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Polishing of secondary treated wastewater using nano-ceramic hybrid PET waste plastic sheets

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ABSTRACT

This work aims to valorize a considerable part of solid waste that accumulates in the environment as well as the safe reuse of the treated wastewater. A non-woven fabric of polyethylene terephthalate (PET) was coated with a thin layer of a nano-ceramic material. The inhibitory effect of the nano-ceramic material was evaluated against gram +ve (*Staphylococcus aureus*) and gram –ve bacteria (*Salmonella typhimurium* and *Escherichia coli*) using the spread plate technique. Domestic wastewater was treated using a combined upflow anaerobic sludge bed (UASB) followed by a downflow hanging non-woven fabric (DHNW) reactor. The DHNW consisted of four segments (compartments). The hydraulic residence time (HRT) of the UASB reactor was 5 h. The coated sheets were used as a packing material for the DHNW reactor. The levels of chemical oxygen demand (COD), biochemical oxygen demand (BOD), and total suspended solids (TSS) were decreased in the final treated effluent (UASB/DHNW) from 386, 293, and 192 mg/L to 45, 30, and 12 mg/L with total removal states of 88%, 90%, and 90%, respectively. The fecal coliform and *E. coli* counts were reduced from 4.6×10^7 and 3.5×10^5 to 4.8×10^6 and 7×10^4 MPN/100 mL, respectively. The results showed that the effluent of the combined UASB/DHNW treatment system with the nano-ceramic sheets could be safely reused for different purposes.

Keywords: Upflow anaerobic sludge bed (UASB); Downflow hanging non-woven fabric (DHNW); Polyethylene terephthalate; Polyacrylic acid

1. Introduction

Plastic materials constitute an important part of our daily life. About 335×10^6 tons were produced globally [1] in 2016. They are expected to reach $1,124 \times 10^6$ tons in 2050 [2]. About 32×10^6 tons of plastics were collected in Europe as waste. Among these, 42% incinerated, 31% were collected for recycling, and 27% was landfilled. Thus, for the transition toward the circular economy, reusing, and recycling of plastic is marked as an essential step [3,4]. This is carried out to close the plastic loop. As a result, the concept of recycling has been widely applied in Europe for municipal solid waste, including plastic [3,4].

Polyethylene terephthalate (PET) is synthesized by the interaction of trans-esterification of dimethyl terephthalate and ethylene glycol (EG) or polycondensation of terephthalic acid (TPA) with EG. PET products have become one of the major produced thermoplastic materials [5]. Since PET is a very stable polymer and resistant to enzymatic or hydrolytic degradation, it has become a major component of plastic waste found in the environment [6].

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Egypt generated about 90 × 10^6 ton/y of solid waste [7]. According to previous calculations for the State of Kuwait (as a model), about 6.13 × 10^6 ton/y of plastic bottles were produced in Egypt [8]. Because of its very poor biodegradability, this form of plastic waste has accumulated in the environment for a long time. The reusing and recycling of such waste are important solutions. The recycling of non-woven fabric as packing materials for aerobic and anaerobic wastewater treatment was carried out in 2018 [9] and 2019 [10]. Unfortunately, biological treatment systems showed some deficiency for the removal of bacteria from the finally treated effluent [11–20]. Porous organic polymers act as favorable materials upon being used in environmental and antimicrobial applications [21–24].

Bacterial infections appear to be a major cause of morbidity and death. Thus, particular attention has been paid to new and emerging disinfection materials based on nano-particles. Extensive researches on metal nano-particles have revealed their wide range of antibacterial properties, high area to volume ratio, strong, and broad-spectrum antimicrobial activities [25]. The combination of transition metals such as (Cu^{2+}, Ag^{+}, Ce^{4+}, and Zn^{2+}) with biomaterials has recently reduced *in situ* cytotoxicity and microbial development. Copper, for example, may present a high antibacterial ability by keeping low cellular toxicity. For various activities in living organisms, a small amount of copper is required. Copper, in its elemental and associated forms, maintains anti-proliferative, anti-inflammatory, and anti-microbial properties. The use of various concentrations of copper doped Wollastonite ceramic material showed a wide range of antibacterial activity against gram +ve and gram –ve bacteria [26,27].

The method of preparing nano-ceramic sheets plays an important role in adjusting the properties of nano-particles. These properties are believed to be well-adjusted by adapting the wet precipitation method parameters. All these parameters could be easily controlled in the wet precipitation method to tolerate the properties of the nano-particles as confirmed previously [28]. Besides, low price precursors were used in the wet precipitation method compared to the sol–gel one. Our current research is focused on the addition of different copper oxide portions to the calcium silicate ceramic during the preparation process. This is performed by using the wet precipitation method (economic and flexible method). Under this perspective, doping of calcium silicate ceramic with copper would promote antibacterial as well as an antifungal process [27]. Upon introducing ceramics to polymers, they are capable of providing efficient materials. These substrates may contribute in the processes of water treatment [28]. Organic polymers can be loaded with graphene oxide [29] or ceramic clays to produce effective membranes for wastewater purification [30,31]. Acrylates as organic monomers are considered successful materials for coating organic and inorganic substrates to modify their surfaces for specific applications. Trimethylolpropane trimethacrylate (TMPTMA) was used for coating the surface of olive stones waste to be introduced as filler inside acrylonitrile-butadiene rubber [32]. Polyacrylic acid was used to enhance the efficiency of polyvinylidene fluoride with acceptable antifouling properties [33]. Acrylic acid was employed to improve the surface of waste rubber powder employing gamma radiation to be mixed with waste polyethylene as a low-cost treatment and reusing of polymeric wastes [34].

This work aims to evaluate the new idea of using nano-ceramic materials integrated with non-woven PET sheets as a disinfectant instead of conventional chlorination step.

2. Materials and methods

2.1. Materials

The used materials include polyvinyl alcohol (PVA), polyvinylpyrrolidone (PVP), acrylic acid, potassium persulfate and *N*,*N*′-dimethylene bisacrylamide purchased from (Sigma-Aldrich, Germany) and commercial starch from the local market. Used PET was washed with hot water and dried.

2.2. Methods and instrumentation

Complete physicochemical characteristics, fecal coliform (FC), and *Escherichia coli* (*E. coli*) analyses of the influent sewage and effluents were carried out according to American Public Health Association (APHA) [35]. The physicochemical analyses covered pH, organic loads (chemical oxygen demand (COD), biological oxygen demand (BOD)), nitrogenous load (ammonia–nitrogen (NH_3) and total Kjeldahl nitrogen (TKN)), oxidized nitrogenous compounds (nitrates (NO_3) and nitrites (NO_2)), total suspended solids (TSS), and volatile suspended solids (VSS).

2.3. Reactor configuration

The combined UASB/DHNW reactors (Fig. 1) were used in this study. The packing material was added at the lower part of the UASB reactor above 10 cm above the base. About 20 g/L VSS sludge was used at the beginning of the study. Table 1 depicts the HLR and OLR of the UASB

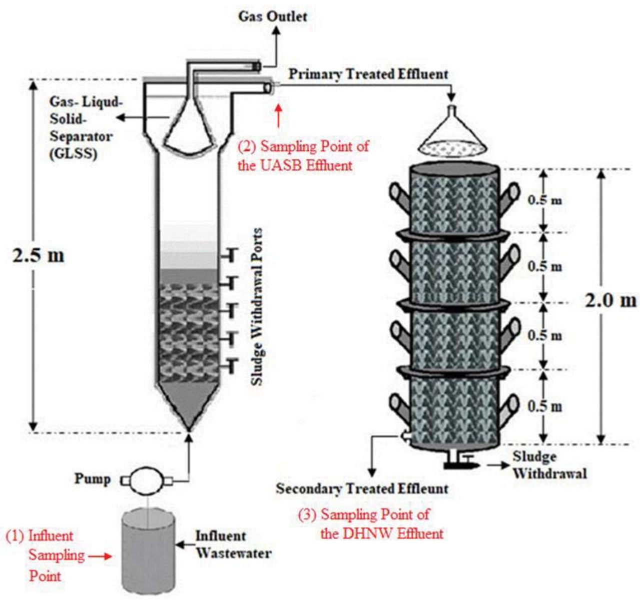

Fig. 1. Combined UASB/DHNW reactors [9,10].

reactor. Samples were collected weekly from the sampling points as indicated in Fig. 1.

The DHNW reactor was used as a post (secondary) treatment step for the UASB reactor. The dimension and operating conditions of the UASB and DHNW reactors were described by El-Khateeb et al. [9,10]. Fig. 2 shows the configuration of the DHNW reactor. The base of compartments 1, 2, and 3 was perforated to allow (seepage) the flow of water.

2.4. Non-woven (PET) fabrics

El-Khateeb et al. [9,10] investigated the non-woven (PET) fabrics as packing material. In the present study, the chosen pattern of non-woven fabric was corrugated for the UASB reactor and Bakelite hair rollers for the DHNW reactor.

2.5. Morphology of the packing material

JEOL JXA-840A electron probe micro-analyzer (Tokyo, Japan) scanning electron microscopy (SEM) was used to investigate the fiber of the packing material and the adhering of the nano-ceramic materials.

Table 1
Operating conditions for the UASB/DHNW reactors

Parameter	UASB	DHNW
Hydraulic loading rate (HLR), $m^3/m^3/d$	4.8	0.013
Organic loading rate (OLR), COD $kg/m^3/d$	1.9	0.672

2.6. Nano-ceramic sheets preparation

The nano-ceramic material was prepared according to Ammar et al. [26]. Different starting materials were used to achieve the coat effective membranes. The used materials for this study were different pure clays, aluminum silicate, and calcium silicate or calcium aluminate powders. The used polymers are PVA, polyvinyl pyrrolidone (PVP), and commercial starch. To improve the effectiveness of the bacterial disinfection cycle, various proportions of transition metals have been added. The starting materials were prepared, followed by the sintering of some doped transition metals and the processing of reinforced ceramic composites. Nano-ceramic preparation steps took place as follows: 5 g of PVA was dissolved in 30 mL of deionized water at 70°C for 30 min. The nano-ceramic powder was added step wisely to the solution with continuous stirring forming a slurry. The slurry will be kept to cool the added to the mold. The composite was dried at 100°C and then ignited at 500°C–600°C. The previous step was repeated by changing the (PVA) content to reach the optimum porous membrane for application in water treatment experiments. On parallel, another kind of organic polymers; PVP will be mixed with the prepared nano-ceramic powder to produce another porous ceramic sheet.

2.7. Coating of nano-ceramic sheets onto the plastic non-woven fabric

The plastic PET fabric sheets were immersed in distilled water at 70°C for 15 min. Potassium persulfate was added

Compartments of the DHNW reactor The DHNW (combined)

Fig. 2. Configuration of the DHNW reactor.

to the hot bath with stirring for 5 min. Acrylic acid was mixed with *N*,*N*′-dimethylene bisacrylamide then introduced to the solution containing PET sheets with continuous stirring for 30 min. The sheets were then moved from the solution and spread on a dry glass sheet. The wet surface of the fabric covered with the polyacrylic was then sprayed with the prepared ceramic powder on both sides. This was followed by keeping the modified non-woven fabric at 85°C for 3 h. The previous step is done for drying and confirming complete polymerization with cross-linking for polyacrylic acid. At this stage, avoiding the dissolution of polyacrylic acid which has an important role at which it attaches the ceramic powder and spreads it homogenously on the PET surface. Hence, the modified PET non-woven fabrics loaded with the nano-ceramic sheets are ready for experimental investigations.

3. Results and discussion

3.1. Morphological investigation via SEM

Fig. 3 shows the packing material before and after adhering to the nano-ceramic materials. The pure fabric in Fig. 3a displayed a smooth surface without any solid particle observation. The fibers of the non-woven fabrics were randomly distributed in all directions. This could enhance the possible mechanism of filtration of wastewater besides the biological treatment. Meanwhile, the treated non-woven fabrics in Fig. 3b revealed solid white CuO-doped ceramic. It was homogeneously distributed within the textile matrix. Such white ceramic-CuO particles are closely bound to the fiber surface via the formed thin layer of the polyacrylic acid layer. Such results confirm the possible application of the combined nano-ceramic and non-woven fabric sheets for targeted purposes.

3.2. Biological sewage water treatment

Sewage water was treated using a combined UASB and DHNW reactor. The system was in operation for 3 y and reached a steady-state condition. The characteristics of the effluent were consistent. Both reactors represent anaerobic/aerobic biological treatment techniques. By using the UASB reactor, the organic loads indicated through COD, BOD, and TSS were reduced from 386, 293, and 192 mg/L to 140, 100, and 61 mg/L, respectively. While in the final treated effluent, the residual concentrations of COD, BOD, and TSS were 45, 30, and 19 mg/L, respectively. The performance of the treatment system was comparable to that obtained by the fourth generation DHS reactor [36]. The entrapped TSS was degraded in/by non-woven fabric in the DHNW reactor under the long sludge retention time (SRT). The concentration of TSS in the final effluent was 19 mg/L, reflecting a 93% reduction. Regardless of the characteristics of the influent wastewater, the DHNW effluent quality was stable.

Fig. 4 shows the counts of FC and *E. coli* in raw sewage as well as the treated effluents. The counts of both FC and *E. coli* have reduced from 4.6×10^7 and 3.5×10^5 MPN/100 mL to 4.8×10^6 and 7×10^4 MPN/100 mL by using UASB reactor, respectively. The DHNW reactor treated the effluent of the UASB reactor extensively. The FC and *E. coli* residual counts were 2.7×10^5 and 2.5×10^3 MPN/100 mL, respectively. Just 2 logs order decreased by the UASB and DHNW reactors. The key removal method is the retention of suspended solids that may bind bacteria [9,10].

3.3. Performance of the nano-ceramic sheets as a tertiary treatment step

The secondary treated effluent still contains high FC and *E. coli* counts. To valorize the treated sewage water, a tertiary treatment step using nano-ceramic sheets was used. Figs. 4 and 5 show the nano-ceramic sheets output of the DHNW reactor effluent treatment.

Surfaces doped with Cu-nano-particles are capable of disinfecting liquid which is evaluated against gram +ve (*Staphylococcus aureus*) and gram –ve bacteria (*Salmonella typhimurium* and *E. coli*) using spread plate technique (Fig. 6). The mode of Cu-nanoparticles actions was summarized in El-Gendi et al. [37] resulted from denaturation of protein when contacted sulfhydryl groups in the bacterial outer membrane or cell wall. Copper ions interact with amines and carboxyl groups in *N*-acetylglucosamine and *N*-acetylmuramic acid in the peptidoglycan layer [38].

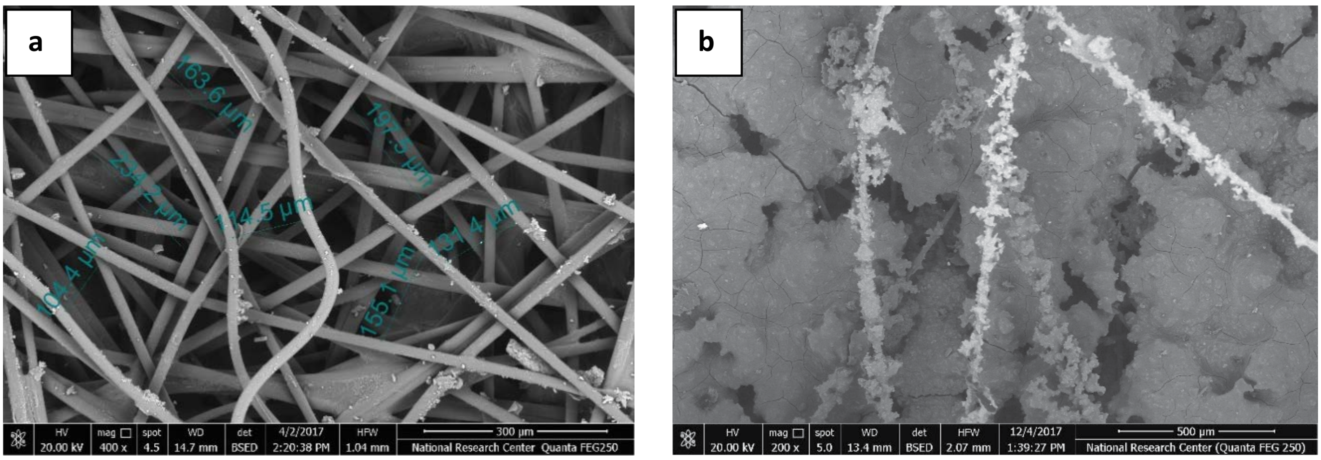

Fig. 3. SEM of the packing material (a) with (b) without ceramic nano-materials.

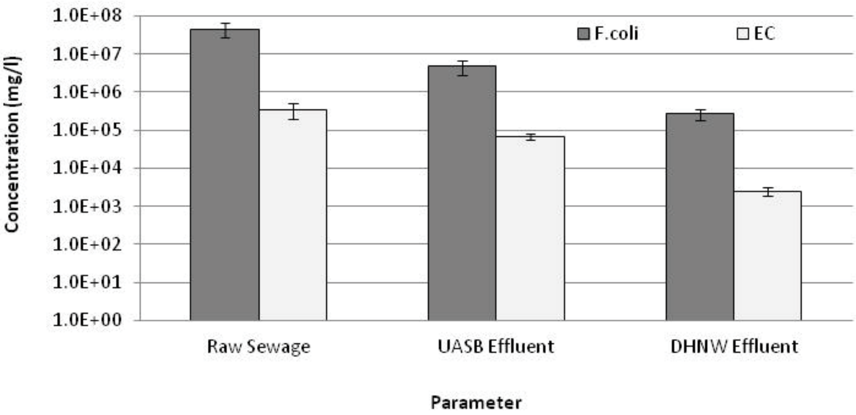

Fig. 4. FC and *E. coli* counts in sewage water and treated effluents.

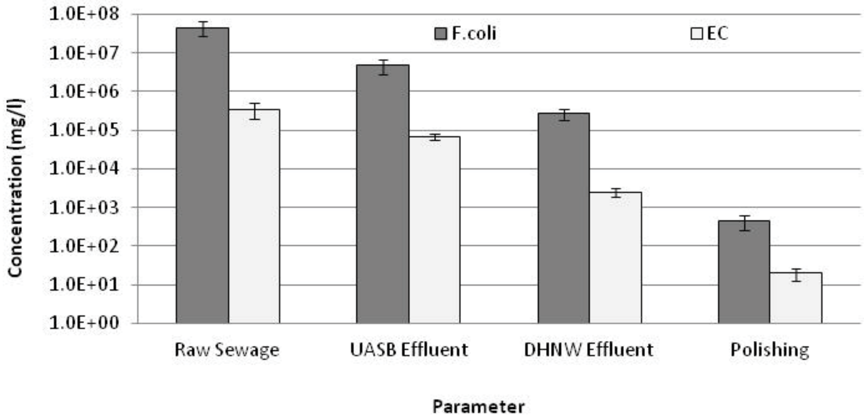

Fig. 5. Efficiency of the nano-ceramic sheet as a polishing step.

These interactions may cause destabilizing or breakage of bacterial cell wall and membrane, which is defined as the bacteriolytic effect. Copper ions disorganize helical structures of DNA that are involved in cross-linking within nucleic acid strands [39]. It causes lipid peroxidation and/or protein oxidation in case forming reactive oxygen species and reduces biofilm formation, where nanoparticles are embedded on the surface of the sheets rendering a much more active outer layer and reduce significantly the cell surface hydrophobicity, and consequently altering the attachment of bacteria. The surface of Cu NPs stops the exopolysaccharides, which has an axial role in biofilm formation and maturation [38,39]. The antibacterial effect is size-dependent [40] where nano-particles (about 8 nm) were found to be effective for the removal of bacteria via the formation of free radicals contributed to damaging the bacterial membranes. Results illustrated that the activity was enhanced upon introducing calcium silicate doped with copper. This may be due to the dissolution actions of the doped ions with copper samples from calcium silicate.

3.4. Valorization of the finally treated effluent

The FC, as well as E. coli counts, were reduced from 2.7×10^5 and 2.5×10^3 MPN/100 mL to 4.9×10^2 and 1.9×10 MPN/100 mL with removal rates of 99.8% and 99.2%, respectively. Egyptian Code No. 501/2005 [41] for wastewater reuses classified the treated wastewater into three grades: *A*, *B*, *C*, and *D* according to the level of treatment as shown in Table 2. Accordingly, it assigns the agricultural groups that can be irrigated by treated wastewater depending on the grade. Effluent limit values for microbiological parameters are suitable for reuse in irrigation of crops in category (*B*) including fodder/feed crops, trees producing fruits with epicarp, nursery plants, and trees used for green belts, etc.

4. Conclusions

The treatment of wastewater was carried out using combined UASB/DHNW reactors. The results showed that the bacterial count of both FC and *E. coli* were still high.

Therefore, the attached nano-ceramic material on the surface of non-woven (PET) fabric was employed to eliminate the remaining bacterial loads from the biologically effluent.

The counts of FC, as well as *E. coli*, were reduced by removal rates of 99.8% and 99.2%, respectively.

The quality of the final treated effluent could be used for irrigation of crops in category (*B*) including fodder/feed

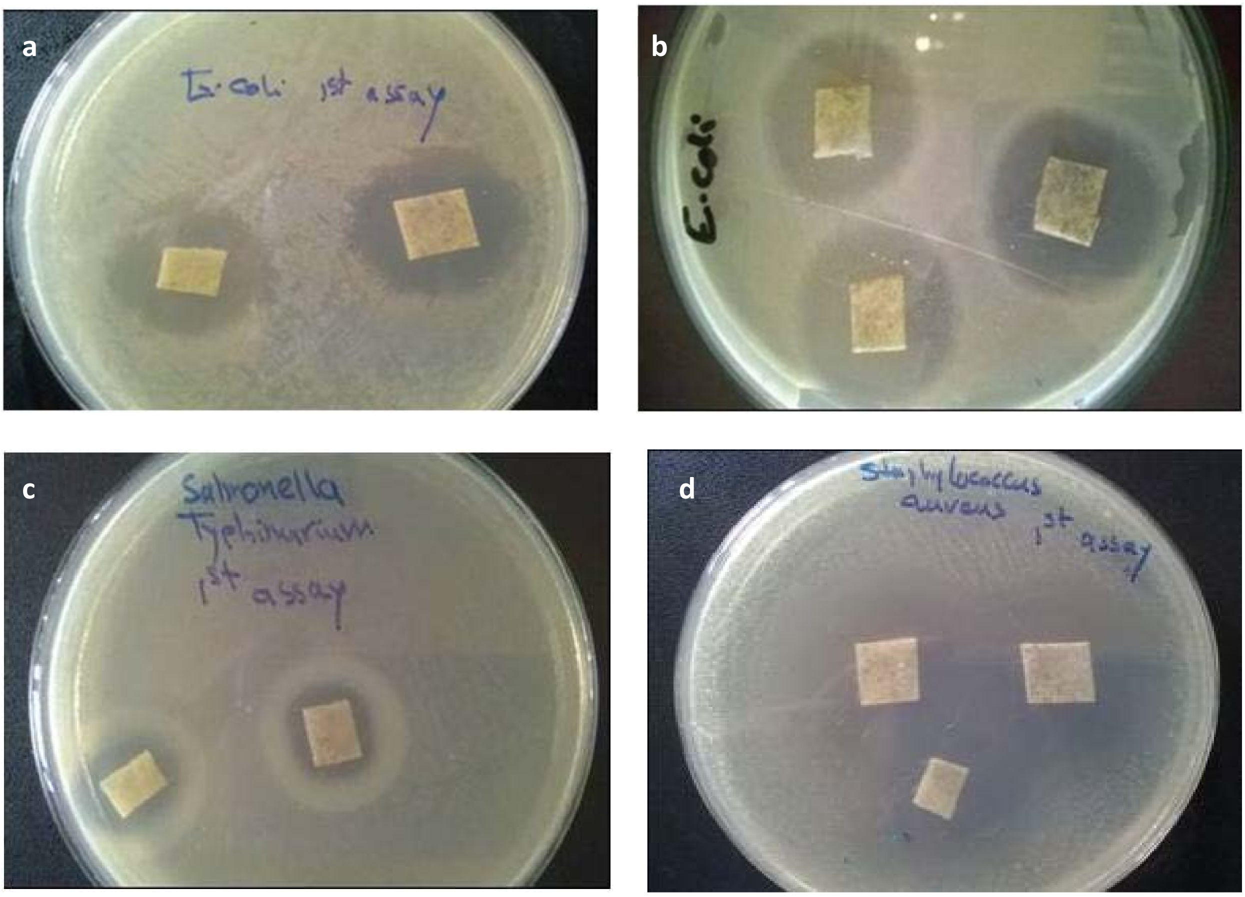

Fig. 6. Antibacterial activity of ceramic nano-material sheets against both gram +ve and gram –ve bacteria: (a and b) gram –ve bacteria (*E.coli*), (c) gram –ve bacteria (*Salmonella typhimurium* ATCC 6538), and (d) gram +ve bacteria (*Staphylococcus aureus*).

Table 2
Egyptian Code No. 501/2005 for wastewater reuses [41]

Treatment grade		*A*	*B*	*C*	*D*
Maximum allowable physical and chemical standards	TSS (mg/L)	15	30	50	300
	Turbidity (NTU)	5	ND	ND	ND
	BOD_5 (mg/L)	15	30	80	350
Maximum allowable bacteriological standards	Fecal coliform (*E. coli*)/100 mL	20	100	1,000	ND

ND: not determined.

crops, trees producing fruits with epicarp, nursery plants, and trees used for green belts [40].

Valorization of both liquid waste (sewage water) as well as solid waste such as waste PET, took place.

The application of such technology will reduce the accumulation of a major part of solid waste in the environment.

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Conflict of interest

The authors certify that there is no conflict of interest with anyone. This is a confession of us to do so.

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