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Comparative study of different solar-based photo catalytic reactors for disinfection of contaminated water

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ABSTRACT

In this work, two solar-based photo catalytic reactors having different geometries and configurations for the disinfection of contaminated water were designed and tested. These solar-based reactors are stair-type open exposure and parabolic trough concentrator (PTC). For the first type, the stairs were coated with TiO_2 using sol-gel method, whereas the parabolic type concentrator used a titania-coated rod. For monitoring of disinfection process, the parameters such as solar irradiation exposure time and bacterial deactivation counts were investigated carefully. On contrary to commercial slurry-based photo reactor reported in the literature, thin film titania-coated reactor's performance was investigated in this work. Moreover, detailed designed aspects of the reactors, process parameters, and reactor operation are presented and discussed. The target aqueous matrix was drinking water developed in laboratory having 10^6 CFU/mL bacterial strain. Experimental results revealed that during 20 min of solar radiation exposure, 28 and 75% of the initial bacterial strain disinfection was recorded in staircase and PTC photo reactor, respectively. Moreover, results revealed that 1.75 kJ/L of UV energy is required in PTC photo reactor to achieve 75% reduction of initial bacterial population, whereas 4.1 kJ/L of UV is required to produce the same result in staircase photo reactor. The PTC showed more effective inactivation of *E coli* bacteria than the staircase-type photo reactors. This study is highly significant for designing of large-scale PTC reactors for clean water supplies for rural population and remote locations.

Keywords: Stair case photo reactor; Parabolic trough concentrator photoreceptor; Titania thin films; Solar disinfection

1. Introduction

Clean water is essential for the living organism (humans, animals, and agricultural plants), sustainable development of any country, and for energy/power generation. The supply of safe drinking water is an issue of great concern, particularly in developing countries. Although several methods have been developed to supply safe drinking water to urban populations, efforts are still needed to achieve required

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targets for sustainable development and supply of enough quantity of safe water in these countries to millions of people living in rural areas. According to WHO, in Africa, Asia, Latin America, and the Caribbean, nearly 1.5 billion people have no access to clean drinking water [1]. The lack of access to safe drinking water is directly related to poverty due to poor health. The direct and indirect human costs of water born diseases due to drinking the polluted water are enormous, including widespread health problems, heavy labor costs leading to severe limitations for economic development. Fortunately, most of the clean water deficit countries lie in the region having high average solar irradiation flux round the year. Hence, it is blessing by nature to develop some cost-effective solar based disinfectant units for clean water supplies to these communities using solar radiations.

The increasing concern for pollution (or pathogenic)-related water diseases has promoted the implementation of more stringent standards on polluted water and wastewater to sufficiently meet existing safe standards for requirements in bathing area protection, drinking water, wastewater discharge, and reuse for better public health and environmental protection. Consequently, specific waste water treatment techniques need to be developed in the treatment chains by considering the water quality objectives in terms of technical reliability, economic, and environmental criteria. Various chemical processes such as activated carbon, coagulation, and multimedia sand filtration have been applied for removing the microorganism or other pollutants. These conventional technologies only separate the contaminated substances from the treated water to another solid form which requires further treatment and dispose offs. This increases the additional cost incurred for the decontamination and cleaning of water. Chlorination is a method which is universally accepted for water disinfection process in preventing waterborne infectious diseases, bearing the advantages of low operating cost in a broader range of pH, lower dosages, and contact times with chlorine dioxide as effective disinfectant. However, the main disadvantages of chlorination for microorganism inactivation could be summarized as follows: (a) production of toxic by-products such as trihalomethanes, haloacetic acids, and other dissolved organic halogens as potential human carcinogens even at low concentration; (b) poor inactivation efficiency of spores, cysts, and some viruses at low dosages used for bacterial coliform removal; (c) varying efficiency affected by the presence of nitrite (nitrification effect) and suspended solid concentrations; and (d) high investments for scrubbing safety equipment and need for dechlorination to meet stringent EPA regulatory limits [2–4].

Reverse osmosis technology has been also reported as a cost-effective technology accommodating the above-mentioned needs, for maintaining excellent water quality with higher reliability. However, the presence of impurity, particularly bacteria fouling on membrane surface, has been commonly known as one of the major problem responsible for disrupting the efficiency of the membrane over the course of a lifetime, where the fouling status is critically influenced by variations in wastewater quality [5].

To overcome the above-mentioned drawbacks of existing conventional methods, specific clean water technology needs to be developed in the water treatment systems and this is exactly what we developed at our laboratories using elegant photo catalytic process, novel nano-engineered materials, and using abundant natural resource of solar radiations as an excitation source. This technology is based on "Advanced Oxidation Processes (AOP)" which is considered as innovative water treatment technologies [6,7]. Therefore, recently, research focus is more and more on those AOPs which can be driven by solar irradiation and heterogeneous catalysis [8]. Among the semiconductor catalysts applied in AOP, titanium dioxide (TiO_2) has been focus and shown great interest by scientist working to develop photo catalysis technology. The TiO_2 is the most active photo catalyst under the photon energy of ($300\ nm < \lambda < 390\ nm$) and remains stable after the repeated catalytic cycles, whereas CdS or GaP are degraded along with to produce toxic products [9].

Initially, when the semiconductor catalyst like titania is excited with sun radiation having energy higher than the band gap of catalyst, the electron hole pairs are generated by photoexcitation which can move to the surface of semiconductor particle to form a highly oxidizing radicals such as $OH^{\bullet}$ (hydroxyl radical) and $O_2^{\bullet -}$, (super-oxide radical) and these radicals effectively oxidize the cell membrane and damage the microbial organism [10]. The hydroxyl radical generates oxygen while H+ ions form hydrogen by capturing conduction band electrons. These highly active species, e.g. hydroxyl radical ($OH^{\bullet}$) and $O_2^{\bullet -}$, can mineralize the organic compounds [11–16], disinfect the pathogens and other living organism [17–19]. Titania slurry-based bacterial disinfection has been reported in [20]. The main disadvantage of slurry-based photo reaction after the successful disinfection process is to remove the slurry by post-treatment filtration process which adds cost and extra work.

There is a consensus in research community regarding the mechanism of destruction of bacteria by photo catalysis [9]. For the photo catalysis disinfection process, the microorganism outer wall is thought to be

first site of attack by highly reactive hydroxyl radicals. Rapid leakage of potassium ions from the bacteria parallel to the decrease in cell viability has been reported [21]. Maness reported some results which can be explained by peroxidation of the polyunsaturated phospholipid component of the cell membrane leading to a loss of essential cell functions, e.g. respiratory activity of bacteria, and in the end, to the cell death [22]. In another report, the cell inactivity is explained that damage takes place on the lipopolysaccharides layer of the external cell wall and on the peptidoglycan layer of bacteria. Next, the peroxidation of the lipid membrane (the radicals oxidize to fatty acids), the oxidation of the proteins membrane (amino acids) and of polysaccharides take place, consequently leading to the cell death [9].

The main objective of this work was to develop an autonomous solar photo catalysis system for the disinfection in contaminated water. Two different types of photo catalytic reactors were developed and tested: staircase and parabolic type. In the staircase design, all the stairs were coated with titania thin film, and in parabolic concentrator design, titania-coated rod was used as active photo catalytic reactor. Experiments were carried out to study the *E. coli* bacteria disinfection process against the solar irradiation exposure time and absorbed UV energy for both designed photo catalytic reactors.

2. Materials and methods

2.1. Experimental setup

2.1.1. Stair-type collectors

Non-concentrating stair-type or flat plate collector is the simplest and cheapest among all other type of collectors because they do not concentrate the solar radiations. Stairs and its support surfaces along with water storage rectangular vessel are made of stainless steel. Solar radiation collector area of the designed photo reactor is 7,670 cm^2 and is mounted on a portable table top surface. Flat plate collector designs are simple, robust, and economical in manufacturing and be able to withstand the fluid pressure during operation. The dimensions of the each stair are 50, 10.2, and 5.14 cm in length, width, and height, respectively. Ten stairs were fabricated to cover the area of 7,670 cm^2. Stair assembly inclination angle is set to the latitude of the site, 26.08° degrees due south for the maximum collection of solar radiations. A complete flow control mechanism was devised and installed in the system and is shown in Fig. 1a. Installed pump circulates the water throughout the reactor, and a flow control valve

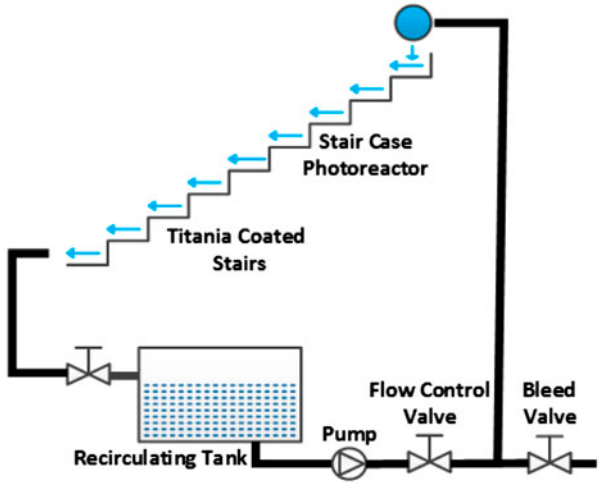

Fig. 1a. Process flow schematic of staircase photo reactor design.

Fig. 1b. Pictorial view of staircase reactor.

was used to regulate the water flow rate. The complete experimental setup is illustrated in Fig. 1b.

2.1.2. Concentrating collectors

The parabolic trough collector consists of a structure that contains reflective concentrating parabolic surface. An absorber tube containing a central rod coated with TiO_2 is fixed at the geometrical focal line of the parabolic trough concentrator (PTC) on which all the reflected solar radiations are focused. The structure is usually driven by a sun tracking system to keep the parabolic collector aperture perpendicular to the solar radiations. Fig. 2 shows the path for direct solar radiations until it enters the absorber tube for

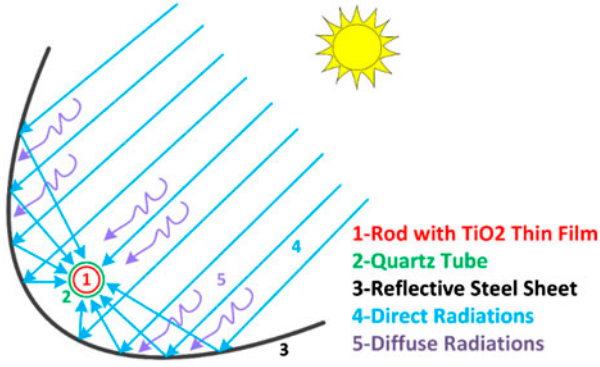

Fig. 2. PTC photo reactor with solar radiation path travel.

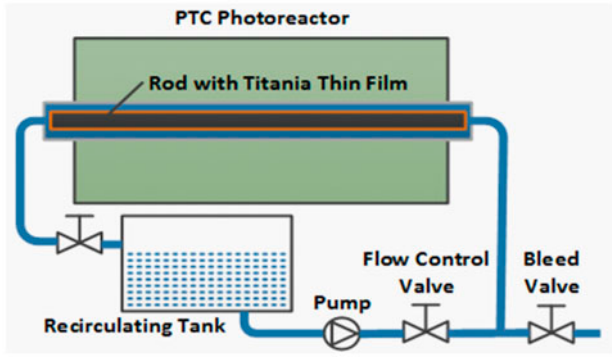

Fig. 4a. Schematic of water flow system for PTC photo reactor.

the photo catalytic process. Two factors are considered to calculate the optical efficiency of the reactor, first the mirror reflectivity and second the UV transmittance of the absorber tube material. The global radiations are affected by the transmittance of the absorber glass tube. Within the glass absorber tube, a solid rod supporting titania thin film is inserted for the photo catalysis reaction and shown in Fig. 3. To attain the better quantum efficiency of the parabolic concentrator, the ratio of the inner diameter of the tube to the diameter of the rod should be close to the refraction index of the introduced water. In our design, rod diameter with 24.5 mm and the glass tube with 31.5 mm inner dimension having 2.5 mm thickness were selected to attain the $n = 31.5/24.5 = 1.285$, which is very close to the induced water refractive index (n=1.33) at temperature of 20°C. A water circulation mechanism and flow control system were designed and installed to run the reactor in continuous or batch mode operation. The complete schematic of water flow control is depicted in Fig. 4a. The complete PTC with photo reactor along with tracker system and flow control system is illustrated in Fig. 4b.

2.2. Photocatalyst preparation and coating procedure

In order to investigate the effect of TiO_2 as a photo catalyst, a stable coating of the titania was deposited in both reactors. Uniform microstructure coating of TiO_2 on stairs (346 L Steel) and on rod (304 L Steel) was carried out by sol-gel method using Tetra n Butyle Titanat (TBT) as alkoxide material. TiO_2 solution was prepared from TBT ($C_{16}H_{36}O_4Ti$) as a starting material. 91 mol% of ethanol (C_2H_5OH) and 2 mol% of ethyl aceto acetate (EAcAc, $C_6H_{15}O_3$) were mixed at room temperature, and then, 4 mol% of TBT was added. The formed solution was stirred for 8 h continuously and aged for 24 h to complete the reaction.

Steel stair structure and rod surfaces were prepared by grinding them with 1,500 emery paper

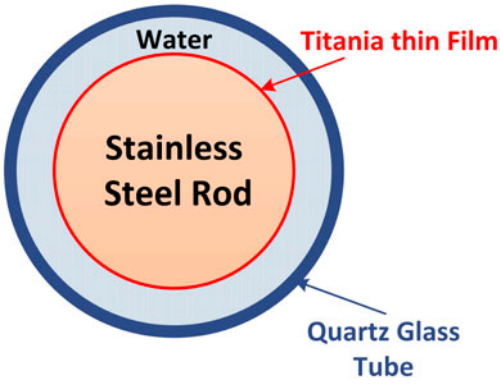

Fig. 3. Thin film titania supported rod for PTC photo reactor.

Fig. 4b. Pictorial view of experimental setup of PTC for photo catalysis.

followed by polishing with 0.3 μm Al_2O_3 powder. The surfaces were cleaned by ethanol and acetone and prepared them for dip coating. Steel rod was dipped into the solution for 10 s, removed back, and allowed to dry at room temperature. Steel stairs were coated using pressurized spray gun to provide a thin layer of solution and dried at room temperature. After natural drying at room temperature, both photo reactors were heated at 150˚C for 30 min and then calcinated at 450˚C for 1 h to remove the organic residuals. The prepared rod was inserted and fixed into the quarts glass tube. The tube and rod assembly was installed within the parabolic structure at its focal point.

2.3. Bacterial preparation and enumeration

E. coli K12 wild-type (MG 1655) bacteria were used in our studies due to its widespread used as a fecal indicator. First, nutrient broth media were sterilized at 121˚C at 2 bar pressure for 30 min duration and then, its 8 g amount was dissolved into 1,000 mL of distilled water (pH adjusted to 7.5). Later on 15 g of nutrient agar was added to the nutrient broth media and sterilized. The whole agar media were kept to cooldown before pouring into petri dishes that is further used to test the bacteria. Single colony of bacteria was grown overnight in the nutrient broth at 37˚C on a rotary shaker (160 rpm). Aliquots of the pre-culture were inoculated into fresh medium and incubated. The culture was harvested by centrifugation at 4,000 rpm for 15 min at 4˚C and washed twice with a sterile 0.9% NaCl solution at 4˚C to ensure the elimination of broth medium. Finally, pellet was resuspended to a final concentration of 1×10^9 CFU/mL in sterile water. The calculated amount (1 mL) of bacteria is added in the each liter of water used in the experiment to have the initial concentration of 1×10^6 CFU/mL.

Viable cells of bacteria in the samples taken during the experiment were analyzed by plating them on nutrient agar plates after serial dilution in 0.9% saline. Colonies were counted after 18 h incubation at 37˚C [23].

2.4. Photo catalytic bacteria removal studies

All the experiments were carried out under the sunlight having insolation 650–750 W/m^2 (700 W/m^2 average) at Dhahran (latitude 26.28 N, longitude 50.21 W), Saudi Arabia. The stair-type photo reactor was kept due south with inclination angle (26.28˚) equal to the latitude of the site where as the PTC was tracked to keep its aperture perpendicular to the sun during the experimental run. A centrifugal-type pump was used to circulate the water from reservoir to the reactor continuously, and flow control valve is used to adjust the water flow rate at 1.45 L/min for both reactors.

At the start of experiment, both reactors were cleaned and 20 L of filtered water was admitted into the reservoir tank of each reactor. 20 mL of cultured *E. coli* bacteria was added to the each reservoir tank and its water was circulated through the reactor for 10 min in the dark until the uniform bacterial concentration of 10^6 CFU/mL was attained in the whole reactor. Each of the experiment is characterized as continuously stirred tank reactor in series with the plug flow solar photo reactor. The initial uniform bacterial concentration in each experiment was 10^6 CFU/mL. At the beginning of each experiment, water sample was taken from the reservoir at $t = 0$, photo reactor was uncovered, and solar light was allowed to incident upon the collectors. The collected sample was marked and kept in the complete dark as a control throughout the whole experimental run to ensure that the bacteria die-off in the samples was only due to the solar photo catalytic reactions and not from the other mechanism such as heat or any contamination. The water was recirculated in the reactor with sunlight exposure for 2 h. Water samples were collected after every 20 min duration and taken directly to the laboratory for bacteria enumeration. It was ensured that the no bacteria die-off takes place outside the photo reactor tube. Each reactor was operated in continuous mode of operation. The average UV intensity in the solar radiations was taken the 5% of the whole solar spectrum, and it comes out to be 30–35 W/m^2 at the site of experiment.

3. Results and discussion

Before we present the results, it is worth mentioning to describe the killing phenomenon of bacteria. The electron hole pairs, generated by photoexcitation, can move to the surface of semiconductor particle to form highly oxidizing radicals such as $OH^\bullet$ (hydroxyl radical) and $O_2^{\bullet -}$, (super-oxide radical) and these radicals effectively oxidize the cell membrane and damage the microbial organism [10]. The hydroxyl radical generates oxygen, while H^+ ions form hydrogen by capturing conduction band electrons. The super-oxide radical $O_2^{\bullet -}$ and $OH^\bullet$ generated through solar radiation induced photocatalysis process as described in Eqs. (1)–(5) kill the bacteria in contaminated water, that is,

$$SC(\text{semiconductor catalyst}) \xrightarrow{h\upsilon > E_g} SC(h^+_{vb} + e^-_{cb}) \quad (1)$$

$$H_2O + h^+_{vb} \rightarrow H_2O^+ \rightarrow OH^{\cdot} + H^+ \quad (2)$$

$$OH^{\cdot} + OH^{\cdot} \rightarrow H_2O + 1/2O_2 \quad (3)$$

$$H^+ + e^-_{cb} \rightarrow H^{\cdot} + H^{\cdot} \rightarrow H_2 \quad (4)$$

$$O_2 + e^-_{cb} \rightarrow O^{\cdot}_2 \quad (5)$$

Moreover, the extent of microbial damage depends on how effectively the cell walls and cell membrane succumb to the oxidative process [24]. Data collected from the experimental runs in both reactors were plotted. Figs. 5 and 6 show the survival of bacteria population (normalized values, actual bacterial concentration/initial bacterial concentration) as a function of time for the staircase and PTC reactor, respectively. For the staircase photo reactor, it took about 20 min to disinfect the 28% of starting value of bacteria concentration. As the time progressed, bacterial deactivation was increased and approached to 93% after 140 min. Bacterial disinfection rate for PTC was faster than the staircase. Using the PCT, 75% decrease in the deactivation of bacteria is observed in the first 20 min of the experiment and it approached to 97% after 140 min of photoactivity. The initial fast decay in inactivation activity is attributed due to the high insolation flux at the photo reactor tube leading to more photo catalytic activity.

The bacterial inactivation against the cumulative solar UV radiation is also calculated according to the following relation.

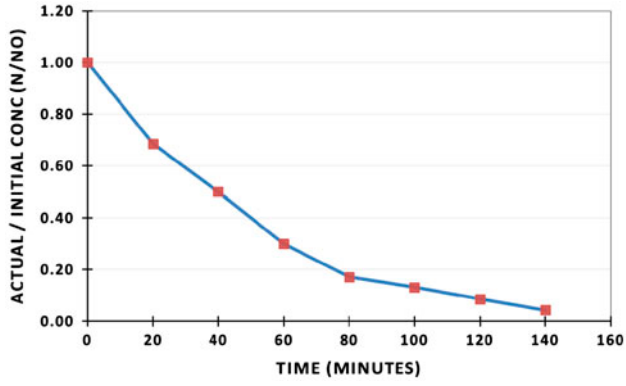

Fig. 5. Bacterial disinfection activity in staircase reactor.

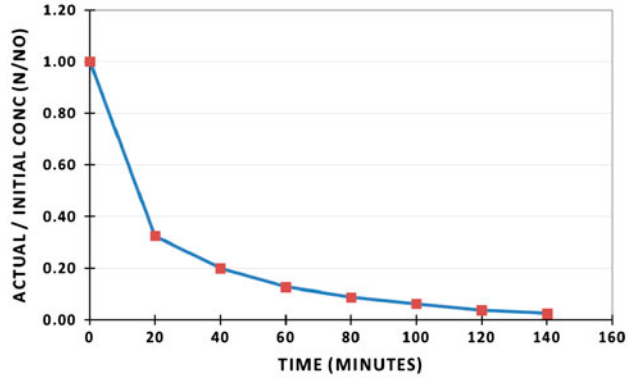

Fig. 6. Bacterial disinfection activity in PTC photo reactor.

$$Q_{UV_n} = Q_{UV_{n-1}} + \frac{\Delta t_n UV_{GN} A}{V_T}, \Delta t_n = t_n - t_{n-1}$$

In this equation, $Q_{UV_n}$and $Q_{UV_{n-1}}$ are the cumulative received UV energy per liter in the samples at time n and $n-1$. $\Delta t_n$is the time interval between two collected samples. $UV_{GN}$is the average incident ultraviolet radiation on the photo reactor during the experimental run, and its value is taken as 30 W/m^2. A is the irradiated area and, V_T is the total volume (20 L in each reactor) of the circulated fluid in the reactor. Figs. 7 and 8 show the bacterial inactivation as a function of the cumulated solar UV radiation in the staircase and PTC photo reactors, respectively. The plotted results revealed that PTC reactor performs more efficiently than the staircase reactor design. The results showed that 1.75 kJ/L of UV energy is required to achieve 75% reduction of bacterial population in PTC whereas only 4.1 kJ/L of UV energy is required to produce the same result in staircase

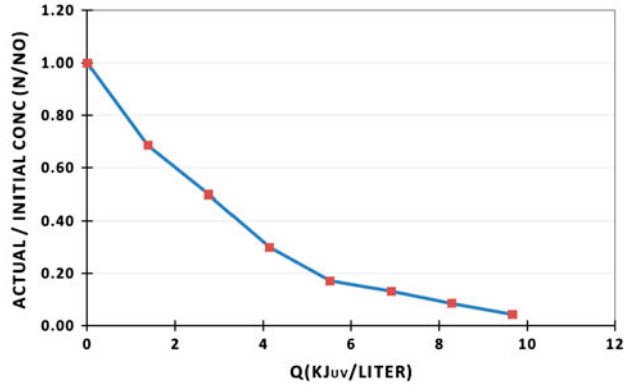

Fig. 7. Survival of *E. coli* v/s cumulative UV radiation for staircase reactor.

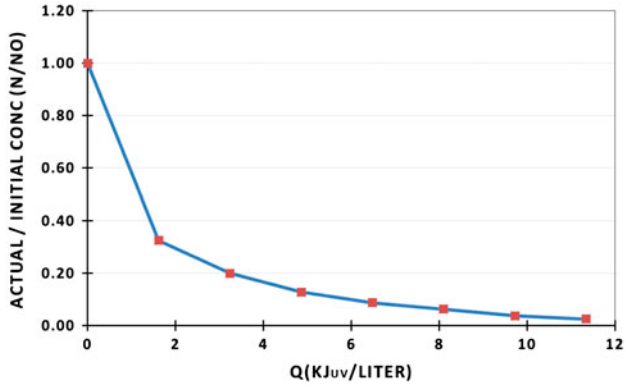

Fig. 8. Survival of *E coli* v/s cumulative UV radiation for PTC reactor.

reactor. It should be noted that these results consider the difference in collector area (and thus solar UV radiation flux) between the CP and stair reactors. The cumulative dose of UV radiation is a function of both solar intensity and collector irradiated area, consequently providing the valid comparisons between both photo reactors.

The temperature rise in the reactors during the experiment was observed from 25 to 31˚C in staircase reactor where as in parabolic reactor, it increased from 25 to 40˚C. The literature shows that any temperature related die-off bacteria starts to have synergetic effect at minimum level of 45˚C [25]. The observed photo catalytic disinfection of the bacteria is due to the overall effect of solar irradiation and oxidant species produced by the supported titania in form of thin layer through photo catalytic process and not due to the rise of temperature.

These experiments clearly reveal the benefits of PTC photo reactor over the staircase one and confirm that the PTC profile is the optimum collector configuration for the disinfection purpose. The relative difference of these two collector geometries needs some further investigation in terms of full/large-scale implementation while considering the manufacturing/fabrication, reflective material quality, tracking, and overall system cost.

4. Conclusions

Two solar disinfection photo reactors, staircase, and PTC were designed, fabricated, and demonstrated for disinfection of *E coli* bacteria. The results revealed that adequate disinfection is attained using sunlight with the use of supported catalyst in both reactors. Both collector profiles in continuous mode of flow operation were evaluated, with the PTC demonstrating a clear superiority over the staircase in terms of disinfection efficiency. Experimental results revealed that 28 and 75% of the initial bacterial concentration is disinfected in staircase and PTC photo reactor, respectively, during the first 20 min of operation. Moreover, results show that 1.75 kJ/L of UV energy is required to achieve 75% reduction of initial bacterial population in PTC photo reactor whereas 4.1 kJ/L of UV is required to produce the same result in staircase reactor. The fast decay of bacterial inactivation activity in PTC is attributed due to the high insolation flux at the tube leading to more photo catalytic activity. The attained results were only for a single bacterial species, and more work is envisaged to assess the photo catalytic disinfection process on other strains of bacteria and pathogenic organism, i.e. protozoa and viruses.

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