effect was determined by measuring viable

Effect of solar radiation and reactor surface treatment:

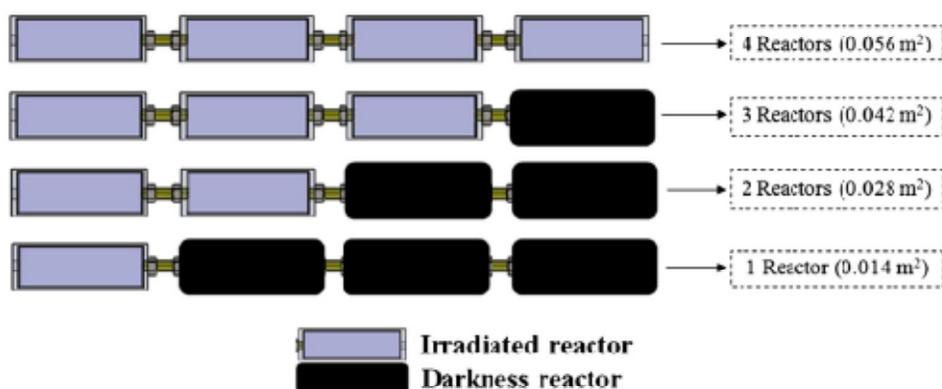


Fig. 4 Reactor configurations

E. coli cells after irradiation for 15 to 90 min. Experiments were designed to quantify disinfection efficacy of the reactor with time. For each day of testing, the feed pump was turned on at 9:00 a.m. at a flow rate of 10 mL/min.

The feed suspension of bacteria used for reactor testing was prepared in 6.0 L autoclave-sterilized de-ionized water in a 7.5-L container. Approximately 400 mL *E. coli* culture was added to the 6 L water to produce a suspension of initial volume 6.4 L. The initial cell count was 10^7 CFU/mL. During operation, the progress of disinfection was monitored by sampling the reactor contents at 15-min intervals. The feed was sampled at 10:45 a.m. and 12:15 p.m. The effluent was sampled at 15-min intervals starting at 11:00 a.m. and ending at 12:15 p.m. A 10-mL sample was collected in a sterilized tube; feed samples were obtained from the feed reservoir.

Photocatalytic disinfection on treated surfaces

Experiments were conducted with the CPC operated in recirculation mode. The total volume of the reactor was 6.04 L, of which 1.04 L was irradiated. The reactor was connected to a pump to maintain a continuous flow of 10 mL/min at 32 °C. The four reactor blocks were placed at the centre of the CPC to maintain the total radiated volume at 260 mL/reactor or 1.04 L for four reactors. Before each experiment, the reactor was cleaned by pumping sterilized deionized water through it for approximately 10 min. To follow the effect of photocatalytic treatment and to evaluate bactericidal activity, bacterial cell concentrations ranging from 10^7 to 10^8 CFU/mL were inoculated, and every 15 min during radiation 10 mL was sampled to monitor the effects of the photocatalytic treatment on the number of cultivable cells and on viability. The antibacterial activity of TiO₂-coated surfaces was evaluated the reactor for different exposure times. The TiO₂-coated surface reactors were inoculated with the bacterial suspension and placed in the center of the CPC exposed to solar radiation. After the experimental treatment, bacteria were recovered by washing twice with sterile deionized water.

Enumeration of cultivable cells

The number of cultivable cells was determined by use of a standard plate count method. The bacterial cells were cultivated in an appropriate dilution of the suspension, in triplicate, on agar medium, for 24–48 h, at 37 °C. Additional verification counts were performed after 48 h. Each experiment was repeated at least three times. The number of viable bacteria was expressed as the logarithm of the number of CFU.

Solar radiation measurement and assessment

Global solar radiation was measured with a pyranometer (model-CM11; Kipp and Zonen), which was mounted at the same angle as the CPC, i.e. 14°; the units of solar radiation were incident W/m².

Results and discussion

Effect of global solar radiation and accumulated solar energy on inactivation of bacteria

Figure 5a shows bacteria concentration (log (CFU/mL)) plotted against time.

The concentration of bacteria decreased with almost the same kinetics under different doses of solar energy under each condition in 90 min. The reactor surface (area 0.056 m²) absorbed photons from solar radiation, the highest amount of which was 47 MJ/m² in 90 min, but had no significantly greater effect on inactivation of bacteria. The amount of solar energy on the reactor surface areas of 0.042, 0.028, and 0.014 m² was approximately 4.0 MJ/m² in 90 min, and reduced the number of bacteria by 2 logs CFU. In all four experiments, results for inactivation of bacteria were not significantly different, as reported elsewhere [31]. Therefore, a larger reactor surface area does reduce the concentration of bacteria better than a smaller reactor surface area. The smallest reactor surface area had the highest bacteria disinfection efficiency in 90 min. The results were analyzed to determine the cause of inactivation of bacteria, and showed that a small number of incoming photons from solar energy can cause inactivation of bacteria. Another reason was that the number of incoming photons was insufficient to have more effect on inactivation of bacteria in a continuous flow system. Moreover, the bacteria inactivation results also show the bacteria inactivation rate as a function of accumulated solar energy, because in this work analyzed the effect of water temperature and accumulated solar energy on inactivation of bacteria. Global solar radiation was measured as solar irradiance incident per unit surface area (W/m²). Moreover, the reactor surface area received illumination by solar energy dose (J/m²) and the accumulated solar energy per unit (kJ/L):

$$Q_n = \sum_n Q_{n-1} \cdot \frac{A}{Vt} \cdot \Delta t_n \quad \Delta t_n = t_n - t_{n-1}$$

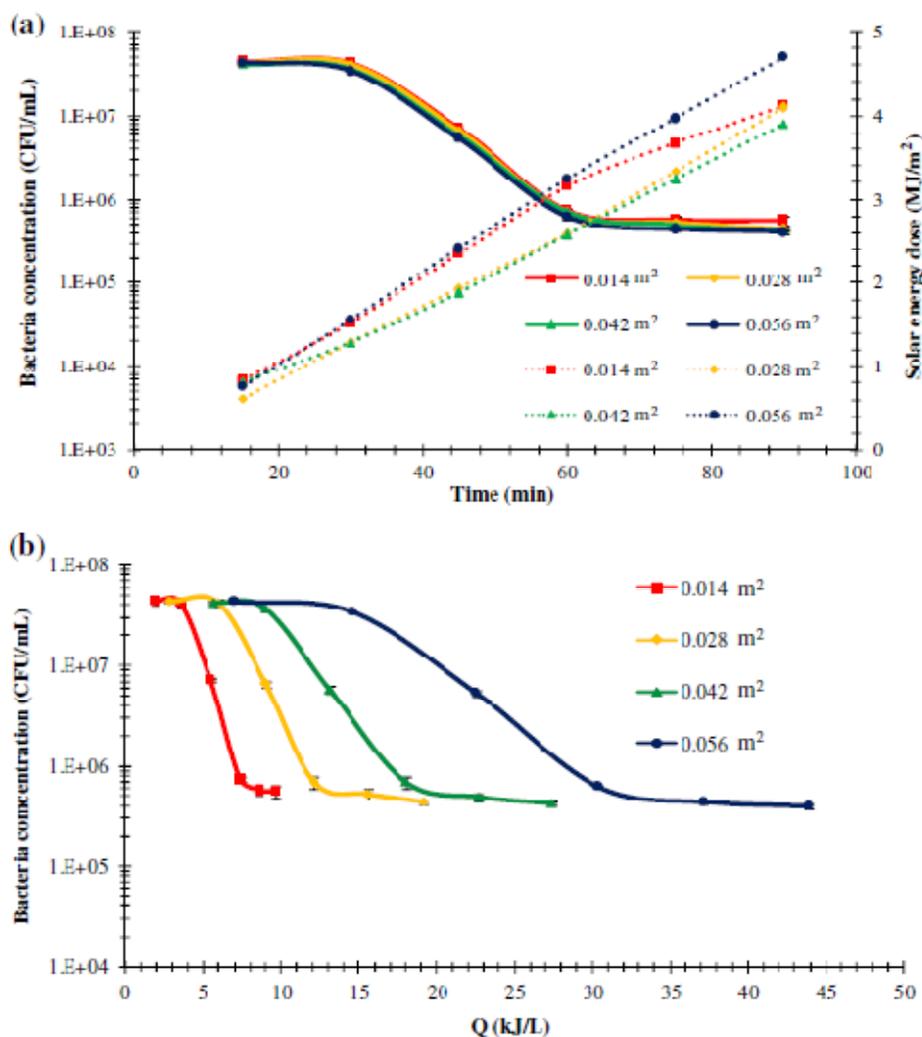


Fig. 5 **a** Effect of amount of solar energy (dotted lines) and time on solar disinfection of water containing *E. coli* (log (CFU/mL)) for different reactor surface areas (straight lines). **b** Effect of accumulated solar energy on inactivation of bacteria of *E. coli* (log (CFU/mL)) for different reactor surface areas (straight lines)

where Q_{n-1} and Q_n are accumulated solar energy received, Δt_n is the time period between sampling, A is the area of the irradiated surface, and V_t is total water volume.

All four results for accumulated solar energy in Fig. 5b show that the low-surface-area reactor has more effect on inactivation of bacteria. The reactor surface area of $0.014\ m^2$ illuminated by accumulated solar energy of only 10 kJ/L reduced the number of bacteria by 2 logs CFU whereas the other reactors with different surface areas, which received more accumulated solar energy, resulted in less efficient inactivation of bacteria. Incoming photons resulting in accumulated solar

energy of only 10 kJ/L was enough to reduce the amount of *E. coli* by 2 logs CFU. Therefore, the low light intensity of solar radiation was sufficient to generate photons on reactor surfaces. This effect could be explained on the basis that the system was tested under continuous flow conditions. Therefore, *E. coli* could recover when it was not exposed to solar irradiation. Moreover, reactor surfaces received discontinuous solar light intensities and hence inactivated bacteria less efficiently, in agreement with previous work [1, 26, 31] which showed that in continuous flow systems a larger illuminated surface area results in no significant improvement of solar disinfection. Therefore, these results could be used for cost saving in terms of smaller surface areas which is very interesting for low-cost processes for inactivation of bacteria.

Effect of global solar radiation and accumulated solar energy on black reactor surfaces on inactivation of bacteria

It is well known that black objects can absorb more solar energy. Therefore, in this study, a reactor with a black surface area was used for solar disinfection. All the experiments showed that inactivation of bacteria in 90 min decreased slightly differently, as shown in Fig. 6a. However, for the black reactor the surface area was 0.056 m², which was illuminated by a solar energy dose of only 2.5 MJ/m² in 90 min; the result showed that bacteria concentration decreased by 2 logs CFU. Nevertheless, the black reactor surface area of 0.014 m² was illuminated by a high solar energy dose of 4.5 MJ/m² in 90 min and inactivation of bacteria decreased only slightly and less than in other experiments because it had less area to absorb photons to attack bacteria cells. So, the black reactor surface could be used to receive solar energy to absorb more photons to attack bacteria cells. Therefore, increasing the black reactor surface area was slightly more effective for inactivation of bacteria. Moreover, this experiment showed that the black reactor surfaces could increase light photons to damage bacteria cells which is a superior method to apply for inactivation of bacteria. Moreover, the results in Fig. 6b show the black reactor surface area of 0.014 m² had the highest efficiency of inactivation of bacteria because it reduced the concentration of bacteria by almost 2 logs CFU for an accumulated solar energy of 11 kJ/L. However, black reactor surface areas of 0.028, 0.042, and 0.056 m² were illuminated by accumulated solar energy of 14, 20, and 16 kJ/L, respectively, and were able to reduce the concentration of bacteria by 2 logs CFU. This result showed that the efficiency of a black surface area of 0.014 m² is sufficient to inhibit growth of bacteria. The results reveal *E. coli* received discontinuous solar irradiation and could recover during flow in the dark. Therefore, the results show that a lower surface area is more efficient at inactivation of bacteria, as reported elsewhere [26, 31]. However, the reactor surface area of 0.056 m² is slightly more effective on inactivation of bacteria than the reactor surface of 0.042 m² with accumulated solar energy of 15 kJ/L because it received solar light of lower intensity. Therefore, a larger surface area could be used for disinfection for low light intensity. The reactor surface area of 0.056 m² performed slightly better in reducing bacteria concentration. Therefore, these results can be applied to save costs, and so use of black reactor surface areas is a good technique for low-cost inactivation of bacteria.

Effect of solar radiation and reactor surface treatment

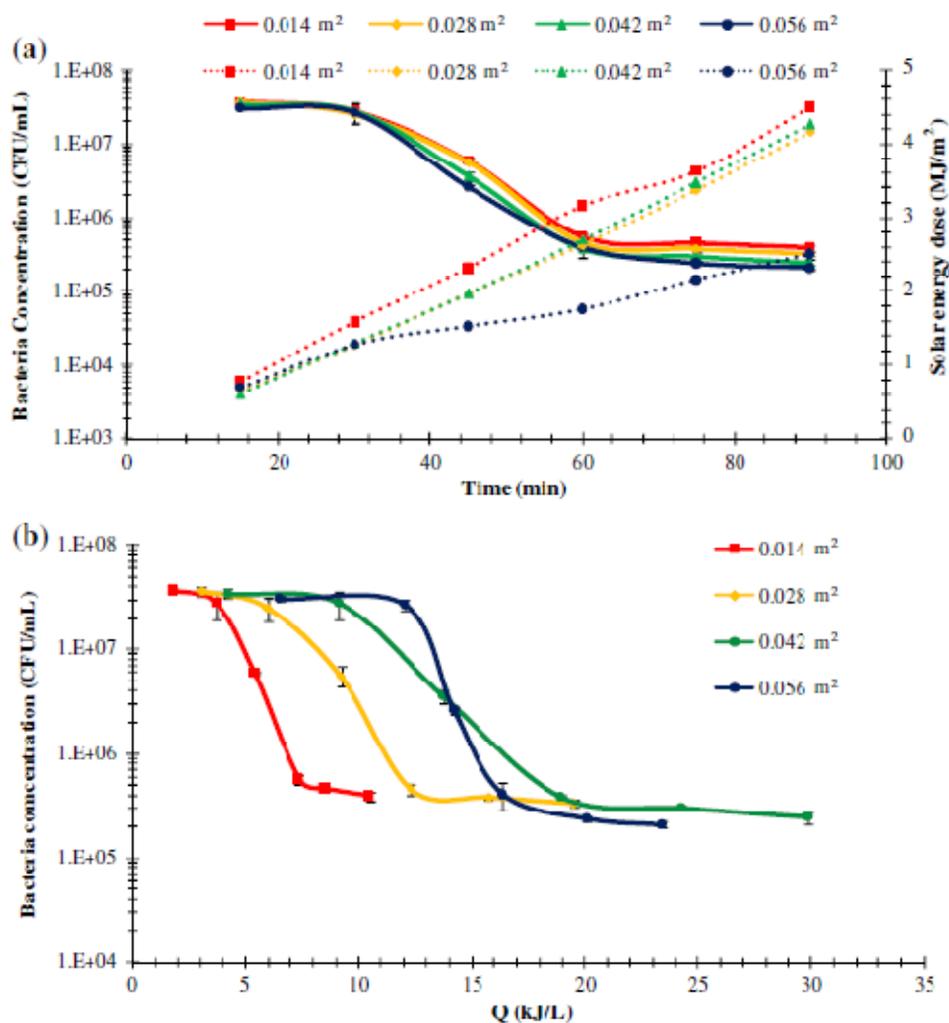


Fig. 6 **a** Effect of amount of solar energy (*dotted lines*) and experiment time on inactivation of bacteria of *E. coli* (log (CFU/mL)) by use of different reactor surface areas (*straight lines*). **b** Effect of accumulated solar energy on inactivation of bacteria (log (CFU/mL)) by use of different reactor surface areas (*straight lines*)

Effect of global solar irradiation and accumulated solar energy on inactivation of bacteria in the presence of TiO₂

It has been proved that the catalyst TiO₂ can reduce the concentration of bacteria when illuminated by solar radiation for photocatalytic disinfection [10, 11, 22]. A CPC solar reactor has been shown to enable efficient inactivation of bacteria by solar photocatalysis with immobilized TiO₂ during treatment times of 15–90 min. These experiments were set to enhance the effectiveness of inactivation of bacteria by adding TiO₂. The results showed the catalyst-coated reactor surface area of 0.056 m² was illuminated by low light intensity by a solar energy dose of only 3.6 MJ/m² in 90 min,

but this enabled the most effective inactivation of bacteria, by approximately 4 logs CFU, because the large surface area received incoming photons to attack bacteria cells, whereas a catalyst-coated reactor surface area of 0.014 m^2 with the highest solar energy dose of 5.2 MJ/m^2 reduced *E. coli* concentration by only 2 logs CFU in 90 min because of the small number of photons. Moreover, the catalyst-coated reactor surface area of 0.028 m^2 was illuminated by a solar energy dose up to 4.5 MJ/m^2 more than the catalyst-coated reactor surface area of 0.042 m^2 , which was illuminated by a solar energy dose of 4.3 MJ/m^2 . However, all experiments show that reactors with larger surface areas could inactivate bacteria with smaller amounts of solar energy, as shown in Fig. 7a. Therefore, an increase in catalyst-coated reactor surface area was more effective for photocatalytic disinfection. Moreover, the graph in Fig. 7b shows the catalyst-coated reactor surface area of 0.014 m^2 was illuminated by accumulated solar energy of 12 kJ/L . This experiment shows that the lowest amount of incoming photons could reduce bacteria concentration and had the highest effect on inactivation of bacteria of 2 logs CFU. However, the lower accumulated solar energy was no more effective at reducing the concentration of bacteria, and the catalyst-coated reactor surface area of 0.028 m^2 illuminated by accumulated solar energy of 22 kJ/L performed slightly at reducing the concentration of bacteria than the catalyst-coated reactor surface area of 0.014 m^2 . Moreover, the catalyst-coated reactor surface area of 0.042 m^2 , which received more incoming photons by accumulated solar energy up to 32 kJ/L , could reduce the concentration of bacteria by 3 logs CFU. Furthermore, the catalyst reactor surface area of 0.056 m^2 reduced the concentration of bacteria by 4 logs CFU with solar energy of 35 kJ/L . These results showed the larger catalyst-coated reactor surface area received more incoming photons to attack bacteria cells. Therefore, the reactor surface area of 0.056 m^2 reduces the concentration of bacteria more effectively than reactor surface areas of 0.042 , 0.028 , and 0.014 m^2 . This is because *E. coli* moves into the dark, where it receives discontinuous solar light intensity and is not affected by photocatalytic disinfection. Moreover, previous work has shown that intermittent solar light required more time to complete photocatalytic disinfection [14]. Deactivation of bacteria by solar radiation in a CPC occurs whether or not the catalyst was present. Total photocatalytic deactivation of bacterial cell suspensions was a consequence of the combined effect of solar radiation and the oxidant species generated in the immobilized TiO_2 . However, although solar radiation deactivated bacterial cells, deactivation was not complete, because bacterial regrowth was detected. The method might be improved by photocatalytic action under solar radiation to deactivate bacteria completely, enabling photocatalytic disinfection of water. The irradiated area of the reactor is of crucial importance in the inactivation of bacteria. There was also a synergistic effect of run time and flow rate of the suspension in the reactor, because of stress from the flow rate on the bacteria suspensions.

Effect of water temperature, in the absence and presence of TiO_2 , on inactivation of bacteria

These experiments were conducted to observe the effect of water temperature on inactivation of bacteria by solar energy and different reactor surface area. The results in Fig. 8a show that water temperature was increased by solar radiation. The

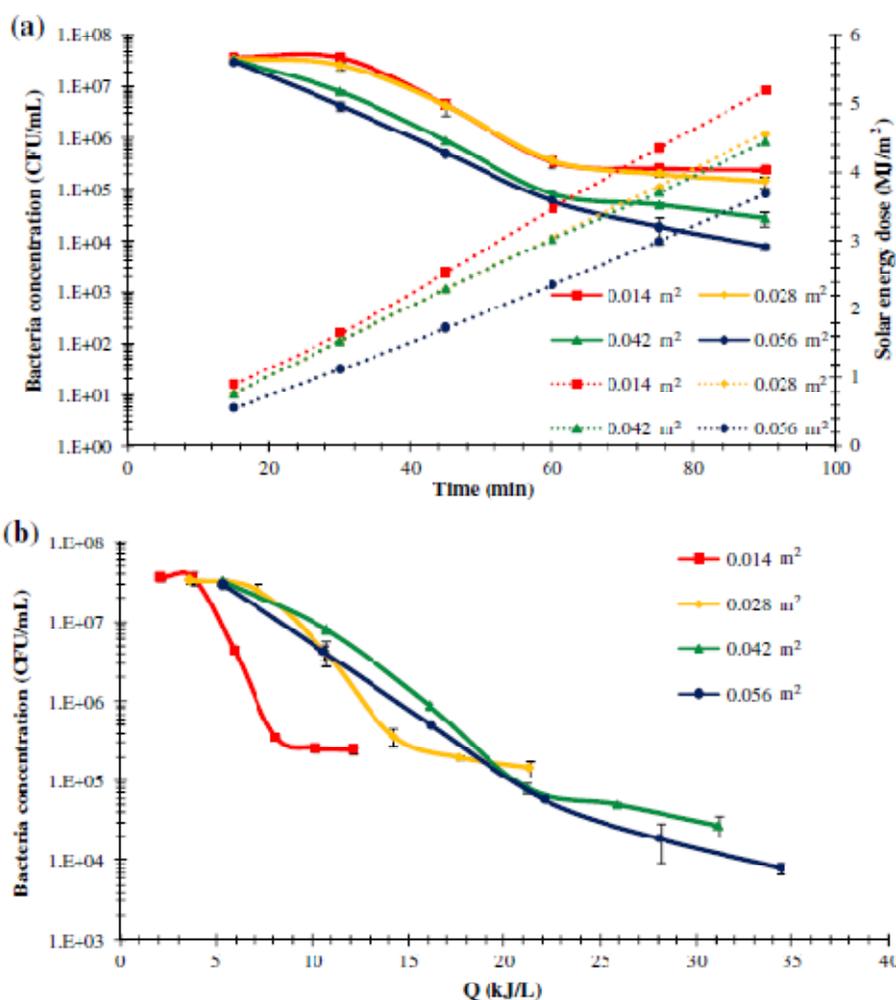


Fig. 7 **a** Effect of solar energy dose (*dotted lines*) and experiment time on photocatalytic disinfection of *E. coli* (log (CFU/mL)) with different reactor surface areas (*straight lines*). **b** Effect of accumulated solar energy (kJ/L) on inactivation of *E. coli* bacteria log (CFU/mL) by use of different reactor surface areas (*straight lines*)

water temperature for a reactor surface area of 0.056 m², the largest reactor surface area, was increased to 45 °C. However, this was no more effective than other temperatures at inactivation of bacteria. Water temperature for reactor surface areas of 0.014, 0.028, and 0.042 m² was increased to approximately 39 °C but this had the same effect on the concentration of bacteria as the 0.056 m² reactor surface. Therefore, the experiments showed that water temperatures up to 45 °C did not affect inactivation of bacteria, in contrast with the pasteurizing effect of radiation at temperatures higher than 45–50 °C [33]. Our results also showed that water temperature in the reactor with a catalyst-coated surface area of 0.014 m² was increased to much higher than 45 °C but in this experiment bacteria concentration

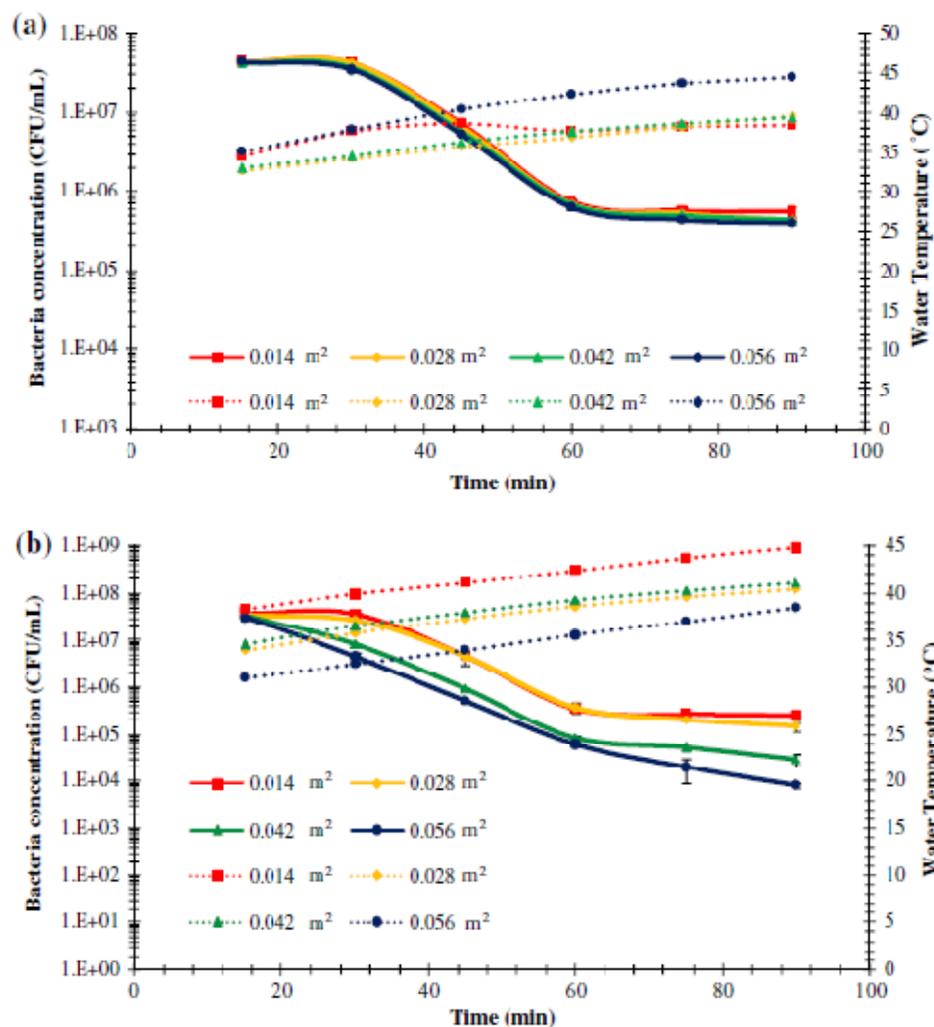


Fig. 8 Effect of water temperature (*dotted lines*) and experiment time on inactivation of *E. coli* (log CFU/mL) with different reactor surface areas (*straight lines*): **a** in the absence of TiO₂ and **b** in the presence of TiO₂

was reduced by 2 logs CFU only because it had the least surface area to absorb solar irradiation. Moreover, water temperature in reactors with surface areas of 0.028 m² and 0.042 m² was increased equally, to 40 °C. However, a reactor surface area of 0.042 m² was more effective at inactivation of bacteria. Another result in support of this observation was that water temperature in the reactor with a surface area of 0.056 m² was increased by the lowest amount, to 37 °C only, but the reactor surface area of 0.056 m² had the most effect on photocatalytic disinfection. Therefore, water temperatures not above 45 °C did not affect photocatalytic disinfection, as shown in Fig. 8b.

Conclusions

Solar-only disinfection and photocatalytic disinfection in conjunction with a compound parabolic concentrator (CPC) were demonstrated to inactivate *Escherichia coli* K12 (TISTR 780) in a continuous flow reactor. Global solar radiation is a major light source for use as a photon generator. A series of experiments were set up to examine the effect of a cumulative global solar energy and reactor surface treatment. The effectiveness of the two systems in reducing the concentration of bacteria was compared. The effectiveness of solar disinfection showed that smaller solar-illuminated surface areas and black surface areas were more efficient at deactivating bacteria in continuous flow systems. However, the larger black surface area reactor was slightly more effective at inactivation of bacteria because more photons to damage bacteria were absorbed by the black surface reactor. Similarly, the larger catalyst-coated surface area received more incoming photons from solar energy, which resulted in high performance in deactivation of bacteria cells. Furthermore, the reactor coated with TiO₂ by thermal spraying enabled high-performance inactivation of bacteria. The results showed that low-cost inactivation of bacteria can be achieved by use of a continuous flow process with proper design of a CPC module and reactor surface.

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