Disinfection of a farmstead roof harvested rainwater for potable purposes using an automated solar photocatalytic reactor

Nwoke Okala Okala$^{1,5}$*, Ezema Fabian Ifeanyichukwu$^2$, Chigor Nnamdigadi Vincent$^3$, Anoliefo Chukwudi Edward$^4$, Mbajiorgu Constantine Crowner$^5$

(1. National Centre for Energy Research and Development, University of Nigeria, Nsukka;
2. Department of Physics and Astronomy, University of Nigeria, Nsukka;
3. Department of Microbiology, University of Nigeria, Nsukka;
4. Department of Electronic Engineering, University of Nigeria, Nsukka;
5. Department of Agricultural and Bioresources Engineering, University of Nigeria, Nsukka)

Abstract: Access to safe and reliable water supply is a challenge in many parts of the developing world. The use of roof harvested rainwater for domestic and potable purposes is common in households and farmsteads in the rainforest and some parts of savannah ecological zones of Nigeria, in spite of the health risk associated with the drinking of water from an unimproved source such as untreated roof harvested rainwater. Solar photocatalysis has been proven to be effective in water treatment devoid of the shortcomings of high fuel cost and formation of carcinogens associated with conventional households water disinfection methods such as boiling and the use of chemicals. In this study, an automated photocatalytic batch reactor which uses solar radiation as its photon source was developed with materials sourced locally in Nigeria. The automated system injects contaminated water into the reactor tube and evacuates the water into the treated water tank upon the reception of a preset solar radiation dose. Roof harvested rainwater from a farmstead inoculated with \textit{Escherichia coli} (E.coli) was used in the evaluation. Results from the experiments show that the automated photocatalytic batch reactor was faster in bacteria inactivation than other reactors without TiO$_2$ insert. A solar radiation dose of 160 kJ L$^{-1}$ received on the photocatalytic reactor effectively and consistently inactivated \textit{E. coli} concentrations of $10^7 \pm (1.3 \times 10^6)$ CFU mL$^{-1}$ to concentrations below the detection limit of 4 CFU mL$^{-1}$ as well as prevent \textit{E. coli} regrowth after 24 h storage in the dark. The use of this system as a point of use water disinfection system could enhance access to potable water in remote places such as farmsteads.

Keywords: solar radiation dose, \textit{Escherichia coli}, water disinfection, TiO$_2$, photocatalytic reactor


1 Introduction

Water borne diseases are a major public health challenge especially in the rural areas of developing countries. The World Health Organisation (WHO) and the United Nations International Children’s Emergency Fund estimated that 663 million people worldwide, majority of whom live in developing countries use unimproved drinking water sources which include but not limited to unprotected wells, springs and surface water (WHO/UNICEF, 2015). Majority of evident water-related health problems are the result of microbial (bacteriological, viral, protozoan or other biological) contamination and are associated with ingestion of water contaminated with human or animal faeces (WHO, 2008).

Roof harvested rainwater is a major source of water for many households in the rainforest and some parts of savannah ecological zones of Nigeria. Nnaji and Mama (2014) reported that over eighty percent of water demand
of those living in bungalows in these zones could be met by rainwater. However, roof harvested rainwater quality often fall short of the microbiological standard set by regulatory bodies for potable water (Tobin et al., 2013; O’Hogain et al., 2012). At Adani, Enugu State of Nigeria, many farmsteads are involved in rice production with irrigation water sourced from a nearby river while roof harvested rainwater is used for domestic and potable purposes though the water quality is questionable. As shown in Figure 1, a typical rainwater harvesting system in the area consists of a roof catchment, a delivery structure, an above ground storage structure and an outlet. A filter is usually included within the delivery structure to minimize debris while disinfection is occasionally done after water has been collected from the storage structure.

![Figure 1](image.png)

Disinfection which is defined as the removal of harmful microbial contaminants (Backer, 2008) is conventionally implemented in many places including farmsteads in the developing world with methods such as boiling and the use of chemicals. Water boiling is hampered by the unavailability/high cost of fuels while formation of carcinogens is the shortcoming associated with the use of chemicals. Solar photocatalysis for water treatment could be a better alternative especially in places with abundant, all-year-round solar radiation because of the advantages of ‘solar photocatalysis for water treatment’ over the shortcomings of the conventional water disinfection methods.

Photocatalysis using a semi-conductor as applied in water disinfection technology is an Advanced Oxidation Process (AOP) in which low energy ultraviolet (UV- A (315- 400nm), UV-B (280-315 nm)) light impinge on a semi-conductor acting as catalyst in the presence of water to generate electron-hole pairs which facilitate the in situ production of highly reactive transitory species (i.e. H$_2$O$_2$, OH$,\cdot$, O$_3$) for mineralization of refractory organic compounds, water pathogens and disinfection by products (Ray, 2009). The UV spectrum of solar radiation has also been shown to have direct biocidal effects on microbial water contaminants (Choi and Choi, 2010; Hijnen et al., 2006).

Among the semiconductor catalysts (TiO$_2$, ZnO, Fe$_2$O$_3$, CdS, GaP and ZnS), titanium dioxide (TiO$_2$) has received the greatest interest in research and development of photocatalysis technology because TiO$_2$ is the most active photocatalyst under the photon energy of 300 nm$<\lambda<$390 nm in addition to its strong chemical and mechanical properties (Chong et al., 2010). The semiconductor catalysts can either be utilized in aqueous form or suspended on a solid substrate in batch systems and flow through systems. While utilization in aqueous form is more efficient because of the availability of more surface area of the catalysts, the requirement of post treatment retrieval of the catalyst makes such system expensive and complex to operate (McGuigan et al., 2012). On the other hand, suspension on a solid substrate removes the requirement for post recovery but limits the surface area of the catalyst available for reaction.

For applications involving the use of solar radiation for water disinfection, it is usually recommended to experimentally determine the minimum solar radiation dose (J L$^{-1}$) known as ‘lethal solar radiation dose’ which invariably is related to the exposure time to solar radiation for the inactivation of a given quantity of pathogen(s) in water with known physical and chemical properties (Polo-López et al., 2011). In SoDis (Solar water disinfection) applications, transparent bottles are usually filled with contaminated water and subsequently exposed to sunlight for some period of time. Meierhofer et al. (2002), recommends exposing the filled containers to full sunlight for six hours otherwise for two consecutive days if cloudiness exceeds 50% of the sunshine hours in the first day of exposure.

The aims of this study were (a) to determine the ‘lethal solar radiation dose’ for the inactivation of pathogen(s) in roof harvested rainwater in a farmstead using an automated batch process photocatalytic reactor and (b) to compare the rate of bacteria inactivation in the automated batch reactor to other SoDiS reactors.
2 Materials and Methods

2.1 Materials

The materials used in developing the automated batch photocatalytic reactor are: (a) Reactor tubes made of 64 cm in length borosilicate glass tube with internal and external diameters of 36 mm and 40 mm respectively (b) The substrate on which TiO$_2$ was deposited was a borosilicate glass rod of length 60 cm and diameter of 6.9 mm. The reactor tube and the glass rod were procured from Scientific Equipment Development Institute (SEDI) Enugu, Nigeria.(c) Car windshield washer pumps procured from an automobile repair shop in Nsukka, Nigeria were used for pumping water into and out of the reactor tube.(d) Titanium dioxide (Degussa P-25) procured from the Institute of Chemistry Education, University of Wisconsin–Madison, was used as the photocatalyst. (e) An Arduino® microprocessor procured from a local electronic components shop was used for the control of automation involving water pumping, solar irradiance sensing and logging, reactor temperature measurement and digital display of data on a Liquid Crystal Display (LCD) screen. (f) Three lengths (7 cm each) of copper wire were used as water level sensors in the reactor tube. (g) Solar irradiance sensing was done with a photovoltaic cell (5 cm by 5 cm) originally made for the charging of a hand-held solar rechargeable lantern. (h) Temperature sensor (waterproof LM 35) procured from a local shop, for sensing temperature of the reactor. (i) A liquid crystal display (LCD) screen for displaying instantaneous solar irradiance (W m$^{-2}$), accumulated radiation (J L$^{-1}$), duration of the disinfection process (seconds) and reactor temperature (°C). (j) A 2 GB memory card procured from a local shop for data storage. (k) A CPC reflector made from an aluminum roofing sheet procured from a local building materials shop (l) Two buckets with lids (one bucket for the contaminated water while the other is for the treated water). (m) 12 volts, 18 Ah battery for powering the system (n) signal and power cables (o) two 10 Wp PV modules for charging the battery bank. The schematic of the setup is shown in Figure 2 while the pictorial view of the automated batch-process solar water disinfection system is shown in Figure 3.

2.2 The water disinfection system set up

The borosilicate glass rod on which TiO$_2$ has been deposited is held in position at the centre of the glass tube with the aid of rubber corks at the two ends of the tube. The reactor tube with the glass rod is positioned at the focus of a compound parabolic concentrator (CPC) with aperture width of 10.7 cm and a concentration ratio of 1.6 with respect to the reactor tube diameter. The water pumps are activated by the signal from the reactor water level sensors. When the reactor tube is not full, current flow from a 5 V signal source to the ‘water level full’ sensor is truncated which in turn activates pumping of untreated water into the reactor tube as a result of the encoded programme in the Arduino® micro processor. Upon fullness of the reactor tube with contaminated water, current flow from the 5 V signal source to the ‘water level full’ sensor is restored which deactivates water pumping into the reactor tube. The outlet pump is activated when the reactor has received a given solar radiation dose (J L$^{-1}$) preset at the beginning of the experiment/disinfection process.

2.3 Solar radiation sensing

2.3.1 Calibration of an improvised solar irradiance sensing unit
The output voltage of a photovoltaic (PV) cell measuring 5 cm by 5 cm originally developed for a d.light® rechargeable lantern with product number “S2” was used for solar irradiance sensing and solar radiation logging. Preliminary investigation had shown that a plot of the output voltage due to solar radiation of the d.light® PV cell against time, is similar in shape to the graph of solar irradiance plotted against time for a Davis® made solarimeter in use at National Centre for Energy Research and Development, University of Nigeria, Nsukka. The output voltage from the d.light® PV cell was therefore amplified, calibrated and validated with different data sets from the Davis® made solarimeter. A statistical analysis using Student’s t-test showed no significant difference at $p=0.05$ for different data sets from a Davis® made solarimeter and the d.light PV cell output voltage after the necessary conditioning. A plot of the two data sets (400 to 1100 nm) as recorded on the 25th of August, 2016 is shown in Figure 4.

Figure 4 Comparison of solar irradiance measured with Davis® made solarimeter to the improvised sensing unit

2.3.2 Micro processor coding

According to Polo-López et al. (2011), the ‘lethal UVA dose’ can be calculated as:

$$Dose (J \text{ m}^{-2}) = \int \text{UVA (W m}^{-2}) \text{dt (s)} C$$

(1)

where, UVA is the solar UV irradiance incident upon the reactor; $dt$ is the exposure time (s); and $C$ is the concentration factor of the mirror. For the case where global solar irradiance is to be measured instead of UVA, and no CPC is incorporated, Equation (1) can be expressed as:

$$I (J \text{ m}^{-2}) = \int G (W \text{ m}^{-2}) \text{dt (s)}$$

(2)

where, $I$ is the global solar radiation ($J \text{ m}^{-2}$) received on the reactor for an exposure time (s) of $dt$ and for varying degrees of solar irradiance ($W \text{ m}^{-2}$) designated as $G$. If $I_n$ is used to represent radiation for a time period ($\Delta t$)$_n$ for which irradiance $G_n$ is constant, Equation (2) can be rewritten as:

$$I = G_1(\Delta t_1)+G_2(\Delta t_2)+G_3(\Delta t_3)+…+G_n(\Delta t_n)$$

(3)

The solar radiation dose (J L$^{-1}$) received on the reactor can be expressed as:

$$\text{Dose (Joules/Liter)} = I (J \text{ m}^{-2}) \times A (\text{m}^2)/V (\text{L})$$

(4)

where, $A$ is the rectangular cross sectional area of the reactor and $V$ is the volume of the reactor. The arduino micro processor was then programmed based on Equations (3) and (4), for the implementation of solar radiation logging as well as computation of the received solar radiation on the reactor. A preset value of solar radiation dose ($S_d$) is set at the beginning of each experiment such that when $S_d$ is equal to the dose computed from Equation (4), water is evacuated from the reactor into the treated water tank. Thereafter, fresh untreated water is pumped into the reactor from the untreated water tank. A water quality test on water samples taken from the water evacuated from the reactor for different values of $S_d$ would then show the optimal solar radiation for the disinfection of water from a given source with noted physical, chemical and microbial contamination levels.

2.4 Immobilization of TiO$_2$ unto the substrate

The borosilicate glass rod surface used as the substrate for TiO$_2$ deposition was roughened with sandpaper to improve adhesion of the TiO$_2$ unto its surface given that earlier trials without roughening were not very successful. The rod was then washed with a 5% detergent solution and rinsed with tap water. The washing and rinsing were repeated five times after which the rod was rinsed with distilled water. The glass rod was then sonicated in a distilled water bath for 15 mins. The sonication process was repeated three times. The rod was then dried in an oven at 105°C for 1 h before being weighed. A 1% solution of Degussa P-25 TiO$_2$ was prepared with distilled water and sonicated for 15 min. The rod was swirled in the TiO$_2$ solution until coated. The swirling of the glass rod in the TiO$_2$ solution and drying was repeated 10 times for adequate coating. The
rod was then annealed at 300°C for 10 h. The initial (before TiO₂ deposition) and final weight (after TiO₂ deposition and annealing) of the rod were 51.701 and 51.744 g respectively. Given the effective volume of the reactor as 0.6 L, the concentration of the TiO₂ in the reactor was approximately 0.07 gL⁻¹.

2.5 Rainwater collection

The rainwater used in this experiment was sourced from an above-ground tank measuring 4546 litres in volume and made of Polyvinly Chloride (PVC) from a rainwater harvesting system situated in a farmstead at Adani (6.73 °N, 6.99 °E), Enugu State, Nigeria. The farm measures approximately 15 hectares and cultivates rice. The roof catchment of the rainwater harvesting system is made of corrugated sheet metal (aluminium) and measures 9.14 by 7.31 m. A single batch of rainwater (approximately 50 L) was collected from the rainwater storage to ensure that the same quality of water was used for all experiments. The quality parameters of the water are shown in Table 1. To maintain the chemical quality of the water, the water was not autoclaved before each experiment. Turbidity was measured with a turbidity meter (AL 250T-IR) while temperature was measured with a mercury in glass thermometer. Dissolved oxygen (DO), Electrical Conductivity (EC) and pH were measured with YSI EcoSense® (DO 200), PCE® (CM 41) and Vantakool® digital pH meter respectively. Microbial parameters were determined with standard direct plate count methods with the appropriate medium.

Table 1 Physiochemical and microbial quality of the rainwater

<table>
<thead>
<tr>
<th>Physiochemical parameters</th>
<th>Unit</th>
<th>Value</th>
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</tr>
<tr>
<td>Odour</td>
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<td>Dissolved Oxygen (DO)</td>
<td>Mg L⁻¹</td>
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</tr>
<tr>
<td>Electrical Conductivity (EC)</td>
<td>μS cm⁻¹</td>
<td>230</td>
</tr>
</tbody>
</table>

Microbial parameters

| Total Coliform (TC)             | CFU/100 mL | 920   |
| Fecal Coliform (FC)             | CFU/100 mL | 412   |
| Heterotrophic Plate counts (HPC)| CFU/mL     | 1890  |

2.6 Bacterial preparation and enumeration

Escherichia coli was used as the model bacterium in this study due to its widespread use as a faecal indicator. A single colony of E. coli (K – 12 (ATCC 23631)) taken from refrigerated stock was sub-cultured in 100 mL of sterile nutrient broth (TITAN TM 350) and incubated for 18 h at 37°C. The cells were harvested by centrifugation at 3000 rpm for 10 min and washed with sterile normal saline. The bacterial pellet was then resuspended in the 10 L of the rainwater to give an initial bacteria concentration of 10³±(1.4×10⁶) Coliform Forming Units per millilitre (CFU mL⁻¹) and was used as the contaminated water in the reactors. Water samples were taken from the “untreated water tank” as well as the other batch reactors for enumeration before the experiments. Water samples were also taken from the automated batch reactor and the other reactors during the experiment and from the “treated water tank” of the automated system at the end of each experiment. Using spread plate method and appropriate dilutions, enumeration were done on the water samples plated in triplicate on standard plate count agar plate (CM 463; Oxoid, UK). Colonies were counted after incubation at 37°C for 18 h. The mean of the results were plotted with error bars indicating standard deviation (SD) of the results.

2.7 Construction of Compound Parabolic Collecto (CPC)

A CPC profile generated with ray tracing technique by Márquez et al. (2010) presented in Figure 5 was normalized to a scale of 1:1 and traced on ten blocks of wood. Following the pencil trace of the CPC shape on the blocks of wood, a wood band saw was used to cut the shapes. The cut blocks of wood were arranged side by side on a
wooden plank and held onto the plank with screws fastened through holes drilled by a 3 mm diameter drill bit such that the arranged blocks of wood formed the female side of a mold for shaping the aluminium sheet into a CPC. This arranged blocks of wood is shown in Figure 6.

**Figure 6** Female side of a mold for shaping the aluminium sheet into a CPC

A commercially available aluminium roofing sheet (uncorrugated) with a thickness of 0.8 mm was then cut into sizes of 21 cm by 60 cm and forced into the mold with the aid of the male part of the mold. Finally the CPC formed with the aluminium reflector was held in position with screws fastened at its two rear ends onto two blocks of wood of the same dimension and shape with the ones used as the female side of the mold.

### 2.8 The experiment

For each experimental run, the automated sequential batch reactor designated as T₁ (Figure 3) was set up alongside four other batch reactors. These reactors presented in Figure 7 were; (a) a reactor tube of the same dimension as the reactor in automated batch process, fitted with a CPC but does not contain TiO₂ designated as T₂, (b) a reactor tube of the same dimension as the automated batch process reactor but is not fitted with a CPC and does not contain TiO₂ designated as T₃, (c) a bottle made of transparent polyethylene terephthalate (PET) which does not contain TiO₂ and is not fitted with a CPC designated as T₄, (d) a reactor tube of the same dimension as the automated sequential process reactor kept in the dark as control, designated as T₅. All the reactors except T₅ were aligned in the East-West axis, and inclined at 22° (local latitude plus 15°) to the horizontal facing South. On the experimental days, the solar radiation sensor at NCERD weather station (located ≈ 30 m from experimental site) was inclined at 22° to the horizontal and faced South.

### 3 Results and Discussion

#### 3.1 Bacteria inactivation results

The results of the experiment performed on 24th, 28th and 31st of July, 2017 for 120, 160 and 160 kJ L⁻¹ of solar radiation dose respectively are presented in Figure 8, Figure 9 and Figure 10.
3.2 Discussion

3.2.1 Rate of bacteria inactivation between the photocatalytic process and the other batch reactors

The automated photocatalytic batch reactor was consistently faster than the other batch reactors in the inactivation of *E. coli*. This result is consistent with reports in literature (Rizzo et al., 2014; Dunlop et al., 2011; Méndez-Hermida et al., 2007) Arranged from the fastest in *E. coli* inactivation to the slowest are; the automated photocatalytic batch reactor (*T*₁), the PET bottle (*T*₄), the batch reactor with a CPC reflector but without TiO₂ insert (*T*₂), the batch reactor without a CPC reflector and TiO₂ insert (*T*₃), the control reactor (*T*₅). The PET bottle was faster than the other batch reactors without TiO₂ insert exposed to solar radiation in the inactivation of *E. coli* though it had a bigger water volume (≈0.7 L) and smaller solar collector area (6.5 cm × 19 cm = 124 cm²) compared to the other reactors with 0.6 L volume each and collector area of 240 cm² (60 cm × 4 cm). It could have resulted from the relatively smaller wall thickness of the PET bottle (≈0.2 mm) compared to the other batch reactors made of borosilicate glass with 4 mm wall thickness. This could have made it easier for the penetration of the UV band of the solar radiation spectrum into the PET bottle than the other batch reactors. The photocatalytic reactor was faster than the PET bottle in *E. coli* inactivation because of the TiO₂ insert.

3.2.2 Solar radiation dose for bacteria inactivation in the photocatalytic reactor

Consistent solar radiation doses were observed for bacteria inactivation to concentrations below the detection limit of 4 CFU mL⁻¹ for the water exposed to solar radiation in the automated photocatalytic reactor. From the results presented above, 120 kJ L⁻¹ of solar radiation was able to effect approximately 7-log reduction in *E. coli* concentration in the water in the automated photocatalytic reactor for all the experiments carried out on sunny days as can be seen in the water samples collected at; 11:43 am on the 24th of July, 2017 (Figure 8), and 12 pm on the 31st of July, 2017 (Figure 10). For the experiments conducted on a cloudy day (Figure 9), a higher dose of solar radiation was required to effect the same level of inactivation of the *E. coli* in the photocatalytic reactor. As presented in Figure 9 which was not a sunny day, 120 kJ L⁻¹ of solar radiation could only effect 4-log reduction in the *E. coli* concentration of the water in the automated photocatalytic reactor. Estimation of the solar radiation dose received on the photocatalytic reactor for the water sampled at 3 pm on the 28th of July (Figure 9) was based on solar radiation data from the Davis® made solarimeter at the weather station of the National Centre for Energy Research and Development, which indicated that approximately 3.5 MJ m⁻² was received in the time period (10 am to 3 pm) which using Equation 4 is equivalent to 140 kJ L⁻¹ of solar radiation dose. This method of estimating solar radiation dose was always used when the water sample under discussion was not collected at the end of the experiment. This suggests that on sunny days, a lower solar radiation dose is required for bacteria inactivation in the photocatalytic reactor than on a cloudy day.

3.2.3 Solar radiation dose for *E. coli* regrowth prevention

There were bacteria regrowth in the water samples in which *E. coli* concentration had been reduced to a concentration below the detection limit of 4 CFU mL⁻¹ with the receipt of approximately 120 kJ L⁻¹. As shown in Figure 11, the samples marked; 24/07/2017, 28/07/2017(a), 31/07/2017(a) had bacteria regrowth though *E. coli* reduction to a concentration below the detection limit had been reached with the receipt of 120 kJ L⁻¹ of solar radiation on the automated reactor. Also from Figure 11, the sample marked 28/07/2017(a) had regrowth though it had attained *E. coli* reduction to a level below the detection limit at 3 pm (Figure 9) with the
receipt of approximately 140 kJ L\(^{-1}\) of solar radiation dose. However, the last sample collected at the end of the experiment from the automated photocatalytic reactor on the 28/07/2017 at 3:11 pm (Figure 9) which had received 160 kJ L\(^{-1}\) of solar radiation had no bacteria regrowth after 24 hrs storage in the dark. The samples which had no regrowth after the 24 hrs storage in the dark are marked; 28/07/2017(b), 31/07/2017(b), 31/07/2017(c) in Figure 11 and had received approximately; 160 kJ L\(^{-1}\), 140 kJ L\(^{-1}\) and 160 kJ L\(^{-1}\) of solar radiation. From these results, a solar radiation dose of 160 kJ L\(^{-1}\) received on the photocatalytic reactor was observed to effectively inactivate \(E.\ coli\) concentration of approximately \(10^7\) CFU mL\(^{-1}\) to a level below the detection limit as well as prevent \(E.\ coli\) regrowth after 24 hrs storage in the dark irrespective of the experimental period being cloudy or sunny.

![Figure 11 Chart of \(E.\ coli\) regrowth](chart.png)

Thermal bacteria inactivation did not seem to have played a synergistic role in the entire inactivation process for the reactors. The maximum water temperature recorded was 41°C in the PET bottle while the maximum temperature recorded in the other borosilicate glass made reactors was 39°C. Therefore the synergy between optical and thermal inactivation process which was reported to have effect at approximately 45°C (Navntoft et al. 2008) could not have been responsible for the bacteria inactivation in the reactors for the entire experiment though sub-lethal effects due to the increased temperature could have contributed.

The aluminium reflector used as CPC did not enhance the bacteria inactivation considerably. From the results presented from Figure 8 to Figure 10, there didn’t seem to be a significant difference between the rate of bacteria inactivation in the batch reactor with a CPC reflector but without TiO\(_2\) insert (T\(_2\)) and the batch reactor without a CPC reflector (T\(_3\)). This could have been due to the use of a commercially and locally available grade of aluminium sheet which was not anodised and electropolished. Hence, the use of an anodised and electro polished aluminium surface as CPC as reported by Tanveerand Tezeanli Guyer (2013) could have produced better bacteria inactivation rate.

4 Conclusion

Roof harvested rainwater from a farmstead, inoculated with \(E.\ coli\) was successfully disinfected with a locally developed automated solar photocatalytic reactor. The reactor was faster in bacteria inactivation than other batch reactors tested in this study. A solar radiation dose of 160 kJ L\(^{-1}\) was effective for the inactivation as well as regrowth prevention of \(E.\ coli\) concentration of \(10^7±(1.3×10^6)\) CFU mL\(^{-1}\) in water exposed to solar radiation in the automated solar photocatalytic reactor. On a sunny day at the site of the experiment, this solar radiation dose could be received in about 3 h of sunshine.

Acknowledgements

The authors are grateful to Dr. Chidinma Ezema (Nee Okpara) (Energy Research Centre, University of Nigeria, Nsukka (UNN)) for providing the TiO\(_2\) (Degussa P25) used in this study and Mr. Offia Solomon (Energy Research Centre, UNN) for assistance during immobilization of the TiO\(_2\) on the substrate. We are also grateful to Mr. Ngene Augustine Arinze of the Faculty of Veterinary Medicine (UNN) for the assistance offered on bacteria growth and enumeration. We also appreciate the management of the National Centre for Energy Research and Development, UNN, for access to laboratory equipment.

References


