Potential and prospects of photocatalytic disinfection: Using sustainable solar-energy-driven photocatalysts

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ABSTRACT

The use of solar-energy-driven photocatalysts for sewage disinfection treatment was investigated, and the photocatalyst RGO/Fe,N-TiO₂/Fe₃O₄@SiO₂ (RGOFeNTFS) was specifically developed for sewage disinfection. The study involves examining the photocatalyst's potential for treating real sewage samples from four different local sewage treatment works. Sewage samples are classified by region based on the use of different types of water for toilet flushing, such as low-salinity sewage for regions using freshwater and high-salinity sewage for regions using seawater. The applicability of the technology for sewage disinfection was identified based on the required reaction time necessary for the treated sewage to comply with the corresponding *E. coli* discharge standards. Under simulated solar irradiation, RGOFeNTFS shows potential for treating low-salinity sewage, for which a relatively short reaction time is needed to meet the relevant standards. The reaction can damage the cell structure of the bacteria, causing damage indicated to be irreversible by the live/dead assay. When evaluating the potential of the technology for practical application, sewage characteristics such as suspended matter and organic matter on photocatalytic disinfection should be carefully reviewed. The nontoxicity of the RGOFeNTFS photocatalyst to the marine environment was indicated by the natural-born phytoplankton in seawater.

KEYWORDS Disinfection; photocatalysis; sustainable; sewage; solar energy; TiO₂

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1. Introduction

Clean water is essential for human beings and wildlife. Pathogenic microorganisms in water can cause diseases and threaten the ecosystem, and thus the disinfection of water and access to clean water is of great concern to society. In this regard, the bacterial density (commonly indicated as the *E. coli* density) of treated water/wastewater is a mandatory criterion for assessing water quality and the efficiency of disinfection methods (HKDSD, 2020; APHA, 2017; Wen et al., 2020). For sewage treatment, disinfection of bacteria is necessary to reduce the potential risks that some bacteria pose to human beings and the environment. Conventional disinfection methods like chlorination and UV disinfection have limitations such as being chemical- or energy-intensive, and have the drawbacks of producing disinfection byproducts and bacterial re-growth (Khadgi et al., 2017; Li et al., 2016). The quest is continuing for the development of other suitable disinfection technologies which comply with simple and effective implementation and maintenance, and mainly use environmentally friendly and sustainable energy sources such as solar energy. Photocatalysis is a promising technology with these advantages. An investigation of the application of photocatalysis driven by solar light, which has the benefit of low energy consumption and low greenhouse gas emissions, is of great value for the improvement of sewage disinfection.

TiO₂ is one of the most explored photocatalysts in previous decades due to its physiochemical inertness,

nontoxicity and low price (Asahi et al., 2014). However, the application of photocatalysis using TiO₂ is limited due to its restricted response to only the UV region, which covers a mere 5% of the solar spectrum, and the difficult separation of the photocatalytic materials from the treated solution (Asahi et al., 2014; Dong et al., 2015). Further development of TiO₂ is necessary for practical application. The co-doping method using Fe and N can modify TiO₂'s bandgap, hence allowing it to respond to a larger range of wavelengths (He et al., 2019). The basis of photocatalytic disinfection is the redox potential of the photocatalytically generated electrons and holes (Asahi et al., 2014). The introduction of reduced graphene oxide (RGO), which has high conductivity, promotes the opportunities for separation of the electrons and holes by inhibiting their recombination in the photocatalyst bulk (He et al., 2021). Moreover, to address this concern about the separation of the photocatalyst from the treated sewage, introducing magnetic properties has proved an efficient strategy, in which Fe₃O₄@SiO₂ has been designed as the magnetic core (He et al., 2019; He et al., 2021). In this study, TiO₂ has been innovatively engineered through the abovementioned strategies, obtaining the titanium dioxide-based magnetic photocatalyst (RGO/Fe,N-TiO₂/Fe₃O₄@SiO₂, its short form RGOFeNTFS will be used herein).

Rather than using synthetic water, this study evaluates the photocatalyst and examines the practical potential of the technology in real sewage, whose complex components are difficult to mimic in the lab. In such real sewage, the

practical value of photocatalysis under practical conditions is better understood and evaluated. The effects of sewage characteristics on the disinfection performance imply to what extent the technology can be applied in a real environment, with the fluctuation of sewage characteristics. As the practical application is a concern, the discharge standards (*E. coli* density) of the sewage effluent are the criteria for evaluating the performance of photocatalytic disinfection using RGOFeNTFS driven by solar energy.

In the sewage treatment process, sewage effluent is finally discharged into a body of water. The safety of the photocatalyst materials is a concern for society. Therefore, a toxicity assessment of the photocatalysts is crucial in understanding photocatalytic disinfection's practicality (Sendra et al., 2017; Rueda-Marquez et al., 2020). Phytoplankton is the basic species of the coastal food web and can rapidly reflect the effects of the discharge on marine water, due to its high productivity and quick response to environmental variation (Miller et al., 2010). The toxicity of a photocatalyst to the ecosystem after the photocatalytic disinfection process is indicated by phytoplankton in seawater.

In this study, solar-energy-driven RGOFeNTFS has been developed for photocatalytic disinfection (under simulated solar light) for four different types of sewage samples collected from local sewage treatment works (STWs). In Hong Kong, different types of water (freshwater and seawater) are used for toilet flushing in different regions, which produces different types of sewage. Low-salinity and high-salinity sewage samples are classified in this study based on the sewage sources that using freshwater and seawater for toilet flushing, respectively. STWs-1: low-saline sewage after tertiary treatment; STWs-2: low-saline sewage after secondary biological treatment; STWs-3: high-salinity sewage after secondary biological treatment; STWs-4: high-salinity sewage after chemically enhanced primary treatment. The effects of the sewage characteristics on the disinfection efficiency were also studied to understand the tolerance of the technology for the fluctuation of sewage quality. Finally, as a vital aspect of evaluating the photocatalytic disinfection of real sewage using RGOFeNTFS, a toxicity assessment of the photocatalyst material was also conducted using phytoplankton as an indicator.

2. Methodology

2.1. Chemicals and reagents

Ferrous chloride tetrahydrate (99%), ferric chloride hexahydrate (99.9%), tetraethyl orthosilicate (98%), titanium (IV) butoxide (97%), urea (99%), graphene oxide dispersion in H₂O (4 mg/mL), and m-lauryl sulfate broth were purchased from Sigma-Aldrich. Trisodium citrate dihydrate (99%) was obtained from VWR. Ethanol (99.9%) and ammonia hydroxide solution (25%) were obtained from

Merck. All the chemicals and reagents were used without any further purification. Throughout the synthesis process, deionised water was used.

2.2. Synthesis and characterisation of RGOFeNTFS

The RGOFeNTFS was synthesised via a multi-step method, which is a similar synthesis process to our previous study (He et al., 2021). Briefly, the magnetic core of Fe₃O₄@SiO₂ is firstly synthesised via the co-precipitation method. Then co-doping of TiO₂ is performed via the sol-gel procedure for obtaining TiO₂ from titanium butoxide, and this process concurrently occurs with the formation of the Fe,N-TiO₂/Fe₃O₄@SiO₂ composite. Finally, RGO is combined with the above photocatalyst composites, obtaining RGOFeNTFS after calcination under a N₂ gas atmosphere.

The structure, magnetisation, optical property and morphology of RGOFeNTFS (using XRD and Raman, UV-vis DRS and TEM-EDS mapping, respectively) have also been reported in a previous study (He et al., 2021). The morphology of RGOFeNTFS has been further analysed using TEM.

2.3. Collection and characterisation of sewage samples

All the sewage samples used in the study were collected upstream from the disinfection operation unit, in four different types of STWs: (1) STWs-1: low-saline sewage after tertiary treatment; (2) STWs-2: low-saline sewage after secondary biological treatment; (3) STWs-3: high-salinity sewage after secondary biological treatment; (4) STWs-4: high-salinity sewage after chemically enhanced primary treatment. They cover most of the sewage types in Hong Kong. The collection of sewage samples was carried out following the standard sewage sample collection and storage method (APHA 1060). The discharge standards of *E. coli* (95 percentile) for different Hong Kong local STWs are shown below, STWs-1 and STWs-2: 1,500 CFU/100 mL, STWs-3: 15,000 CFU/100 mL, STWs-4: 900,000 CFU/100 mL (HKDSD, 2020). The characteristics of the sewage samples were measured in detail after collection, with three parallel measurements for each parameter (n=3). The constituents of real sewage are complicated and there may have been certain factors affecting the final photocatalytic disinfection performance. In this study, sewage samples were collected in different months to observe the fluctuation of water quality and to study the photocatalytic disinfection capacity under different sewage conditions.

2.4. Photocatalytic disinfection procedure

2.4.1. Photocatalytic disinfection reaction

The experimental setup was developed in a previous study (He et al., 2021). The reaction was performed in a 125

mL conical flask, with a solution volume of 50 mL, under vigorous stirring. The light source was a xenon light (300 W, PLS-SXE300UV, PerfectLight Technology, Beijing) with an Air Mass 1.5 Global filter, producing the simulated solar irradiation for the photocatalytic disinfection. The irradiation intensity of the incident light is 816 W/m^2 , which is measured at the centre of the flask where the reaction took place. The reaction temperature was about 25°C for all tests. Three parallel samples were taken at each sampling point at the designed time intervals. Photocatalytic disinfection was conducted in the original sewage samples within one week after the collection. As discussed in the aforementioned previous study, the optimum dosages of RGOFeNTFS for the disinfection reaction were 1, 3, 3 and 3 g/L for STWs-1, STWs-2, STWs-3 and STWs-4 sewage samples, respectively (He et al., 2021). The tests in this study were performed in triplicate.

2.4.2. Live/dead assay before and after disinfection

To investigate the bacterial cell damage before and after the photocatalytic disinfection reaction, a live/dead viability assay was used, employing two fluorescent nucleic acid dyes, diaminophenylindole dihydrochloride (DAPI) and propidium iodide (PI), to stain the DNA of the bacteria. Since bacterial cells have cell membranes, only certain types of fluorescent staining dyes (such as DAPI) can penetrate the intact cell membrane of viable bacteria and stain the DNA in the cytoplasm. PI cannot penetrate an intact bacterial cell membrane and can only reach the DNA in the cytoplasm through holes on the damaged bacterial cell membranes caused by photocatalytic disinfection reaction. Consequently, the cell membrane-permeable DAPI can stain both viable and dead bacteria with blue fluorescence, while the membrane-impermeable PI only stains dead bacteria with red fluorescence. By comparing the fluorescent colours of the stained bacteria, the dead bacteria (fluorescent red) can be identified among all the bacteria (both viable and dead, fluorescent blue). The overlay column shows an overall picture of the viable and dead bacteria.

2.4.3. Changing sewage characteristics

Among the four types of sewage samples, the STWs-1 and STWs-2 sewage samples are more promising for the application of photocatalytic disinfection. The effects of sewage characteristics such as pH, ion concentration, dissolved oxygen (DO), turbidity, and TOC on the photocatalytic disinfection performance was evaluated for these two sewage samples. The sewage characteristics were changed based on the original values. pH values were changed using NaOH (0.1 mM) and HCl (0.1 mM). The turbidity was studied by adding suspended solids isolated from the sewage samples. The TOC value was adjusted by adding commercial Suwannee River NOM

(Suwannee River NOM (2R101N) obtained from the international humic substances society. Different DO levels were controlled by injecting N_2 gas to achieve 4 and 6 mg/L level and by monitoring the DO level using DO meter at the same time. Corresponding sodium and nitrate salts were added to the sewage to change the specific ion concentration. Only one characteristic was changed for each test; the experimental conditions were consistent. As the natural decay of *E. coli* occurs over time, in the tests studying the effects of sewage characteristics on photocatalytic disinfection, natural *E. coli* from the sewage samples was isolated and cultured in Luria-Bertani (LB) broth and then added to the sewage to keep the initial *E. coli* density of each test consistent. The *E. coli* was prepared as described in the previous study (He et al., 2021).

2.4.4. Bacterial enumeration

The *E. coli* densities (criteria of sewage discharge standards) of the sewage samples were enumerated using the membrane filtration technique with m-lauryl sulfate broth as the selective medium, as described in the local sewage discharge standard (HKEPD, 1991). Cultures pertaining to other bacteria are inhibited by the selective m-lauryl sulfate broth, giving it the ability to distinguish *E. coli* from other bacteria in real sewage.

2.5. Bioassay using phytoplankton to indicate toxicity

In this study, to diminish any adverse effects of the ambient environment on the phytoplankton, seawater samples containing phytoplankton were collected on the HKUST campus pier at 17:00. Subsequently, the collected seawater containing phytoplankton was mixed with photocatalyst materials, and kept in the dark at room temperature. The phytoplankton density of the test samples (with the addition of RGOFeNTFS) was monitored for 48 hours, and compared to that of the control sample with phytoplankton in seawater, without any addition. All the samples were kept in a 12-well test plate in the dark at room temperature. Phytoplankton was observed and the density counted under a microscope (Olympus, Model CKX41). Three parallel samples were taken at each sampling point at the designed time intervals.

3. Results and discussion

3.1. Characterisation

The crystalline structure, morphology, elemental properties, magnetism and optical properties of RGOFeNTFS have been discussed in the previous study; that RGOFeNTFS can be solar-energy driven, and the successful reduction of graphene oxide to RGO in the photocatalyst composites, has also been evidenced by

Raman in a previous study (He et al., 2021). The TEM image (Fig. 1) indicates that RGOFeNTFS has a composite structure consisting of RGO, $\text{Fe}_3\text{O}_4@\text{SiO}_2$ core and Fe,N-TiO_2 components.

Moreover, RGOFeNTFS is superparamagnetic and can be magnetically separated from the treated sewage samples in the presence of a magnetic field (200 mT). The magnetic separation efficiency of RGOFeNTFS was retrieved using Equation (1) by measuring the solution turbidity before (initial turbidity) and after (final turbidity) and the magnetic separation of RGOFeNTFS from the treated sewage (He et al., 2021). RGOFeNTFS exhibits a magnetic separation efficiency of the photocatalyst at 99.5% (dosage of 1 g/L) and 99.2% (dosage of 3 g/L) in STWs-1 and STWs-2 sewage samples, respectively after 10 minutes of magnetic separation.

Magnetic separation efficiency (%) =

$$\frac{(\text{initial turbidity-sewage turbidity}) - (\text{final turbidity-sewage turbidity})}{\text{initial turbidity-sewage turbidity}} \times 100\% \quad (1)$$

Figure 1. TEM images of RGOFeNTFS.

3.2. Photocatalytic disinfection of real sewage

An evaluation of the photocatalytic disinfection using RGOFeNTFS for four different types of sewage samples (low-salinity sewage after tertiary treatment, low-salinity sewage after secondary biological treatment, high-salinity sewage after secondary biological treatment, and high-salinity sewage after chemically enhanced primary treatment) was conducted to determine the potential of the technology for practical application.

3.2.1. Photocatalytic disinfection of STWs-1 sample

The dosage of RGOFeNTFS used in the photocatalytic disinfection of STWs-1 sewage samples was 1 g/L, the optimised dosage determined previously (He et al., 2021) for this sewage sample. The discharge standard of *E. coli* is 1,500 CFU/100 mL. Sewage samples were collected in different months and characterised as shown in Appendix Table I. The sewage characteristics of these samples were stable with low fluctuation, implying that stable photocatalytic disinfection performance can be achieved at different times due to the stable sewage characteristics. The photocatalytic disinfection of two STWs-1 sewage samples, collected on 9 October 2018 and 9 May 2019, was studied (Figure 2). The sewage samples, after photocatalytic disinfection of around 20 minutes, could meet the discharge standard. After a 30-minute reaction, a 1.08-log reduction of *E. coli* density was achieved in the sewage sample (9 October 2018), in which the *E. coli* density was reduced from (3.79 ± 0.06) -log to (2.71 ± 0.06) -log CFU/100 mL, fully meeting the discharge standard of 1,500 CFU/100 mL (3.18 -log CFU/100 mL). A photocatalytic disinfection performance in line with the discharge standard was also achieved for the sewage sample collected on 9 May 2019. Under the same experimental conditions, the *E. coli* density was reduced from (3.44 ± 0.06) -log to (2.97 ± 0.08) -log CFU/100 mL, which is below the discharge standard of 3.18 -log CFU/100 mL. Moreover, RGOFeNTFS was applied for another two cycles of reaction, in which the treated STWs-1 sample meets the discharge standard of *E. coli* under the same experimental conditions. It indicates that RGOFeNTFS is efficient for photocatalytic disinfection of STWs-1 sewage in different seasons under solar light.

Figure 2. Photocatalytic disinfection performance of the STWs-1 sewage samples.

Damage to the bacterial cells during the photocatalytic disinfection of the STWs-1 sewage sample was examined using a live/dead viability assay. Figure 3 shows the fluorescent images of the stained bacteria measured at different stages. Before disinfection, as shown in the overlay image (Figure 3-1c), the bacteria were stained blue with a limited number of fluorescent red bacteria, indicating that most of the bacteria were viable with an intact cell wall structure. The presence of the dead fluorescent red bacteria was probably due to the natural decay of bacteria in the sewage before observation. After the photocatalytic

disinfection reaction, the majority of the bacteria turned to fluorescent red (Figure 3-2c), indicating that the damage to the bacterial cell walls occurred in the reaction, which subsequently resulted in the disinfection of the bacteria. The bacteria in the samples (24 hours and 48 hours after the reaction) mainly appeared in fluorescent red (Figure 3-3c,4c), consistent with the sample after 30 minutes of photocatalytic disinfection reaction, indicating that the disinfected bacteria in the treated sewage remained disrupted and could not recover after the reaction. It can be concluded that (1) there was no bacterial regrowth in the STWs-1 sewage sample after the photocatalytic disinfection treatment and (2) damage to the bacteria caused by the photocatalytic disinfection reaction is irreversible.

Figure 3. Fluorescence images of bacteria (STWs-1) stained by DAPI and PI: (1) bacteria before photocatalytic disinfection; (2) bacteria after 30 minutes of reaction; (3) bacteria kept for 24 hours after reaction; (4) bacteria kept for 48 hours after reaction. (a) The blue fluorescence illustrates bacterial cells stained with DAPI, (b) red fluorescence illustrates inactivated bacterial cells stained with PI, and (c) merged image of (a) and (b).

3.2.2. Photocatalytic disinfection of STWs-2 sample

The STWs-2 sewage samples have poorer water quality and higher initial *E. coli* density compared to the STWs-1 sewage samples (Appendix Table I). According to the dosage optimisation for the STWs-2 sewage samples, an RGOFeNTFS dosage of 3 g/L was selected for the photocatalytic disinfection of STWs-2 sewage samples (He et al., 2021). As shown in Figure 4, photocatalytic disinfection tests under simulated solar light were performed for the STWs-2 sewage samples collected on 7 September 2018 and 18 October 2019. After around 45 minutes of reaction, the STWs-2 sewage sample (7 September 2018) could meet the discharge standard of 1.5×10^3 CFU/100 mL, (3.18-log CFU/100 mL), showing a 2.88-log reduction of *E. coli* density which was reduced from (4.91 ± 0.08) -log to (2.03 ± 0.05) -log CFU/100 mL.

The sewage sample collected on 18 October 2019 showed a 1.97-log reduction of *E. coli* density after 60 minutes of photocatalytic disinfection, which is also an acceptable treatment time. In the cycle test, consistent performance was obtained over three cycles, with the treated STWs-2 sewage samples meeting the *E. coli* discharge standard after 60 minutes of reaction, illustrating the practical application potential of photocatalytic disinfection technology in the STWs-2 sewage samples.

The content of suspended solids in STWs-2 sewage samples, reflected by TSS and turbidity, is higher than that of STWs-1 sewage samples. They are believed to have a more noteworthy impact on photocatalysis, as the suspended solids may enhance the light scattering losses and hinder the incident light on the photocatalyst surface in the photocatalysis process, hence suppressing the photocatalytic disinfection efficiency (Arya and Philip, 2014; García-Fernández et al., 2015). Considering the acceptable reaction time for treating STWs-2 sewage samples, it was also selected for subsequent study due to its potential for practical application.

Figure 4. Photocatalytic disinfection performance of the STWs-2 sewage samples.

As shown in Figure 5, damage to the bacterial cells was examined using a live/dead viability assay. Bacteria in the STWs-2 sewage sample were stained and monitored during and after the reaction. Before disinfection, as shown in the overlay image (Fig. 5-1c), most of the bacteria were fluorescent blue with few bacteria having red fluorescence, indicating most of the bacteria were viable with an intact cell membrane structure. The presence of the dead bacteria was likely due to the natural decay of the bacteria before the observation. After 60 minutes of photocatalytic disinfection reaction, most of the bacteria showed red fluorescence (Figure 5-2c), indicating damage to the bacterial cell membrane by the photocatalytic reaction. After the reaction had been stopped for 24 hours (Figure 5-3c) and 48 hours (Figure 5-4c), the majority of the bacteria appeared with red fluorescence, being similar to the sample only treated by photocatalytic disinfection (Figure 5-2c). This demonstrates that regrowth of the disinfected bacteria does not occur in STWs-2 sewage sample after the photocatalytic disinfection treatment, and the damage to the bacterial structure is irreversible.

Figure 5. Fluorescence images of bacteria (STWs-2) stained by DAPI and PI: (1) bacteria before photocatalytic disinfection; (2) bacteria after 60 minutes of reaction; (3) bacteria kept for 24 hours after reaction; (4) bacteria kept for 48 hours after reaction. (a) The blue fluorescence illustrates bacterial cells stained with DAPI, (b) red fluorescence illustrates inactivated bacteria cells stained with PI, and (c) merged image of (a) and (b).

3.2.3. Photocatalytic disinfection of STWs-3 sample

The RGOFeNTFS dosage used in the photocatalytic disinfection tests for the STWs-3 sewage samples was 3 g/L, as the initial *E. coli* density of STWs-3 sewage samples was similar to that of the STWs-2 sewage sample. As shown in Figure 6, photocatalytic disinfection tests were conducted in the STWs-3 sewage samples collected at different times (3 October 2018 and 5 September 2019). The required photocatalytic disinfection time for STWs-3 sewage samples (3 October 2018 sewage sample) to meet the *E. coli* discharge standard was 240 minutes, which is impractical for sewage disinfection. For the sewage sample collected on 5 September 2019 with a higher initial *E. coli* density, the treated sewage sample did not meet the discharge standard even after 240 minutes reaction. The STWs-3 sewage samples collected in different years and their characteristics were stable with less parameter fluctuation, as shown in Appendix Table I, especially for the parameters of TSS and turbidity which are crucial to the photocatalytic disinfection efficiency (Arya and Philip, 2014; García-Fernández et al., 2015). Considering the stable sewage characteristics, the photocatalytic disinfection efficiency under the same experimental conditions for the STWs-3 sewage samples is likely consistent, taking a relatively long reaction time for the treated sewage sample to meet the discharge standard. It indicates the poor potential of the technology for the practical application in STWs-3 sewage.

Figure 6. Photocatalytic disinfection performance of the STWs-3 sewage samples.

3.2.4. Photocatalytic disinfection of STWs-4 sample

With an RGOFeNTFS dosage of 3 g/L, the photocatalytic disinfection experiments for the STWs-4 sewage samples were carried out under the same experimental conditions as those of the other three types of sewage samples. In this study, two sewage samples were collected from the STWs-4 STWs on 2 October 2018 and 8 May 2019, respectively (Appendix Table I). As illustrated in Figure 7, after 240 minutes of reaction under the irradiation of simulated solar light, only a 0.2-log reduction of *E. coli* density was achieved in the sewage sample collected on 2 October 2018, being reduced from (7.42 ± 0.05) -log to (7.22 ± 0.04) -log CFU/100 mL, and it is far higher than the required discharge standard of 9×10^5 CFU/100 mL (5.95 -log CFU/100 mL). Moreover, the sewage sample collected on 8 May 2019 had a 0.14-log reduction of *E. coli* density (i.e. reduced from (7.85 ± 0.03) -log to (7.71 ± 0.06) -log CFU/100 mL), which is also higher than the required discharge standard.

The poor performance of photocatalytic disinfection is probably due to the high TOC content, TSS content, turbidity and *E. coli* initial density of these two STWs-4 sewage samples (Appendix Table I). As mentioned previously, high TSS content and turbidity not only increase the light scattering loss, but also induce a shielding effect on the photocatalyst by covering its surface. Consequently, it may result in a reduction of disinfection efficiency (HKEPD, 1991). Photocatalytic disinfection technology may not be suitable for the treatment of STWs-4 sewage.

Figure 7. Photocatalytic disinfection performance of the STWs-4 sewage samples.

Moreover, light control (under solar irradiation without adding RGOFeNTFS) and dark control (with the addition of RGOFeNTFS in dark condition) tests were

conducted to understand the effects of solar irradiation and the presence of photocatalyst on bacteria (Table 1). In the dark control tests, *E. coli* densities are stable with the presence of RGOFeNTFS in the dark, indicating the insignificant effect of the photocatalyst on bacteria without solar light irradiation. Decrease of *E. coli* density was observed in the light control tests for the four types of sewage samples, showing the inactivation effect of solar irradiation on bacteria. However, the disinfection of *E. coli* in the photocatalytic reaction is more significant compared to the light control results (except for the poor efficiency for STWs-4), proving the disinfection is mainly due to the photocatalysis induced by RGOFeNTFS under solar light irradiation.

Table 1. Control tests for different types of sewage samples.

Sewage type	Initial (log10 CFU/100 mL)	Final density (light control(1), log10 CFU/100 mL)	Final density (dark control(2), <i>E. coli</i> viability)
STWs-1	3.72	3.36 (after 30 min)	96% (after 30 min)
STWs-2	5.06	3.96 (after 90 min)	92% (after 90 min)
STWs-3	5.38	4.80 (after 240 min)	93% (after 240 min)
STWs-4	7.86	7.79 (after 240 min)	98% (after 240 min)

Note:

(1) Light control: under solar irradiation without adding RGOFeNTFS; (2) Dark control: with the addition of RGOFeNTFS in dark condition.

Photocatalytic disinfection is based on the redox potential of the reactive species generated by the photocatalysts activated under solar energy. Reactive species approach sewage constituents during the diffusion journey before attacking microorganisms in the sewage. The sewage characteristics can impact the disinfection performance of RGOFeNTFS. Among these four sewage samples, the STWs-1 sewage sample has satisfactory quality with the lowest total suspended solids (TSS) content, turbidity, chemical oxygen demand (COD) and *E. coli* density. Compared with the other three types of sewage samples, the STWs-4 sewage sample exhibits relatively poor quality, not only possessing the highest TSS content and turbidity, but also the highest *E. coli* density and COD. Compared to the STWs-1 sewage sample, the STWs-2 sewage sample has higher TSS, turbidity and COD, and its *E. coli* density is almost an order of magnitude higher than that of STWs-1 sewage. For the STWs-3 sewage sample, its turbidity is a little lower than that of the STWs-2 sewage sample, but the TSS content of these two sewage samples are similar. Moreover, the measured *E. coli* density of the STWs-3 sewage sample is very high, two orders of magnitude higher than that of the STWs-1 sewage sample. The poorer sewage characteristics are in line with the decreasing photocatalytic disinfection efficiency. Regarding the practical operation conditions of disinfection treatment, the technology shows potential for treating STWs-1 and

STWs-2 sewage samples, for which a relative short reaction time (around 20 and 45 minutes, respectively) is needed for the treated sewage to meet the *E. coli* discharge standards.

3.3. Effects of sewage characteristics

In this study, the key sewage characteristics were investigated for their roles in affecting the results as to whether the sewage samples are able to meet the discharge standard of *E. coli* after photocatalytic disinfection. Since the technology is more promising in treating STWs-1 & 2 sewage samples, the effects of sewage characteristics, such as pH, ion concentration, dissolved oxygen (DO), turbidity, and total organic carbon (TOC) on the photocatalytic disinfection technology, were evaluated for the STWs-1 and STWs-2 sewage samples. The discharge standards of *E. coli* (the same standard value for both sewage samples: 1,500 CFU/100 mL) were used as the criteria to evaluate the effects of sewage characteristics on photocatalytic disinfection. For STWs-1 sewage sample, considering the initial *E. coli* density of the sewage sample of 3.8-log CFU/100mL, the treated sewage sample could meet the discharge standard when a reduction of the *E. coli* density value of 0.62-log CFU/100 mL was achieved after 30 minutes of reaction. On the other hand, for STWs-2 sewage sample, a reduction of the *E. coli* density value of 1.86-log CFU/100 mL was achieved after 60 minutes of reaction with the initial *E. coli* density of 5.04-log CFU/100mL.

As shown in Table 2, fluctuation of the sewage characteristics affects the results as to whether the treated sewage samples meet the discharge standard. For the STWs-1 sewage sample, the impacts of variations in pH, ions concentration and DO are insignificant. The variation of these characteristics in the studied range did not cause any non-compliance with the discharge standard. However, the impacts of turbidity and TOC values are significant. The treated STWs-1 sewage sample could not meet the discharge standard when the turbidity and TOC values were increased by three and five times their original values. The STWs-2 sewage sample is less tolerant to the variation of sewage characteristics. The impacts of variations of ion concentration and pH on the treated sewage sample are insignificant. Inorganic ions of the solution can be adsorbed on the RGOFeNTFS surface, and hence occupying the reactive sites for photocatalytic reaction at the interface between the photocatalyst and sewage. As multivalent ions have a more robust adhesion to the photocatalyst surface than the monovalent ions, inorganic ions such as Ca^{2+} , Mg^{2+} and PO_4^{4-} may induce a more adverse effect on photocatalytic efficiency than ions like Na^+ (Marugán et al., 2010; Levchuk et al., 2019). With pH increases or DO decreases, the treated STWs-2 sewage sample could not meet the discharge standard after photocatalytic disinfection treatment. Compliance with the discharge standard was also affected by the increase of TOC and turbidity, which were two and five times their original value. Moreover, the increases of anions and cations were found to be important

factors that prevented the treated STWs-2 sewage sample from meeting the discharge standard. The photocatalytic disinfection of the STWs-1 sewage sample is generally applicable under a wider variation of sewage constituents than the STWs-2 sewage sample.

The suspended solids and organic matter, which affect the turbidity, may cause light scattering and light absorption in the solution. Sewage with high turbidity can promote the loss of incident light to the surface of the photocatalysts and shield the bacteria, resulting in a reduction of photocatalytic disinfection efficiency (Arya and Philip, 2014; García-Fernández et al., 2015). In addition, organic matter, reflected by TOC value, has a higher opportunity to approach reactive species and competes with bacteria for these reactive species during disinfection, inhibiting the disinfection efficiency as a consequence (García-Fernández et al., 2015). As a precursor of reactive species, the content of dissolved oxygen in sewage affects reactive species' production and consequently the disinfection potential (Chong et al., 2010). Ions have an affinity for the photocatalyst surface and can vary the properties of both photocatalyst's and bacterial surfaces. Consequently, interaction between photocatalyst materials and bacteria can then impact on the photocatalysis efficiency (Marugán et al., 2010; Levchuk et al., 2019).

3.5. Bioassay using phytoplankton for toxicity assessment of RGOFeNTFS

After photocatalytic disinfection treatment, the treated sewage is discharged to marine water. Photocatalyst materials are possibly present in the sewage discharge, and then in the marine environment. Considering a magnetic separation efficiency of higher than 99%, and the dosage of 1 and 3 g/L of RGOFeNTFS in the photocatalytic disinfection of STWs-1 and STWs-2 samples, respectively, the possible RGOFeNTFS concentration in the effluent discharge is 0.005 and 0.015 g/L. Effects of RGOFeNTFS on the marine environment is an important consideration, regarding the practical application of photocatalytic disinfection. In addition, a concentration of RGOFeNTFS of 2 g/L was selected for the toxicity assessment to understand the effect of RGOFeNTFS even under any extreme situation. Therefore, the concentrations of photocatalyst added to the seawater in the study were 0.005 g/L and 2 g/L.

The impacts of RGOFeNTFS on phytoplankton were determined by comparing the phytoplankton densities of the samples with the photocatalyst addition to that of the control sample (without any addition). As shown in Figure 8, in the control sample without addition, the phytoplankton density gradually decreased from 4.77 to 4.37-log cell

Table 2. Effects of sewage characteristics on photocatalytic disinfection.

		Types of sewage	Values of characteristics				
pH		STWs-1	√ (4.0)	√ (5.8)	√ (7.5)	-	
		STWs-2	√ (5,3)	√ (7.3)	√ (8.5)		
Dissolved oxygen (DO)		DO (mg/L)	~4.0	~6.0	~8.5		
		STWs-1	√	√	√		
		STWs-2	X	X	√		
			2x Origin value	3x Origin value	5x Origin value	10x Origin value	
TOC (as C mg/L, adjusted by adding NOM)		STWs-1	√	X	-		
		STWs-2	X	X			
Turbidity (NTU, concentrated the suspended solids by filtration)		STWs-1	-			√	X
		STWs-2				X	X
Ion concentration (mg/L)	Cl-	STWs-1	√	-	√	√	
		STWs-2			√	√	
	NO ₃ -	STWs-1			√	√	
		STWs-2			√	√	
	SO ₄ ²⁻	STWs-1			√	√	
		STWs-2			√	X	
	PO ₄ ³⁻	STWs-1			√	√	
		STWs-2			X	X	
	K ⁺	STWs-1			√	√	
		STWs-2			X	X	
	Ca ²⁺	STWs-1			√	√	
		STWs-2			X	X	
	Mg ²⁺	STWs-1			√	√	
		STWs-2			X	X	
Note STWs-1: low-salinity sewage after tertiary treatment STWs-2: low-salinity sewage after secondary biological treatment √ - the treated sewage sample meets the discharge standard (<i>E. coli</i>) X - the treated sewage sample does not meet the discharge standard (<i>E. coli</i>)							

number/mL after 48 hours, due to the natural decay of the microorganisms. After 48 hours, no significant difference in the phytoplankton density was observed in comparing the test samples (with additions of 0.005 g/L and 2 g/L RGOFeNTFS) with the control sample, indicating that the addition of RGOFeNTFS had an insignificant effect on the phytoplankton. The presence of RGOFeNTFS in the seawater is thus shown to be safe to the marine environment, even under the extreme situation in which the magnetic separation fails and the photocatalyst material accumulates to a concentration of 2 g/L. The nontoxicity of RGOFeNTFS to the marine environment shows the high potential of using RGOFeNTFS for long-term applications.

Figure 8. Effects of the presence of RGOFeNTFS on the phytoplankton density in seawater (collected on 12 March 2019; samples kept in the dark at room temperature).

4. Conclusions and engineering implications

The potential of the solar-energy-driven RGOFeNTFS for photocatalytic disinfection of real sewage under simulated solar light was evaluated. The potential of the technology for treating sewage is in line with the sewage quality (from good to poor): STWs-1 --> STWs-2 --> STWs-3 --> STWs-4, regarding the reaction time needed to meet the *E. coli* discharge standards. The technology shows potential for treating STWs-1 and STWs-2 (which have better water quality than STWs-3 and STWs-4), for which a relative short reaction time of around 20 minutes and 45 minutes are required, respectively. After the photocatalytic disinfection stopped, no bacterial regrowth was observed in these treated sewage samples. Moreover, as indicated by the natural-born phytoplankton in seawater, negligible effects of RGOFeNTFS on phytoplankton were observed after 48 hours even under extreme conditions (2 g/L of RGOFeNTFS), demonstrating the nontoxicity of RGOFeNTFS to the marine environment.

The implications of the study are that photocatalytic disinfection using RGOFeNTFS under solar light has the potential for practical application in sewage disinfection. Firstly, the retention time required for the treatment is acceptable, comparable to conventional methods such as chlorination. Secondly, photocatalytic disinfection using RGOFeNTFS is driven by green and renewable energy, specifically solar light. This property makes it a sustainable and environmental-friendly technology, as it saves energy and reduces carbon emissions during sewage treatment, and avoids the energy-intensive drawbacks of conventional treatment methods such as UV disinfection. More importantly, bacterial regrowth, found in UV disinfection, is not shown for photocatalytic disinfection using RGOFeNTFS. Thirdly, the safety of the photocatalyst material has been indicated by phytoplankton, which can rapidly respond to toxic materials in a marine environment. This indicates the safety of the technology's application. Moreover, due to its stability, the photocatalyst can be recycled in practical application, and consequently the cost of material is low in long-term treatment. Regarding these advantages, photocatalytic disinfection using RGOFeNTFS with further development and enhancement is feasible for sewage disinfection in an engineering aspect.

On the other hand, the effects of the sewage characteristics are crucial to the practicality of photocatalytic disinfection. The potential of the technology for treating sewage with poor water quality is still limited. The applicability of the technology for treating sewage with better water quality has higher potential, regarding the disinfection efficiency and retention time. Since suspended matter (turbidity) and organic matter (TOC) significantly affect the photocatalytic disinfection efficiency, they should be carefully examined when considering the practical application of photocatalytic disinfection. In addition, the reactor footprint should be an important consideration for the application of the technology. Moreover, as this technology is driven by solar light, a system simulating solar energy should be considered for operation in the nighttime, or when solar light is inadequate for photocatalytic disinfection.

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Appendix Table 1. Characterisation of sewage samples collected from different STWs.

	STWs-1		STWs-2		STWs-3		STWs-4	
Parameter	20181009	20190509	20180907	20191018	20181003	20190905	20181002	20190508
pH	5.8 ± 0.0	6.64 ± 0.01	6.8 ± 0.1	7.29 ± 0.03	6.1 ± 0.0	7.25 ± 0.05	6.6 ± 0.1	6.50 ± 0.01
TC (mg/L)	3.0 ± 0.1	13.91 ± 0.12	15.6 ± 0.1	17.54 ± 0.30	21.0 ± 0.1	29.28 ± 1.54	83.8 ± 0.7	68.58 ± 23
DO (mg/L)	Not measured	6.81 ± 0.01	Not measured	6.26 ± 0.07	Not measured	7.78 ± 0.03	Not measured	0.71 ± 0.01
TOC (mg/L)	2.1 ± 0.1	4.34 ± 0.05	4.4 ± 0.1	6.32 ± 0.36	5.3 ± 0.0	6.25 ± 0.21	45.6 ± 0.3	28.41 ± 0.40
TN (mg/L)	20.9 ± 0.5	11.32 ± 0.24	4.1 ± 0.1	3.92 ± 0.06	8.3 ± 0.2	7.80 ± 0.79	30.7 ± 0.5	26.64 ± 0.34
TSS (mg/L)	0.9 ± 0.0	0.38 ± 0.03	0.7 ± 0.2	1.71 ± 0.01	5.8 ± 0.1	5.17 ± 0.07	130.0 ± 0.0	92.34 ± 8.01
Turbidity (NTU)	0.4 ± 0.0	0.20 ± 0.02	1.8 ± 0.2	1.69 ± 0.09	1.7 ± 0.2	1.11 ± 0.06	115 ± 2	54.83 ± 0.85
Cl ⁻ (mg/L)	35.8 ± 0.4	60.60 ± 0.11	410.5 ± 0.3	461.26 ± 11.27	5301.4 ± 8.3	3758.23 ± 4.56	8050.0 ± 141.4	9791.51 ± 16.36
NO ₃ ⁻ -N (mg/L)	19.2 ± 0.1	11.57 ± 0.05	3.0 ± 0.0	6.29 ± 0.02	4.6 ± 0.0	3.32 ± 0.02	2.7 ± 0.0	0.52 ± 0.06
PO ₄ ³⁻ -P (mg/L)	2.0 ± 0.1	3.18 ± 0.12	1.0 ± 0.0	2.30 ± 0.02	1.0 ± 0.0	0.86 ± 0.01	1.9 ± 0.0	1.08 ± 0.03
SO ₄ ²⁻ -S (mg/L)	4.8 ± 0.1	9.36 ± 0.01	19.3 ± 0.0	25.25 ± 0.01	206.8 ± 0.4	170.02 ± 2.76	260.1 ± 1.1	534.03 ± 7.35
Ca ²⁺ (mg/L)	35.8 ± 0.6	24.12 ± 0.21	30.8 ± 0.1	31.85 ± 0.16	106.1 ± 0.3	99.15 ± 2.77	135.2 ± 1.0	196.03 ± 2.11
K ⁺ (mg/L)	11.1 ± 0.7	17.74 ± 0.10	18.8 ± 0.0	18.60 ± 0.14	129.3 ± 0.2	88.98 ± 2.53	202.4 ± 4.0	187.93 ± 2.47
Mg ²⁺ (mg/L)	0.6 ± 0.2	3.00 ± 0.15	31.8 ± 0.0	37.13 ± 0.34	295.6 ± 0.5	254.73 ± 6.03	391.6 ± 3.2	576.70 ± 3.50
Conductivity (μS/cm)	Not measured	486 ± 4	Not measured	1909 ± 3	Not measured	11.86 ± 0.05	Not measured	24.72 ± 1.13
COD (mg/L)	Not measured	15 ± 3	Not measured	14 ± 2	Not measured	29 ± 3	Not measured	240 ± 5
<i>E. coli</i> (CFU/100 mL)	(5.1 ± 0.3) × 10 ³	(2.8 ± 0.4) × 10 ³	(8.0 ± 0.1) × 10 ⁴	(1.10 ± 0.14) × 10 ⁵	(2.4 ± 0.1) × 10 ⁵	(6.20 ± 0.95) × 10 ⁵	(3.0 ± 0.5) × 10 ⁷	(7.7 ± 0.6) × 10 ⁷
Discharge standard of <i>E. coli</i> (CFU/100 mL)	1.5 × 10 ³	1.5 × 10 ³	1.5 × 10 ³	1.5 × 10 ³	1.5 × 10 ⁴	1.5 × 10 ⁴	9.0 × 10 ⁵	9.0 × 10 ⁵
Note: STWs-1: low-salinity sewage after tertiary treatment STWs-2: low-salinity sewage after secondary biological treatment STWs-3: high-salinity sewage after secondary biological treatment STWs-4: high-salinity sewage after chemically enhanced primary treatment								

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References

- [1] APHA (2017). *Standard Methods for the Examination of Water and Wastewater 2540 D*. American Water Works Association, Water Environment Federation.
- [2] Arya V and Philip L (2014). Visible and solar light photocatalytic disinfection of bacteria by N-doped TiO₂. *Water Science and Technology: Water Supply*, 14, pp. 924-930.
- [3] Asahi R, Morikawa T, Irie H and Ohwaki T (2014). Nitrogen-doped titanium dioxide as visible-light-sensitive photocatalyst: designs, developments, and prospects. *Chemical Reviews*, 114, pp. 9824-9852.
- [4] Chong M N, Jin B, Zhu H and Saint C (2010). Bacterial inactivation kinetics, regrowth and synergistic competition in a photocatalytic disinfection system using anatase titanate nanofiber catalyst. *Journal of Photochemistry and Photobiology A: Chemistry*, 214, pp. 1-9.
- [5] Dong H, Zeng G, Tang L, Fan C, Zhang C, He X and He Y (2015). An overview on limitations of TiO₂-based particles for photocatalytic degradation of organic pollutants and the corresponding countermeasures. *Water research*, 79, pp. 128-146.
- [6] García-Fernández I, Fernández-Calderero I, Polo-López M I and Fernández-Ibáñez P (2015). Disinfection of urban effluents using solar TiO₂ photocatalysis: a study of significance of dissolved oxygen, temperature, type of microorganism and water matrix. *Catalysis Today*, 240, pp. 30-38.
- [7] He J, Cheng J & Lo I M C (2021). Green photocatalytic disinfection of real sewage: efficiency evaluation and toxicity assessment of eco-friendly TiO₂-based magnetic photocatalyst under solar light. *Water Research*, 190, 116705.
- [8] He J, Zeng X, Lan S and Lo I M C (2019). Reusable magnetic Ag/Fe, N-TiO₂/Fe₃O₄@SiO₂ composite for simultaneous photocatalytic disinfection of E. coli and degradation of bisphenol A in sewage under visible light. *Chemosphere*, 217, pp. 869-878.
- [9] HKDSD (2020). *Effluent Quality of Major Sewage Treatment Works* [Online]. Hong Kong: Drainage Service Department. Available at: <https://www.dsd.gov.hk/TC/Files/tabledate/DSD_8STW_effluent_Mar2020_TCHI.pdf>.
- [10] HKEPD (1991). *Technical Memorandum Standards for Effluents Discharged into Drainage and Sewerage Systems, Inland and Coastal Waters* [Online]. Hong Kong. Available at: <https://www.elegislation.gov.hk/hk/cap358AK!en?xpid=ID_1438403048924_002&IN_DEX_CS=N>.
- [11] Khadgi N, Upreti A R and Li Y (2017). Simultaneous bacterial inactivation and degradation of an emerging pollutant under visible light by ZnFe₂O₄ co-modified with Ag and rGO. *RSC Advances*, 7, pp. 27007-27016.
- [12] Levchuk I, Homola T, Moreno-Andres J, Rueda-Márquez J J, Dizk P, Moríñigo M Á, Sillanpää M, Manzano M A & Vahala R (2019). Solar photocatalytic disinfection using ink-jet printed composite TiO₂/SiO₂ thin films on flexible substrate: applicability to drinking and marine water. *Solar Energy*, 191, pp. 518-529.
- [13] Li G, Nie X, Chen J, Wong P K, An T, Yamashita H and Zhao H (2016). Enhanced simultaneous PEC eradication of bacteria and antibiotics by facilely fabricated high-activity {001} facets TiO₂ mounted onto TiO₂ nanotubular photoanode. *Water Research*, 101, pp. 597-605.
- [14] Marug N J, Van Grieken R, Pablos C and Sordo C (2010). Analogies and differences between photocatalytic oxidation of chemicals and photocatalytic inactivation of microorganisms. *Water Research*, 44, pp. 789-796.
- [15] Miller R J, Lenihan H S, Muller E B, Tseng N, Hanna S K and Keller A A (2010). Impacts of metal oxide nanoparticles on marine phytoplankton. *Environmental Science & Technology*, 44, pp. 7329-7334.
- [16] Rueda-Marquez J J, Levchuk I, Fernández Ibáñez P and Sillanpää M (2020). A critical review on application of photocatalysis for toxicity reduction of real wastewaters. *Journal of Cleaner Production*, 258, 120694.
- [17] Sendra M, Sánchez-Quiles D, Blasco J, Moreno-Garrido I, Lubián, L M, Pérez-García S and Tovar-Sánchez A (2017). Effects of TiO₂ nanoparticles and sunscreens on coastal marine microalgae: ultraviolet radiation is key variable for toxicity assessment. *Environment International*, 98, pp. 62-68.
- [18] Wen X, Chen F, Lin Y, Zhu H, Yuan F, Kuang D, Jia Z and Yuan Z (2020). Microbial Indicators and Their Use for Monitoring Drinking Water Quality—A Review. *Sustainability*, 12(6), 2249.