



Research article

Antibacterial activities of gel-derived Ag-TiO₂-SiO₂ nanomaterials under different light irradiation

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Abstract: Gel-derived Ag-TiO₂-SiO₂ nanomaterials were prepared by sol-gel process to determine their disinfection efficiency under UV-C, UV-A, solar irradiations and in dark condition. The surface morphology and properties of gel-derived Ag-TiO₂-SiO₂ nanomaterials were characterized by X-ray diffraction (XRD), transmission electron microscopy (TEM) and BET specific surface area. The results showed that the average particle size of Ag-TiO₂-SiO₂ was around 10.9–16.3 nm. SiO₂ mixed with TiO₂ (the weight ratio of Si to Ti = 10:90) in the synthesis of Ag-TiO₂-SiO₂ by sol-gel process was found to increase the specific surface area of the obtained photocatalyst (164.5 m²g⁻¹) as compared with that of commercial TiO₂(P25) (53.1 m²g⁻¹). Meanwhile, Ag doped in TiO₂ (the mole ratio of Ag to TiO₂ = 1%) decreased the specific surface area to 147.3 m²g⁻¹. The antibacterial activities of gel-derived Ag-TiO₂-SiO₂ nanomaterials were evaluated by photocatalytic reaction against *Escherichia coli* bacteria (ATCC®25922). Ag-TiO₂-SiO₂ nanomaterials was observed to achieve higher disinfection efficiency than the catalyst without silver since both Ag nanoparticles and ions exhibit a strong antibacterial activity and promoted the e⁻ – h⁺ separation of TiO₂. The bactericidal activity of Ag-TiO₂-SiO₂ nanomaterial under light irradiation was superior to that under dark and only light. The reaction time to achieve a reduction by 6 log of bacteria of UV-C light alone and Ag-TiO₂-SiO₂ with UV-C light irradiation were 30 and 5 minutes, respectively. In addition, the superior synergistic effect of Ag-TiO₂-SiO₂ under both UV-A and solar light as compared to that

under UV-C counterpart could be ascribed to the red-shift of the absorbance spectrum of the Ag doped TiO₂-based catalyst.

Keywords: gel-derived Ag-TiO₂-SiO₂; photocatalyst; *E.coli* inactivation; different light irradiation

1. Introduction

Nowadays, photocatalysis is concerned as one of novel methods in provision of clean drinking water. Among various metal-oxide materials (TiO₂, ZnO, SnO₂, CeO₂ e.g.) being developed for photocatalytic applications, titanium dioxide (TiO₂), a semiconductor photocatalyst, has been widely used because it is relatively efficient, cheap, non-toxic, chemically and biologically inert, and high reactivity under UV light irradiation ($\lambda < 390$ nm). When anatase TiO₂ is exposed to UV light, holes (h_{vb}^+) and excited electrons (e_{cb}^-) are generated. The holes are trapped by water (H₂O) or hydroxyl groups (OH⁻) adsorbed on the surface to generate hydroxyl radicals (OH^{*}) [1,2] which are a powerful and indiscriminate oxidizing agents for bactericidal disinfection [3].

However, due to the wide band gap of the TiO₂ (~3.2 eV), it can only be applied in the UV irradiation which was ~5% of the solar energy, while the visible light contains about 45% of the solar energy. Therefore, to overcome this difficulty, many studies have been done to investigate the deposition of transition metal ions, i.e. Pt, Au, Pd, Ni, Cu, and Ag ions, into TiO₂ as electron acceptors. The results showed that this is an effective way to improve the TiO₂ photocatalytic activity under UV irradiation [4–7] and to improve its visible light sensitivity [8]. Among these ions, Ag⁺ is getting more attention than others because of its capability: (1) to bind, damage, and alter the functionalities of bacterial cell wall membrane, which are slightly negative [9–12]; (2) to interact with the thiol groups of proteins that are important for the bacterial respiration and the transport of significant substances through the cell [13]; and (3) to activate visible light excitation of TiO₂ [14]. As a result, Ag doped TiO₂ highly improved photocatalytic inactivation of bacteria [15,16]. In addition, it is generally believed that Ag nanoparticles enhances photoactivity of TiO₂ by lowering the recombination rate of its photo-excited charge carriers and/or providing more surface area for adsorption [17].

Besides doping Ag⁺ into TiO₂ to increase disinfection efficiency, mixing SiO₂ with TiO₂ to increase the specific surface area of the TiO₂ photocatalyst was also studied in another previous study [18]. A high specific surface area can provide more reactive sites for silver deposition and photocatalytic reactions, which is beneficial to the effective disinfection.

From the aforementioned reasons, the combination of Ag, TiO₂ and SiO₂ in one material shows great promise as efficient and visible light response photocatalytic materials in future. Therefore, in this work, Ag and SiO₂ was doped in TiO₂ by a sol-gel process to determine their disinfection activities against *Escherichia coli* bacteria under UV, solar light irradiations and in dark condition, which was then compared to the corresponding activities of the gel-derived TiO₂-SiO₂ and the commercial TiO₂ Degussa-P25.

2. Material and Method

2.1. Chemical reagents

Tetra-iso propyl orthotitanate (TTIP, Merck, Germany), Tetra-ethylorthosilicate (TEOS, 98%, Merck, Germany), nitric acid (Merck, Germany) and silver nitrate (AgNO₃, Xilong, China) were selected for the preparation of gel-derived Ag-TiO₂-SiO₂ nanomaterials. Ethanol (Prolabo, France) and *iso*-propanol (Prolabo, France) were used as solvents in sol-gel method. Tryptic soy agar (TSA, Indian) was used as growth medium for *Escherichia coli* (*E. coli*, ATCC®25922).

2.2. Synthesis of Ag-TiO₂-SiO₂ catalysts

The detailed preparation procedures of gel-derived Ag-TiO₂-SiO₂ nanomaterials (Ag-TiO₂-SiO₂) are mentioned in our previous study [19]. Briefly, four solutions were prepared: the first solution, S1, was prepared by mixing a mixture of ethanol and *iso*-propanol (solvent mixture, volume ratio of ethanol and *iso*-propanol = 1:1) with water and nitric acid; the second one, S2, included the solvent mixture, water, and Tetra-ethylorthosilicate; the third solution, S3, was a mixture of the solvents and *tetra-iso* propyl orthotitanate (the TiO₂:SiO₂ weight ratio was controlled at 90:10 [18]); and the last one, S4, included the solvent mixture, water, nitric acid and silver nitrate (mol ratio of silver to TiO₂ is 1% [19]). The above solutions were then mixed and refluxed. The obtained sol-gel solution was hydrothermally treated in a lab-made autoclave at 150 °C for 10 h. Solvents were then removed at 50 °C using a vacuum evaporation to collect an aerogel. The aerogel was dried in an oven at 105 °C for 2 h and then calcined in static air at 400 °C for 2 h.

2.3. Characterization of catalysts

Crystallinity of catalyst was characterized by using X-ray diffraction (Rikagu, Cu K α). The catalyst particle size and morphology were obtained by using transmission electron microscopy (TEM, JEM 1400, JEOL, Japan). The BET specific surface area of catalyst was determined by nitrogen adsorption at 77 K using a Chembet 3000. In addition, the mean particle sizes can be calculated by the Scherrer's equation:

$$D = \frac{0.9\lambda}{\beta \cos \theta}$$

where $\lambda = 0.1541$ (nm), β is the half-diffraction angle (rad), θ is the half-peak width (degree), and D is the diameter of the crystalline particle (nm) [20].

2.4. Photocatalytic disinfection of *E. coli*

Photocatalytic disinfection experiments of *Escherichia coli* (*E. coli*, ATCC®25922) was conducted in a reactor containing 300 mL of approximately 10⁶ CFU (colony forming units) *E. coli* /mL under UV-C (15W, Aqua-Pro), UV-A irradiation (15W, Aqua-Pro), solar light (70 to 120 Klux) and in dark (without light) conditions. The concentration of catalyst used in this study was 0.2 g/L. The reaction solution was well mixed for 10 min by using a magnetic stirrer at approximately 200 rpm before the UV lamp was activated. The distance between the UV lamp and the surface of the

bacterial solution was 5 cm. For the photocatalytic experiments under solar light, the reactor was exposed under the sun from 10 am to 12 am. All experiments were stopped after 120 min. The antibacterial activity of the catalyst against *E.coli* was determined by quantitative evaluation methods. 1 mL of the treated solution was diluted with sodium chloride solution (NaCl 0.9%) to obtain the suitable environment for bacterial colonies. The diluted solution was then spread on tryptic soy agar (TSA) plates and incubated at 37 °C for 24 h before the bacterial colonies were counted. Survival of the bacterial population was calculated using the equation :

$$\text{Log survival ratio} = \text{Log} (N_t/N_0)$$

where N_0 represented the initial population and N_t represented the population after irradiation time (T). The obtained data were the average value of three parallel runs or the values showing the best fitting for an exponential reduction (independent experiments \pm standard error of the mean (S.E.M)).

3. Results and Discussion

2.5. Characterization of nanomaterials

Figure 1 shows the XRD patterns of $\text{TiO}_2\text{-SiO}_2$ and 1% $\text{Ag-TiO}_2\text{-SiO}_2$ nanomaterials, obtained by the sol-gel method. XRD patterns show that both $\text{TiO}_2\text{-SiO}_2$ and 1% $\text{Ag-TiO}_2\text{-SiO}_2$ nanoparticle are detected with anatase ($\alpha\text{-TiO}_2$) phase which is the favorable structure than its rutile phase. Anatase with better photocatalytic functional properties was identified as the primary crystalline phase in both samples with the peaks at 2θ of 25.3° , 37.8° , 48.2° , 55.04° , and 62.74° relating to (101), (004), (200), (105), (211), and (204) planes, respectively.

However, there is difference between the patterns of these samples. The order of descending crystallinity is observed from the anatase peaks of two XRD patterns in which $\text{TiO}_2\text{-SiO}_2$ is smaller than Ag doped $\text{TiO}_2\text{-SiO}_2$. XRD pattern of the 1% $\text{Ag-TiO}_2\text{-SiO}_2$ nanomaterials clearly exhibits peaks of the metallic silver at 2θ of 38.1° , 44.5° và 64.3° relating to (111), (200) và (220) diffraction peaks, respectively, while XRD pattern of $\text{TiO}_2\text{-SiO}_2$ shows there is no peak corresponding to Ag. The peaks of Ag in $\text{Ag-TiO}_2\text{-SiO}_2$ are not clear since the percentage of silver was too small.

The mean crystalline size of the $\text{TiO}_2\text{-SiO}_2$ and 1% $\text{Ag-TiO}_2\text{-SiO}_2$ nanomaterials were calculated at about 10.02 nm and 11.35 nm, respectively, based on the peak broadening analysis described by Scherrer's equation for the (101) peak [20]. The crystalline size of $\text{Ag-TiO}_2\text{-SiO}_2$ powder slightly increased as compared with that of $\text{TiO}_2\text{-SiO}_2$. These results indicate that during the drying and calcining process, Ag^+ ions were formed on crystal borders or spreading on the surface anatase grains, which led to the increase in the crystallite size of anatase phase [21,22].

It is found that the BET specific surface area of gel-derived TiO_2 prepared with sol-gel method and hydrothermal treatment ($82.7 \text{ m}^2\text{g}^{-1}$) was higher than that of commercial TiO_2 (P25) $53.1 \text{ m}^2\text{g}^{-1}$. Since the specific surface area has great effect on the photocatalytic activity of TiO_2 , combination of SiO_2 and TiO_2 with the promotion of specific surface area ($164.5 \text{ m}^2\text{g}^{-1}$) has demonstrated better photocatalytic activity than only gel-derived TiO_2 . The specific surface area of $\text{TiO}_2\text{-SiO}_2$ increased around twofold compared to that of bare TiO_2 catalyst. However, when Ag was doped in the $\text{TiO}_2\text{-SiO}_2$ nanomaterials, the specific surface area of obtained $\text{Ag-TiO}_2\text{-SiO}_2$ slightly decreased at $147.3 \text{ m}^2\text{g}^{-1}$. This result may be ascribed to the contribution of silver on the surface of $\text{TiO}_2\text{-SiO}_2$.

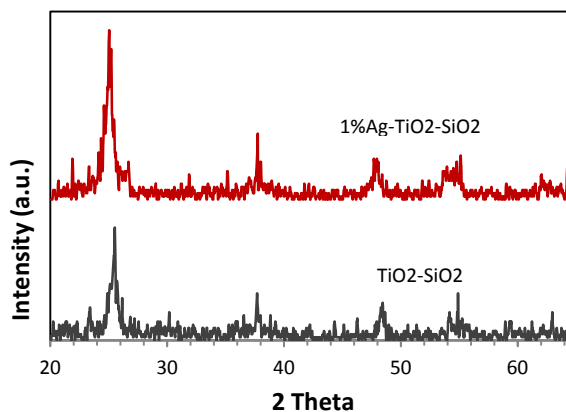


Figure 1. XRD patterns of $\text{TiO}_2\text{-SiO}_2$ and $1\%\text{Ag-TiO}_2\text{-SiO}_2$ powders prepared by Sol-gel method and calcined at $400\text{ }^\circ\text{C}$.

3.2. Photocatalytic antibacterial activity

3.2.1. Disinfection against *E.coli* bacteria under UV-C irradiation

Figure 2 shows the inactivation of *E.coli* under the irradiation of UV-C by using photocatalysts of TiO_2 (Degussa-P25), $\text{TiO}_2\text{-SiO}_2$ or $1\%\text{Ag-TiO}_2\text{-SiO}_2$ for 30 min. It is clear that Ag doped TiO_2 improved the antibacterial activities against *E.coli* in terms of inactivation efficiency and rate of reaction. Among three kinds of catalysts (commercial TiO_2 (P25), $\text{TiO}_2\text{-SiO}_2$ and $1\%\text{Ag-TiO}_2\text{-SiO}_2$), disinfection efficiency of $1\%\text{Ag-TiO}_2\text{-SiO}_2$ was the highest. In addition, the antibacterial efficiency of $1\%\text{Ag-TiO}_2\text{-SiO}_2$ was much higher than that of UV-C only. It took about 5, 10 and 15 minutes to reach 6-log(10) reduction of bacteria colonies by using $\text{Ag-TiO}_2\text{-SiO}_2$ nanomaterials, $\text{TiO}_2\text{-SiO}_2$, commercial TiO_2 (P25), respectively. Meanwhile, 30-minute illumination was required to get 5-log(10) inactivation by using UV-C only. This can be obviously explained that under UV illumination, anatase TiO_2 exposed to UV-C light generates energy-rich electron-hole pairs which are able to degrade cell components of microorganisms. Therefore, the antibacterial activity of commercial TiO_2 (P25) was approximately twice as high as that of UV-C only. On the other hands, SiO_2 mixed with TiO_2 in sol-gel process was observed to significantly increase the BET specific surface area of the gel-derived $\text{TiO}_2\text{-SiO}_2$ ($164.5\text{ m}^2\text{g}^{-1}$) as compared to that of commercial TiO_2 (P25) ($53.1\text{ m}^2\text{g}^{-1}$). The higher the specific surface area of photocatalyst the higher the exposed area between UV-C light and catalysts. This in turn increases the photo-active sites of the catalyst and therefore, improves its photocatalytic activity. As a consequence, the antibacterial activity of gel-derived $\text{TiO}_2\text{-SiO}_2$ was superior to that of commercial TiO_2 (P25) under UV-C irradiation.

In this present work, Ag was deliberately added in sol-gel preparation of $\text{TiO}_2\text{-SiO}_2$ nanomaterials to promote the effective disinfection of the obtained catalyst because Ag nanoparticles [23,24] and ions [25] were observed to exhibit a strong antibacterial activity. In addition, Ag has been proved to be able to modify TiO_2 by effective promotion the $e^- - h^+$ separation, thus enhancing the performance of photocatalytic antibacterial activity [14]. As a result, the $\text{Ag-TiO}_2\text{-SiO}_2$ catalyst showed superior photoactivity against *E.coli* as compared with the gel-derived $\text{TiO}_2\text{-SiO}_2$ counterpart under the same preparation condition.

It is interesting that the rates of reactions were extremely fast in first 5 minutes and there was no significant change in the rate of reactions after 5 minutes (Fig. 2).

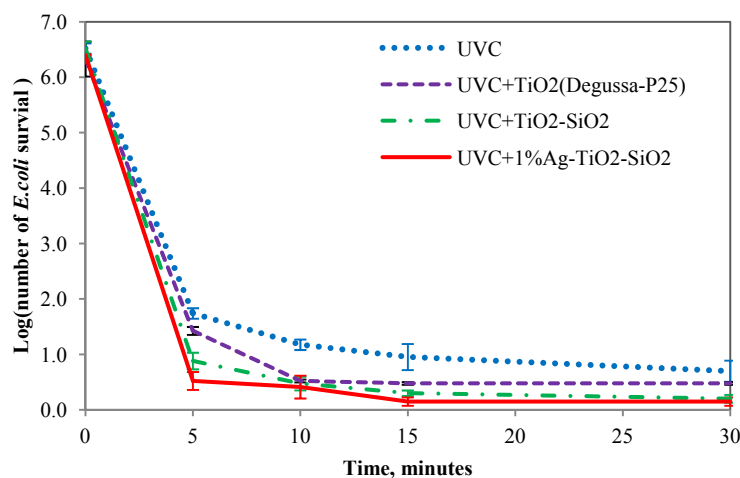


Figure 2. Inactivation effect against *E. coli* by Ag-TiO₂- SiO₂ photocatalysts under UV-C irradiation compared to TiO₂ (Degussa-P25) and TiO₂-SiO₂ (Data are shown as means of n=3 independent experiments ± S.E.M).

3.2.2. Disinfection of Ag-TiO₂-SiO₂ nanomaterials against *E. coli* under different irradiations

UV light is a well-known traditional disinfectant. Therefore, the photocatalytic properties of the catalysts were evaluated by conducting inactivation experiments of *E. coli* bacteria under UV irradiation for 120 minutes and compared to experiments under natural sunlight containing only around 5% of UV light. Based on the observed reduction rate of *E. coli* colonies in figure 3a, it could be seen that the inactivation efficiency under UV-C irradiation (5-log(10)) was extremely higher than those under UV-A irradiation (53.5%) or in dark (0%). It is evident that dark and UV-A were not sufficient to inactivate the bacteria, also confirmed by the previous studies [26,27]. However, after 120-minute exposure to solar light, 4-log(10) reduction of *E. coli* population was recorded while the reduction of viable bacteria was up to about 5-log(10) under 30-minute UV-C irradiation.

Silver has long been known to be one of the most effective doping metals to change the intrinsic band structure of TiO₂, and consequently, to improve its visible light sensitivity [28,29,30] as well as to increase its photocatalytic activity under UV irradiation [31,32,33]. Accordingly, the synergistic effect of UV/solar light and Ag-TiO₂-SiO₂ was also studied in this present work by adding 0.2 g/L of 1% Ag-TiO₂-SiO₂ in water artificially contaminated with pure bacterial culture (10⁶ CFU *E. coli*/mL) (Fig. 3b). As discussed above, UV-C light itself presented the superior disinfection efficiency as compared to other irradiation sources (Fig. 3a). Therefore, a combination of UV-C light with Ag-TiO₂-SiO₂ definitely performed the highest disinfection efficiency against *E. coli* as shown in Fig. 3b.

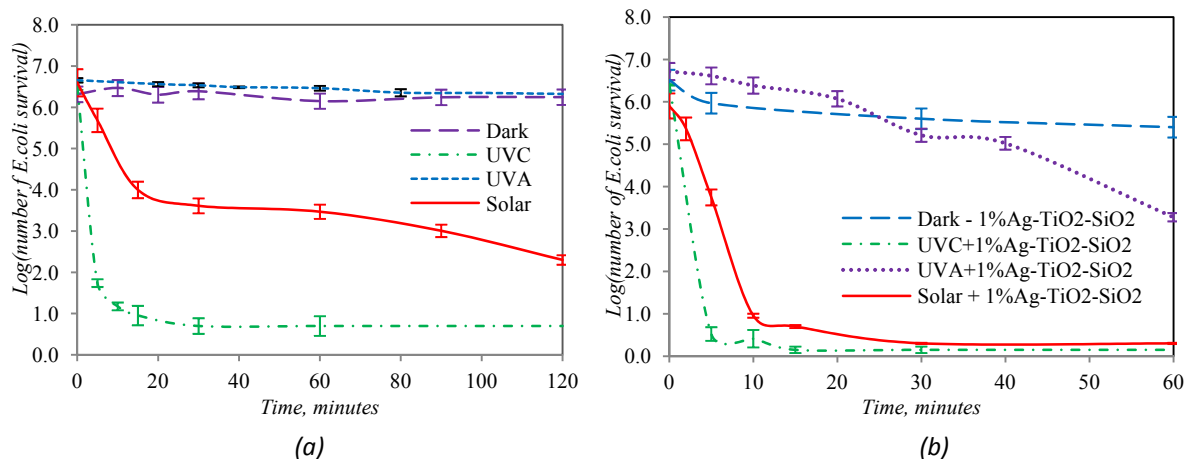


Figure 3. The disinfection efficiency against *E. coli* under different light sources with (a) and without (b) 1%Ag-TiO₂-SiO₂ photocatalysts (Data are shown as means of $n = 3$ independent experiments \pm S.E.M).

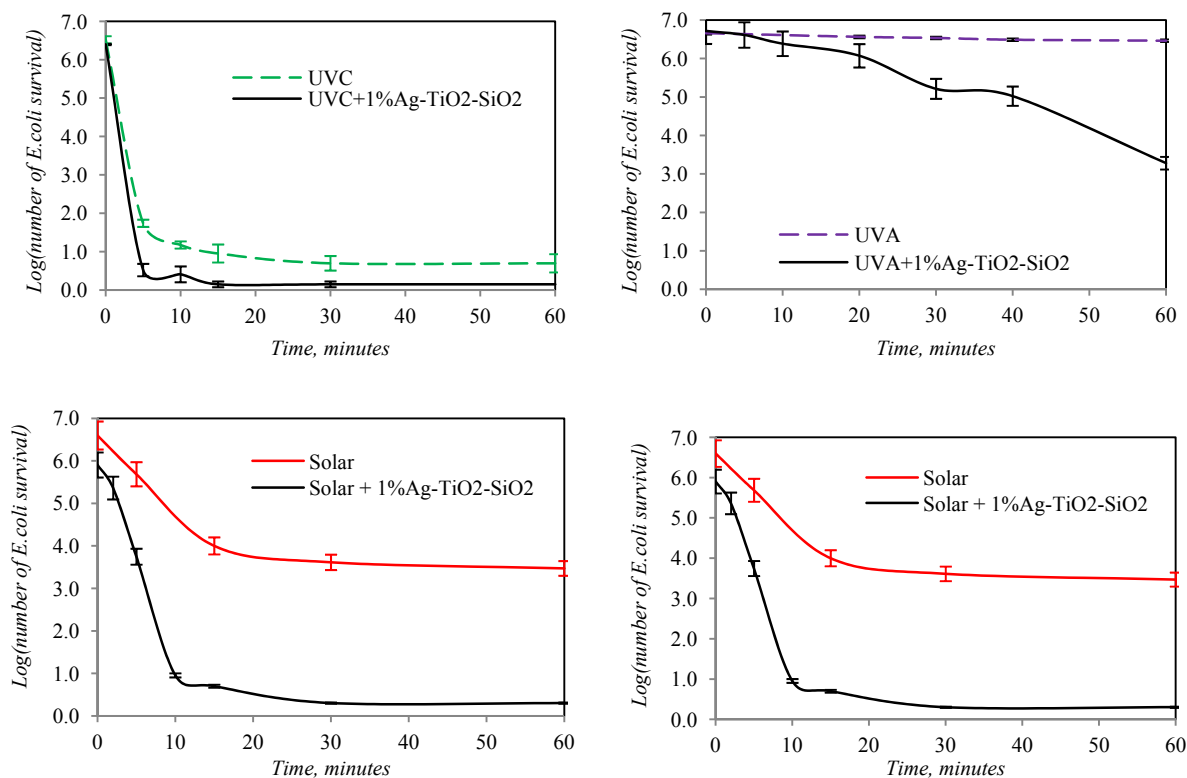


Figure 4. The disinfection efficiency against *E. coli* by 1%Ag-TiO₂-SiO₂ photocatalysts under (a) UV-C irradiation; (b) UV-A irradiation; (c) solar irradiation; and (d) dark condition (Data are shown as means of $n=3$ independent experiments \pm S.E.M).

To further clarify the synergistic effect of Ag-TiO₂-SiO₂ in combination with different light sources, disinfection efficiency of this catalyst against *E. coli* with and without light irradiation was compared and shown in Fig. 4. The presence of Ag-TiO₂-SiO₂ under UV-C was not observed to substantially improve the disinfection efficiency against *E. coli* of this catalyst (Fig. 4a). Meanwhile,

a combination of solar light and Ag-TiO₂-SiO₂ was found to significantly increase the disinfection efficiency as depicted in Fig. 4c. When exposed under natural sunlight irradiation, only 3-log₁₀ reduction of *E.coli* was observed while adding Ag-TiO₂-SiO₂ to the reactor was found to promote the antibacterial activity up to 6-log(10) within 30 minutes. The presence of Ag-TiO₂-SiO₂ under UV-A was not good as that under solar light. Nevertheless, the synergistic effect of such combination was higher than that of Ag-TiO₂-SiO₂ under UV-A irradiation. The superior synergistic effect of Ag-TiO₂-SiO₂ under both UV-A and solar light as compared to that under UV-C counterpart could be ascribed to the red-shift of the absorbance spectrum of the Ag doped TiO₂-based catalyst [27]. This catalyst was consequently more photoactive under UV-A and visible light than under UV-C light irradiation. Although the synergistic effect of Ag-TiO₂-SiO₂ under solar light was superior to that under UV-C, the disinfection efficiency of the latter was observed to be slightly higher than that of the former as shown in Fig. 4a and 4c. These results implied that either UV-C only as a low-cost method or a combination of Ag-TiO₂-SiO₂ with solar light as an energy-saving method could be efficient to inactivate bacteria.

For the inactivation experiments conducted in dark, the reduction rate of *E.coli* colonies by Ag-TiO₂-SiO₂ nanoparticles slightly enhanced (92.4%) after 60 minutes while no change was observed in experiments without catalyst (Fig. 4d). This result could be attributed to: (i) the adsorption of bacteria cells onto TiO₂ surfaces; (ii) the antibacterial activity of silver nanoparticles and ions [23,24,25]; (iii) the synergistic effect of Ag and TiO₂ in dark [19].

4. Conclusion

A feasible method was proposed to overcome the limitation of TiO₂ catalyst in antibacterial activity under sunlight irradiation by preparing the combination of Ag, TiO₂ and SiO₂ in one material. Ag-TiO₂-SiO₂ nanoparticle was found to be an effective catalyst in disinfection for water treatment. UV-A had the disinfection capability of 53.5%; however, when combined synergistically with 1% Ag-TiO₂-SiO₂, the efficiency could increase up to 99.96% in 30 min. In case of doing experiment under UV-C irradiation with Ag-TiO₂-SiO₂, bacterial could be removed completely within 5 min compared with 30 min under the condition of UV-C without catalyst and 10 min with the experiment of using UV-C-TiO₂-SiO₂. In the solar light irradiation, the effective reduction of the viable bacteria for the Ag-TiO₂-SiO₂ was measured 6-log(10) within 30 min. Therefore, Ag-TiO₂-SiO₂ nanomaterial shows great promise as an efficient and visible light response photocatalytic material in future.

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Conflict of Interest

Authors declare that there is not conflict of interest.

References

1. Hoffman MR, Martin ST, Choi W, et al. (1995) Environmental application of semiconductor photocatalysis. *Chem Rev* 95: 69–96.
2. Robertson P (1996) Semiconductor photocatalysis: An environmentally acceptable alternative production technique and effluent treatment process. *J Cleaner Prod* 4: 203–212.
3. Matsunga T, Tamada R, Wake H (1985) Photoelectrochemical sterilization of microbial-cells by semiconductor powders. *FEMS Microbiol Lett* 29: 211–214.
4. Rengaraj S, Li XZ (2006) Enhanced photocatalytic activity of TiO₂ by doping with Ag for degradation of 2,4,6-trichlorophenol in Aqueous Suspension. *J Mol Catalysis A* 243: 60–67.
5. Katsumata H, Sada M, Nakaoka Y, et al. (2009) Photocatalytic degradation of diuron in aqueous solutions of platinized TiO₂. *J Hazard Mater* 171: 1081–1087.
6. Kalathil S, Khan MM, Banerjee AN, et al. (2012) A simple biogenic route to rapid synthesis of Au@TiO₂ nanocomposites by electrochemically active biofilms. *J Nanoparticle Res* 14: 1051–1059.
7. Fang J, Cao S-W, Wang Z, et al. (2012) Mesoporous plasmonic Au–TiO₂ nanocomposites for efficient visible-light-driven photocatalytic water reduction. *Int J Hydrogen Energy* 37: 17853–17861.
8. Yu C, Cai D, Yang K, et al. (2010) Sol-gel derived S, I-codoped mesoporous TiO₂ photocatalyst with high visible-light photocatalytic activity. *J Phys Chem Solids* 71: 1337–1343.
9. Yuranova T, Rincon AG, Bozzi A, et al. (2003) Antibacterial textiles prepared by RF-plasma and vacuum-UV mediated deposition of silver. *J Photochem Photobiol* 161: 27–34.
10. Grieken RV, Marugán J, Sordo C, et al. (2009) Photocatalytic inactivation of bacteria in water using suspended and immobilized silver-TiO₂. *Environmental* 93: 112–118.
11. Sangchaya W, Sikonga L, Kooptarnond K (2012) Comparison of photocatalytic reaction of commercial P25 and synthetic TiO₂-AgCl nanoparticles. *Procedia Eng* 32: 590–596.
12. Ubonchonlakate K, Sikong L, Tontai T, et al. (2011) *P. aeruginosa* Inactivation with silver and nickel doped TiO₂ films coated on glass fibre roving. *Adv Mater Res* 150–151: 1726–1731.
13. Cho KH, Park JE, Osaka T, et al. (2005) The study of antimicrobial activity and preservative effects of nanosilver ingredient. *Electrochim Acta* 51: 956–960.
14. Akhavan O, Ghaderi E (2009) Enhancement of antibacterial properties of Ag nanorods by electric field. *Sci Technol Adv Mater* 10: 015003.
15. Li M, Noriega-Trevino ME, Nino-Martinez N, et al. (2011) Synergistic Bactericidal Activity of Ag-TiO₂ nanoparticles in Both Light and Dark Conditions. *Environ Sci Technol* 45: 8989–8995.
16. Thiel J, Pakstis L, Buzby S, et al. (2007) Antibacterial properties of silver-doped. *Small* 3: 799–803.
17. Scafani A, Palmisano L, Schiavello M (1990) Influence of the preparation methods of titanium dioxide on the photocatalytic degradation of phenol in aqueous dispersion. *J Phys Chem* 94: 829–832.
18. Viet-Cuong N, The-Vinh N (2009) Photocatalytic decomposition of phenol over N-TiO₂-SiO₂ catalyst under natural sunlight. *J Exp Nanosci* 4: 233–242.
19. Hoang T-TN, Suc NV, Nguyen T-V (2015) Bactericidal activities and synergistic effects of Ag–TiO₂ and Ag–TiO₂–SiO₂ nanomaterials under UV-C and dark conditions. *Int J Nanotechnol* 12: 367–379.

20. Sun B, Sun S-Q, Li T, et al. (2007) Preparation and antibacterial activities of Ag-doped SiO₂-TiO₂ composite films by liquid phase deposition (LPD) method. *J Mater Sci* 42: 10085–10089.
21. Chao HE, Yuan YU, Xingfanga HU (2003) Effect of silver doping on the phase transformation and grain growth of sol-gel titania powder. *J Eur Ceramic Soci* 23: 1457–1464.
22. Oliveri G, Ramis G, Busca G, et al. (1993) Thermal stability of vanadia-titania catalysts. *J Mater Chem* 3: 1239–1249.
23. Shahverdi AR, Fakhimi A, Shahverdi HR, et al. (2007) Synthesis and effect of silver nanoparticles on the antibacterial activity of different antibiotics against *Staphylococcus aureus* and *Escherichia coli*. *Nanomedicine* 3: 168–171.
24. Smetana AB, Klabunde KJ, Marchin GR, et al. (2008) Biocidal activity of nanocrystalline silver powders and particles. *Langmuir* 24: 7457–7464.
25. Yamanaka M, Hara K, Kudo J (2005) Bactericidal actions of a silver ion solution on *Escherichia coli*, studied by energy-filtering transmission electron microscopy and proteomic analysis. *Appl Environ Microbiol* 71: 7589–7593.
26. Benabbou AK, Derriche Z, Felix C, et al. (2007) Photocatalytic inactivation of *Escherichia coli*. Effect of concentration of TiO₂ and microorganism, nature, and intensity of UV irradiation. *Appl Catal B Environ* 76: 257–263.
27. Hassan Y, Ishtiaq AQ, Imran H, et al. (2013) Visible light photocatalytic water disinfection and its kinetics using Ag-doped titania nanoparticles. *Environ Sci Pollut Res* 21: 740–752.
28. Anpo M, Kishiguchi S, Ichihashi Y, et al. (2001) The design and development of second-generation titanium oxide photocatalysts able to operate under visible light irradiation by applying a metal ion-implantation method. *Res Chem Intermediat* 27: 459–467.
29. Chen X, Lou Y, Samia ACS, et al. (2003) Coherency Strain Effects on the Optical Response of Core/Shell Heteronanostructures. *Nano Lett* 3: 799–803.
30. Park CH, Zhang SB, Wei SH (2002) Origin of *p*-type doping difficulty in ZnO: The impurity perspective. *Phys Rev* 66: 073202.
31. Choi W, Termin A, Hoffmann M (1994) The Role of Metal Ion Dopants in Quantum-Sized TiO₂: Correlation between Photoreactivity and Charge Carrier Recombination Dynamics. *J Phys Chem* 98: 13669–13679.
32. Mu W, Herrmann JM, Pichat P (1989) Room temperature photocatalytic oxidation of liquid cyclohexane into cyclohexanone over neat and modified TiO₂. *Catal Lett* 3: 73–84.
33. Duonghong D, Borgarello E, Gratzel M (1981) Dynamics of light-induced water cleavage in colloidal systems. *J Am Chem Soc* 103: 4685–4690.



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