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Material integrity of LDPE-based solar water disinfection reactors with improved usability

Supamas Danwittayakul, Supachai Songngam, Tipawan Fhulua, Panida Muangkasem, Phitchaya Muensri, Sittha Sukkasi*

MTEC, National Science and Technology Development Agency, 114 Thailand Science Park, Klong1, Klong Luang, Pathumthani, 12120, Thailand, Tel. +6625646500, Ext. 4261, email: supamasd@mtec.or.th (S. Danwittayakul), Tel. +6625646500, Ext. 4369, email: supachas@mtec.or.th (S. Songngam), Tel. +6625646500, Ext. 4370, email: tipawanf@mtec.or.th (T. Fhulua), Tel. +6625646500, Ext. 4370, email: panidm@mtec.or.th (P. Muangkasem), Tel. +6625646500, Ext. 4371, email: phitcham@mtec.or.th (P. Muensri), Tel. +6625646500, Ext. 4766, email: sitthas@mtec.or.th (S. Sukkasi)

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ABSTRACT

This work investigated the changes in material integrity of solar water disinfection (SODIS) reactors, which had been developed to overcome usability-related short comings of previous designs, for usages in rural areas. During a 12-week period of usage of the new reactors in actual environments, the degradation of the materials was investigated weekly, with respect to surface morphologies, compositions, optical transmissions, tensile strengths, and leaching of organic compounds from the reactors into the treated water. The results showed that there were no scratches on the low-density polyethylene (LDPE) bags (the water-containing components of the reactors), which were problematic in the previous design. The maximum reduction of tensile strength was 28.5%. The optical transmittances in the UVA region of the LDPE and PVC components of the reactors decreased by 11% and 53%, respectively. The composition results indicated that the LDPE bags photo-degraded via an oxidation reaction. 2,4 di-tert-butyl phenol, a known degradation product from antioxidants of polyethylene, was found in some samples of treated water, but at levels close to the method of detection's limit as well as those in the control samples. In addition, the optical transmittances in the UVA region of the LDPE and PVC components of the reactors decreased by 11% and 53%, respectively. While the LDPE bags could be used for up to 12 weeks of SODIS without too much reduction in UVA transmittance, the PVC boxes should be replaced after 4 weeks, or at most 8 weeks.

Keywords: Point-of-use water treatment; LDPE durability; SODIS reactor; Degradation

1. Introduction

The lack of safe drinking water is a serious issue that affects more than 700 million people worldwide, and many people, especially those in developing countries in Africa, South America, and Asia, have to rely on unsafe water that contains pathogenic microorganisms [1,2]. Drinking water contaminated with pathogens leads to a potential risk of water borne diseases, such as cholera, dysentery, and diarrheal diseases [3]. In developing countries, about 50% of the population are exposed to contaminated water sources with inadequate hygiene and poor sanitation, leading to approximately 4 billion cases of diarrhea each year [4]. It has been reported that 0.6 million deaths of children under the age of 5 were a result of diarrhea in 2012 [5]. The most common household water treatment is boiling, which can result in about 35–44% reduction in diarrheal diseases; however, high fuel costs and indoor air pollution can make boiling impractical for many households [6].

Solar water disinfection (SODIS) is a point-of-use water treatment process that has been recommended by the WHO, UNICEF, and Red Cross as an appropriate method for disinfecting drinking water in developing areas, due to its simplicity, affordability, and treatment capability [7–9].

*Corresponding author.

To disinfect water with SODIS, the user fills the water into a transparent container and exposes it to the sun for 6–8 h (or approximately one day) on a sunny day, or 2 consecutive days if the sky is cloudy [10]. During the SODIS process, pathogens in the water are inactivated by ultraviolet (UV) radiation and heat from the sunlight, by optical and thermal inactivation mechanisms, respectively [11]. Many peer-reviewed papers have since been published on SODIS [12].

The most commonly used containers for SODIS are polyethylene terephthalate (PET) bottles, which are easy to find but have several drawbacks. The shapes of the bottles make them bulky to transportation in a large quantity. The circular cross-section of the bottles makes the effective ratio between surface area exposed to the sun and the volume of water being treated low, leading to long SODIS treatment time required. Moreover, PET itself is brittle, allows nearly zero UVB transmittance, and can become easily scratched [13].

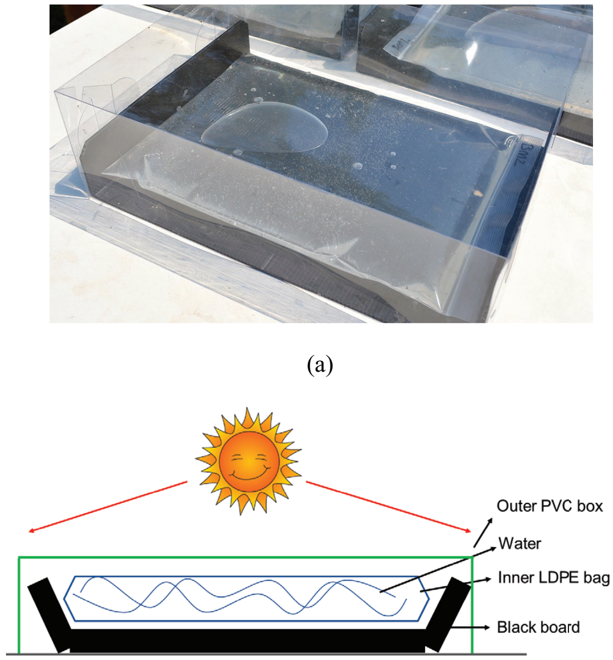
Reactor design is one approach to enhance the disinfection efficiency of SODIS. Low-density polyethylene (LDPE) bags have been used as SODIS containers in previous work [13–15]. Polyethylene (PE) allows high UVB transmittance [13]. The bags can also achieve high radiation-area-to-water-volume ratio, and they can be shipped in a large quantity more efficiently. Another LDPE-bag-based SODIS reactor design used two nesting layers of LDPE bags: the inner layer containing the water to be disinfected and the outer one creating air-insulation to minimize heat loss from the water to the surroundings. The design also included a black board inserted in between the two LDPE bags, on the bottom side, to absorb additional solar radiation and convert it into heat. Although this particular SODIS reactor design overcame the drawbacks of PET-bottle SODIS reactor design, there were still some weaknesses, such as the difficulty of keeping the outer-layer bag inflated to create air insulation. A new SODIS reactor design was subsequently developed, in which an LDPE bag for containing water to be treated was placed on a black plastic board and a clear PVC box covered the entire set. The black plastic board helped convert additional solar radiation into heat, which assists the water disinfection, and the clear PVC box helped create air insulation, which maintained the elevated water temperature. This new design achieved high radiation-area-to-water-volume ratio and high water temperature, while making the setting up operation easy for the users.

In this study, the durability of the LDPE bags and PVC boxes of the new design, as used in actual SODIS conditions for up to 12 weeks, was investigated. The changes in the morphological, physical, and optical properties of the LDPE bags were monitored and characterized as a function of the SODIS usage time. Moreover, the water that was treated in the SODIS containers collected from both SODIS reactors was tested for trace organic contaminants, which could have leached from the LDPE bags into the water over the course of the usage.

2. Materials and methods

2.1. Materials

The SODIS containers had the above mentioned latest design (Fig. 1). The LDPE bag for containing water to be treated had flat dimensions of 20 cm × 30 cm. The PVC box was 25-cm wide, 35-cm long, and 10-cm high. The materials were commercial grades and procured from local suppliers in Thailand. The standards of 2-napthol, 4-ethyl phenol, 4-tert-butyl phenol, 2,6-di-tert-butyl-p-benzoquinone, 2,4-di-tert-butyl phenol, and chloroform were chromatography grade and supplied by Merck and Fluka. All chemicals were used without any purification.



(b)

Fig. 1. (a) SODIS reactor with the new LDPE-bag-based design and (b) schematics of the cross section of the reactor.

2.2. Solar water disinfection process

The SODIS experiments were conducted in Sangkhlaburi district, Kanchanaburi province, Thailand (latitude: 15°10’52.4”N, longitude: 98°19’29.9”E). Each SODIS container was filled with 1500 mL of water, which was procured from a nearby natural source, called Huay Ta Kok, a primary source of drinking water for the local people. The properties of the water are shown in Table 1. The SODIS container with water was exposed to natural solar radiation for 6 h daily. The process was continuously repeated for up to 12 weeks (January–March 2015), in three replicates. At the end of each week, the LDPE and PVC from three SODIS reactors and the water that had been treated in the reactors were collected. The LDPE and PVC samples were then characterized to examine the degradation of the materials, while the treated water samples were analyzed to assess the possible migration of organic components from the bags, and the untreated water samples from the source were analyzed as control samples.

Table 1
The quality of water procured from a local natural river (Huay Ta Kok) as compared to the World Health Organization (WHO) guideline

	WHO guideline	Water sample (Feb. 7, 2014)
pH	–	7.41
Color (Pt-Co)	<15	7.78
Turbidity (NTU)	<5	8.09
Total solids (mg/L)	<1000	133.5
Total hardness (mg/L)	–	87.28
Fe (mg/L)	<0.3	0.1333
Mn (mg/L)	<0.1	0.0108
Cu (mg/L)	<1.0	ND
Zn (mg/L)	<3	0.0066
Pb (mg/L)	<0.01	ND
Cr (mg/L)	<0.05	ND
Cd (mg/L)	<0.003	ND
As (mg/L)	<0.01	<0.0015
Hg (mg/L)	<0.001	<0.0005
Ba (mg/L)	<0.7	0.0353
Se (mg/L)	<0.01	ND
Al (mg/L)	<0.2	0.0604
Ni (mg/L)	<0.02	–
CN^- (mg/L)	<0.07	ND
SO_4^{2-} (mg/L)	<250	ND
Cl^- (mg/L)	<250	ND
NO_3^- (mg/L)	<50	0.12
F^- (mg/L)	<1.5	ND
Bacteria (Coliform) (MPN/100mg)	ND	>23
E. coli (MPN/100mg)	ND	Detected

2.3. Study of the degradation of the plastic materials of the SODIS reactor

The LDPE and PVC samples obtained at the end of each SODIS exposure week were first cleaned by nano pure water (>18.2 MΩ water), then dried with nitrogen gas to remove surface moisture. The plastic films were cut into specific dimensions for various analytical techniques to investigate the degradation mechanisms. Infrared spectra of before- and after-SODIS plastic samples were obtained by Fourier transform infrared (FTIR) spectroscope (Shimadzu, IR Prestige-21 with ATR mode), while a UV-VIS spectrophotometer, (Shimadzu, SolidSpec-3700) was used to examine the optical transmission spectra of the samples. The tensile-strength tests of the plastic samples were carried out following ASTM D638-14 [16] using a universal testing machine (Instron 55R4502) at a cross head speed of 500 mm·min^{-1} for LDPE and of 50 mm·min^{-1} for PVC. For each test, 5 replicate samples were measured and the average values were reported. The surface morphologies of before- and after-SODIS samples were investigated using a field emission scanning electron microscope (FESEM) (Hitachi, SU-8030).

2.4. Study of the leaching of organic contaminants from LDPE bags into the treated water

A gas chromatograph with a mass spectrometry detector (GC-MS) was used to investigate potential migration of organic compounds from the LDPE bags into the water based on the analytical method suggested by Brocca et al. [17]. Each water sample was first extracted by liquid-liquid extraction. One liter of each water sample was added with 100 µl of 50 mg·L^{-1} 2-naphthol, which was used as a surrogate prior to the extraction process. The water sample with surrogate compound was then transferred to a separating funnel and 15 mL of chloroform was then added. The separating funnel was shaken for 5 min to allow organic compounds to move to the chloroform phase. The separating funnel was then kept standing for 30 min. The organic phase at the bottom layer was transferred and stored in a glass bottle. This extraction procedure was then repeated for several times to ensure that all organic compounds were extracted into the chloroform. The organic phase collected in a glass bottle was then gently evaporated by flowing nitrogen gas and finally made up 5 mL of the final volume. The organic compounds in the extracted solution were then analyzed by GC-MS (Shimadzu, QP2010) with a capillary column of RTX-5MS, 30 m, 0.25 mm i.d., 0.25 µm film thickness. The carrier gas was helium with a constant flow of 1.2 mL·min^{-1}. 1 µl of the solution was injected, in the splitless mode, into the injection port at the temperature of 250°C. The temperature was held at 50°C for 1 min and raised at a rate of 5°C/min, then held for 10 min. The MS scan range was from 20 to 650 m/z.

3. Results and discussion

3.1. Tensile-strength analysis

The tensile strength of the starting LDPE bags, before being used for SODIS, was measured at 17.6 MPa. The tensile strength remained stable for the first 4 weeks of SODIS exposure, decreased from 16.7 to 13.8 MPa between weeks 4 and 5, and then remained relatively stable until the end of the 12-week period. The total percentage reduction in tensile strength of the LDPE bags after the 12 weeks was 28.5%. This results indicate that the LDPE bags could be used in actual SODIS conditions for up to 12 weeks without any significant damages while maintaining acceptable strength.

The tensile strength of the PVC box before being used for SODIS was 72.5 MPa. It rapidly decreased after only one week of SODIS exposure to 60.4 MPa, but after that it remained relatively stable until week 12. The total reduction of tensile strength after 12 weeks of SODIS exposure was 16.6%. The PVC boxes, like the LDPE bags, maintained sufficient durability for SODIS usages over the 12-week period.

3.2. FTIR analysis

Fig. 3a–b show the Fourier transform-infrared (FT-IR) absorption spectra of the LDPE and PVC plastic materials as a function of the SODIS exposure times. The absorbance peaks of the LDPE bags at 1020 cm^{-1} of Fig. 3a were C–O stretching peaks, and they tended to increase with longer SODIS exposure, as the peak of W12 was higher than those of W9, W6, and W3. The peak was not found in the W0

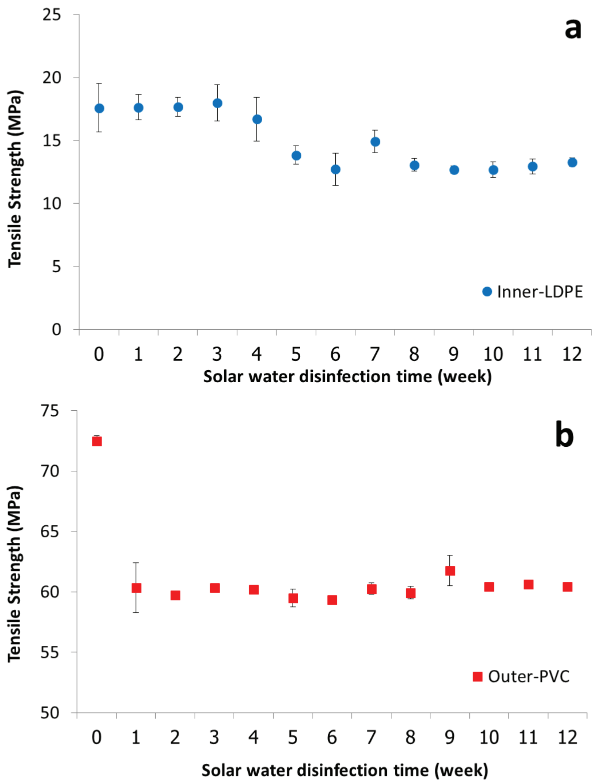


Fig. 2. Tensile strengths of (a) LDPE and (b) PVC materials after 1–12 weeks of SODIS exposure.

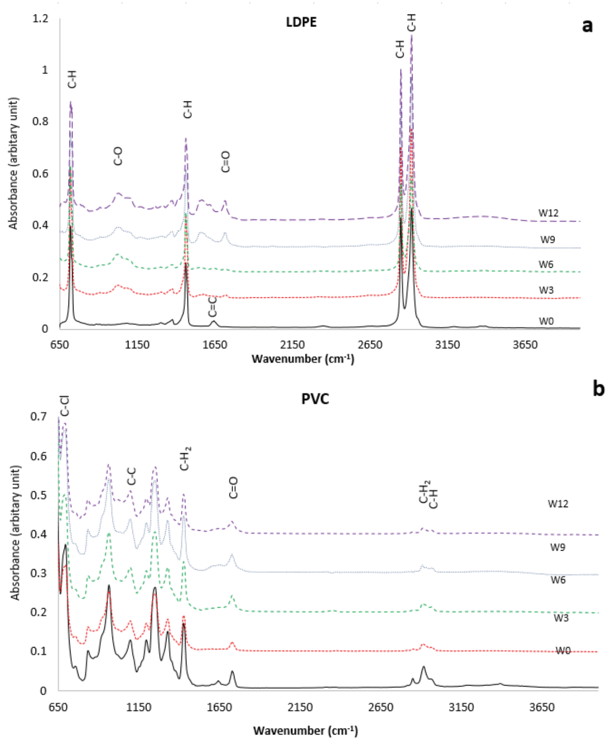


Fig. 3. FTIR spectra of the LDPE and PVC plastic materials after increasing SODIS exposure periods: (a) LDPE and (b) PVC.

LDPE samples (before SODIS exposure). This indicated that the photo-oxidation reactions took place. The photo-oxidation by-product was also confirmed by the observation of a carbonyl group of C=O peak at 1724 cm^{-1} of LDPE samples, even after the SODIS exposure of only 3 weeks (Fig. 3a) [18–23]. The absorption peaks at 1643 cm^{-1} of LDPE samples corresponded to unsaturated hydrocarbons or aromatics (–C=C– stretching peaks) [24,25], which were part of the UV protective additives of commercial LDPE that disintegrated after three weeks of SODIS exposure (Fig. 3a). This could be attributed to the release of the additive into the surroundings by an oxidation reaction. Phenolic compounds containing C=C such as Irgafos 168® and Irganox 1010 are usually added in polyethylene as antioxidants or UV-shield additives, in order to protect the film from UV irradiation [26–28]. These FTIR results suggested leaching of compounds from the LDPE occurred. The leaching was further investigated and reported in the section of analysis of organic compounds migration by GC-MS.

Fig. 3b shows the Fourier transform-infrared (FT-IR) absorption spectra of the outer PVC samples as a function of SODIS exposure time. The absorption peaks of C=O carbonyl group of PVC at 1730 cm^{-1} increased very slowly over the SODIS exposure periods, indicating gradual degradation that leaves ester by-product [29].

The plots of FTIR intensity ratios of C=O group as a function of SODIS exposure time of the LDPE bags are shown in Fig. 4a. An increase in carbonyl group (C=O) could be observed in the LDPE bags due to the photo-oxidation reaction as mentioned earlier. The results corresponded to the increasing peak intensity ratios of C–O in the LDPE samples that were also due to the oxidation reaction. The intensity ratio of C=C peaks at 1643 cm^{-1} of LDPE decreased over time possibly because the additives were released from the bags and migrated into the water. An increase in C=O peaks of the PVC samples could be observed in Fig. 4d, indicating that the PVC samples also gradually produced the C=O group, which was caused by the photo-oxidation degradation during the SODIS exposure. However, the increase in C=O intensity of the PVC samples did not significantly affect the tensile strengths of the samples, as reported in the tensile strength analysis.

3.3. Analysis of organic compounds migration by GC-MS

Following the FTIR results that showed the evidences of degradation of the LDPE bags, as discussed above, the possible leaching of organic compounds into the treated water was investigated by GC-MS. A previous study showed that 4-ethyl phenol, 4-tert-butyl phenol, 2,6-di-tert-butyl-p-benzoquinone, and 2,4-di-tert-butyl phenol could migrate from PE [17], so the presence of these compounds in the water treated by SODIS was investigated. The results of the GC-MS analysis of the treated water samples showed that 4-ethyl phenol, 4-tert-butyl phenol, and 2,6-di-tert-butyl-p-benzoquinone were not found in any of the samples. Only 2,4-di-tert-butyl phenol was found, in the concentrations ranging from 1–3 $\mu g \cdot L^{-1}$ in the SODIS-treated water. Since 2,4-di-tert-butyl phenol is a known degradation byproduct from antioxidants such as Irgafos 168® of polyethylene [26], this result confirmed the FTIR and UV-VIS results that the additive chemicals with C=C bonds in the

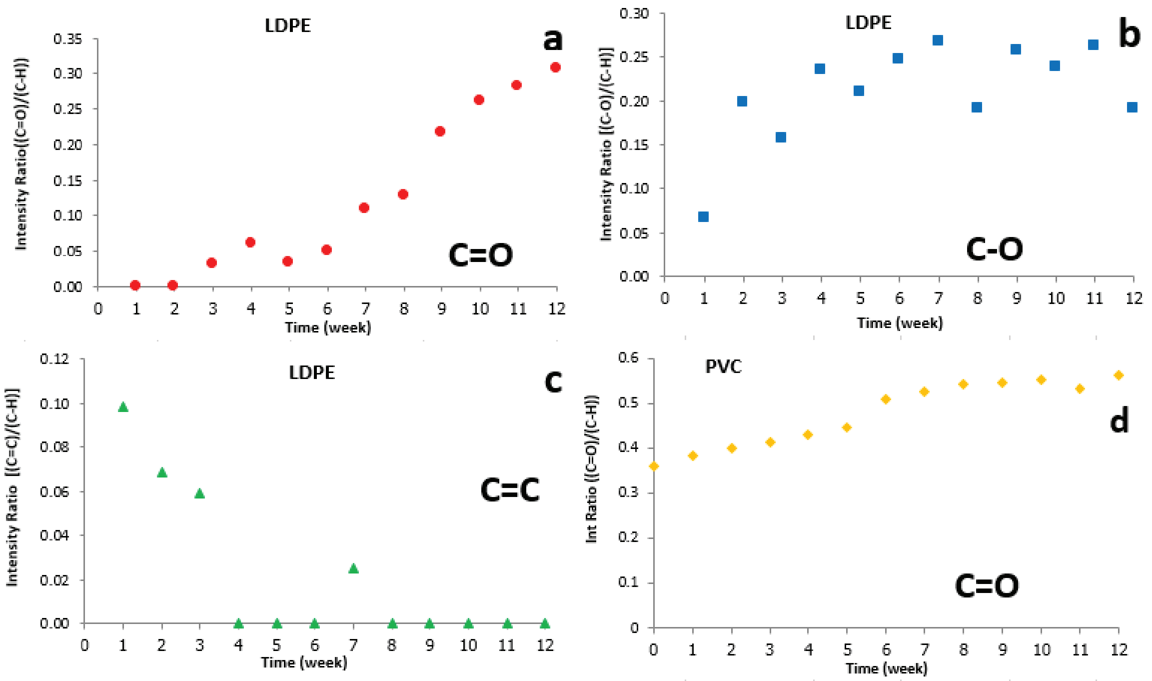


Fig. 4. Plots of FTIR peak intensity ratios of the inner LDPE and outer PVC materials against SODIS exposure time: a) C=O peak/C–H peak of the LDPE, b) C=C peak/C–H peak of the LDPE, c) C–O peak/C–H peak of the LDPE, d) C=O peak/C–H peak of the PVC.

polyethylene films may have been leached from the LDPE bags. However, the levels of the 2,4-di-tert-butyl phenol in the SODIS water samples were in the range of the method detection limit (1 $\mu g \cdot L^{-1}$) and very close to the concentration levels that were also found in the procedural blanks (the control samples that were not SODIS-treated). Also, the compounds were only detected in the samples from Week 2.

3.4. SEM analysis

The SEM micrographs of the LDPE bags are shown in Fig. 5. The surfaces of the before-SODIS samples (Fig. 5a) were very smooth, while those of the LDPE bags after the SODIS exposure exhibited multiple traces of tiny particles, which indicated that the bags had been in contact with contaminants in the water, and the dirtiness increased with longer SODIS exposure (Fig. 5b–c). Fig. 6a–c show SEM micrographs of the PVC boxes after different SODIS exposure periods. It can be observed that the before-SODIS PVC samples had smooth and dense surfaces (Fig. 6a), while the PVC samples after 6 and 12 weeks of SODIS exposures (Fig. 6b and 6c) exhibited rougher surfaces, indicating that the surface structure of PVC changed or slightly degraded, as also indicated by the results of the tensile strength tests.

3.5. UV-VIS transmittance analysis

The optical transmittance spectra of the LDPE and PVC samples are shown in Fig. 7. It can be observed that the before-SODIS LDPE samples (W0 of Fig. 7a) exhibited a strong absorption at 270 nm, since the transmittance plot dipped in that region. However, that absorption dip at 270 nm disappeared in the transmittance spectra of the samples that had undergone the SODIS usage, even after 3 weeks. This phenomenon could be attributed to the loss of the additives, which would normally shield the LDPE bags from the UV, during the SODIS process, due to the UV exposure and turbulence from water during the filling of the bags [30]. These results correspond to the results obtained from FTIR, which showed the decrease of C=C (1643 cm^{-1}) absorption band (Fig. 5a) [31].

The optical transmission spectra of the outer PVC boxes of the SODIS reactor after 3, 6, 9 and 12 weeks of SODIS exposure as compared to the control sample (W0) are shown in Fig. 7b. It can be observed that the control PVC sample showed strong transmittance at 300 nm in the UVB region, and the transmittances of the PVC samples gradually decreased after longer solar exposures.

The changes in the transmittance of the plastic samples in the SODIS reactor were calculated for the ultraviolet range, from 250 to 400 nm, divided into UVA, UVB, and UVC regions (Table 2) by using the average of the transmittance values at of the lowest and highest wavelengths in each UV radiation region as a representative value. During SODIS, solar radiation is utilized to make microbiologically contaminated water safe for drinking. Of the UV radiation that reaches the earth's surface, about 95% is UVA radiation (320–400 nm) and the small remainder is UVB (290–320 nm), which can also cleave the C–C bonds of polyethylene [19,32]. From the results of this study, it can be observed that the highest percentage reduction of the transmittance at UVB of LDPE bags after 12 weeks of SODIS exposure was 16.3%,while in the UVA region (320–400 nm) the reduction was only 10.7%. In the UVC region (250–290 nm), the percentage reduction of the transmittance of LDPE bags was

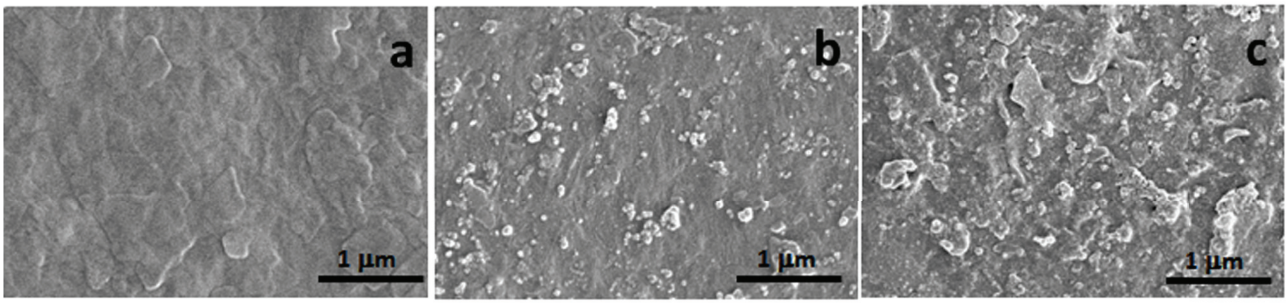


Fig. 5. SEM micrographs of LDPE bags after SODIS exposures: (a) 0 week, (b) 6 weeks and (c) 12 weeks.

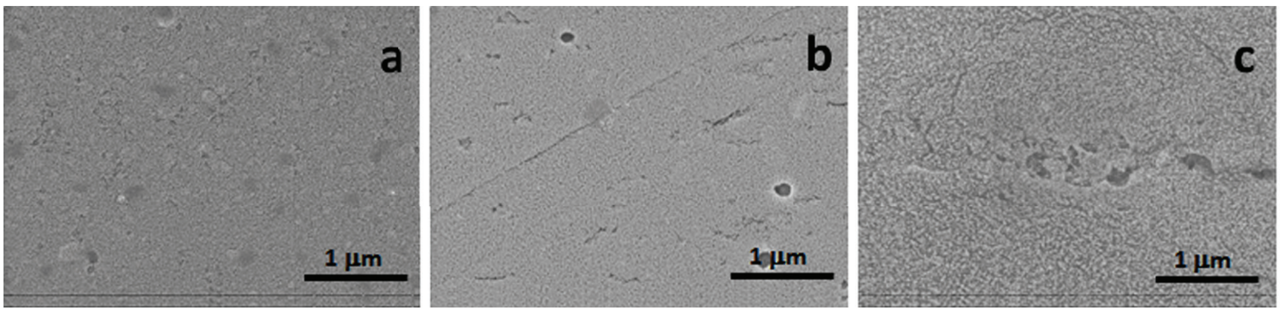


Fig. 6. SEM micrographs of PVC boxes after SODIS exposures: (a) 0 week, (b) 6 weeks and (c) 12 weeks.

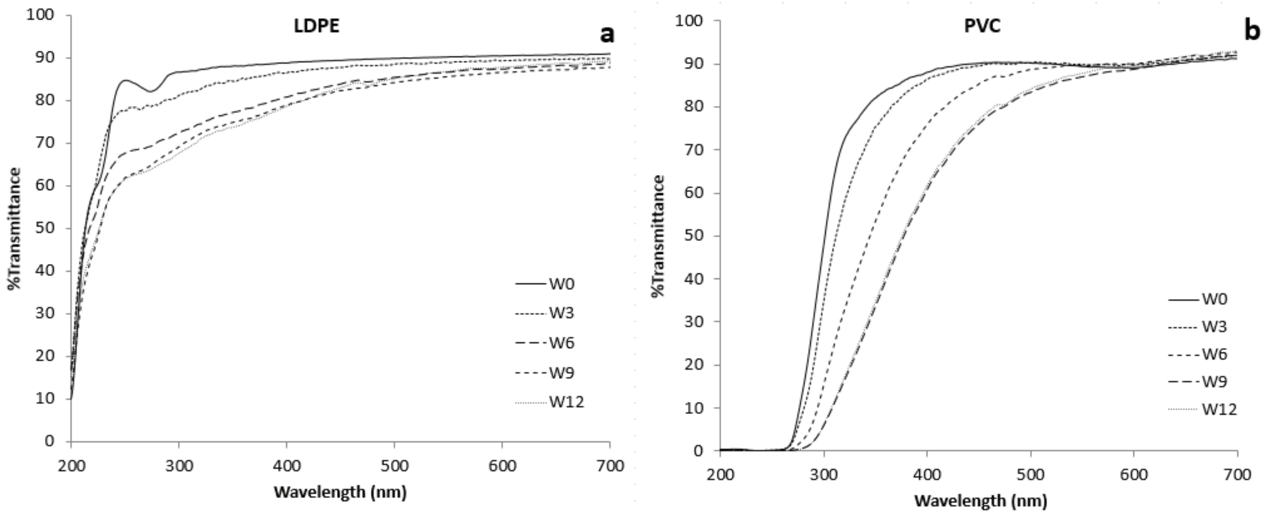


Fig. 7. UV-VIS spectra of the PVC boxes after different SODIS exposure periods, compared to the before-SODIS sample.

16.9%; however, since UVC radiation is mostly absorbed by the atmospheric layer, the transmittance range that is actually significant for SODIS applications is 290–400 nm. Therefore, 10.7–16.3% of significant transmittance of LDPE bags was reduced after 12 weeks of SODIS exposure.

The highest percentage reduction of transmittance in the UVA and UVB regions of the outer PVC boxes was 53.2% and 84.6%, respectively, after 12 weeks of SODIS exposure. These decreases in UV transmittances pose a limitation on the useful lifetime of the PVC boxes. Ideally, the boxes should be replaced after 4 weeks, when the UVA transmittance is 22.2% lower. If necessary, the boxes could be used for up to 8 weeks, when the UVA transmittance is nearly 50% lower, but the required time for SODIS would need to be lengthened.

3.5. Practicality

The use of a PVC box, in lieu of a bigger LDPE bag, as an outer layer of the new SODIS reactor design has some practical advantages. The PVC boxes make the SODIS set up much easier and faster, reducing burdens on the users. They are also

Table 2
The percentage reduction of transmittance at different wavelengths after different of SODIS exposures of LDPE and PVC

Radiation	%Reduction of transmittance of LDPE					
	W2	W4	W6	W8	W10	W12
UVC (250–290 nm)	4.0	20.4	22.8	20.0	17.8	16.9
UVB (290–320 nm)	3.9	5.2	14.5	14.7	14.0	16.3
UVA (320–400 nm)	2.1	3.0	10.4	10.0	9.6	10.7
Radiation	%Reduction of transmittance of PVC					
	W2	W4	W6	W8	W10	W12
UVC (250–290 nm)	6.6	48.7	53.4	55.9	56.7	54.0
UVB (290–320 nm)	9.4	50.7	65.9	78.8	86.2	84.6
UVA (320–400 nm)	4.1	22.2	34.4	46.4	55.7	53.2

more scratch-resistant and easier to clean, so they can remain attractive to the users for longer. The boxes are also strong and robust, so they do not need to be replaced very often. Nonetheless, their UV transmittances decrease significantly by week 4 of usage, so they should be replaced after 4 weeks.

4. Conclusions

This work investigated the changes in material integrity of solar water disinfection (SODIS) reactors, which had been developed to overcome usability-related shortcomings of previous designs, for usages in rural areas. During a 12-week period of usage of the new reactors in actual environments, the degradation of the materials was investigated weekly, with respect to surface morphologies, compositions, optical transmissions, tensile strengths, and leaching of organic compounds from the reactors into the treated water. The results showed that there were no scratches on the low-density polyethylene (LDPE) bags (the water-containing components of the reactors), which were problematic in the previous design. The maximum reduction of tensile strength was 28.5%. The optical transmittances in the UVA region of the LDPE and PVC components of the reactors decreased by 11% and 53%, respectively.

SEM micrographs showed that the LDPE surfaces became deteriorated after the SODIS usage but did not accumulated any scratches. The FTIR results indicated that the LDPE bags were photo-degraded via oxidation reactions, as carbonyl groups could be observed. On the other hand, no significant changes could be observed in the FTIR results for the PVC boxes. The tests for migration of organic compounds in the treated water found only 2,4 di-tert butyl phenol, which could be a photo-oxidation by-product from the LDPE bags, in the range of 1–3 $\mu g \cdot L^{-1}$, which was close to the method of detection's limit and still much lower than the harmful level set by the safety standard. The UVA transmittance results of the LDPE bags and PVC boxes showed that this property could be the limiting factor of the useful lifetime of the parts. While the LDPE bags could be used for up to 12 weeks of SODIS without too much reduction in UVA transmittance, the PVC boxes should be replaced after 4 weeks, or at most 8 weeks.

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