Research Article

The Effect of Molar Ratios of Ti/Si on Core-Shell SiO₂@TiO₂ Nanoparticles for Photocatalytic Applications

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After the core-shell SiO₂@TiO₂ nanoparticles (CSTNs) were synthesized by hydrothermal method, we investigated the influence of different molar ratios of Ti/Si on morphology, structure, and photocatalytic activity of the CSTNs. It was found that the CSTNs showed different size and surface morphology as the Ti/Si molar ratio changed. Besides, the TiO₂ and the CSTN had the anatase phase after hydrothermal process and calcination at 450°C for 2 h. The N₂ adsorption-desorption isotherms demonstrated the CSTNs with the molar ratio of Ti/Si increased from 1:1 to 8:1 can be categorized as type IV with hysteresis loop of type H2 and showed to be mesoporous materials. In addition, the CSTNs with the Ti/Si molar ratio of 5:1 had the highest surface area of 176.79 m²/g. Surface charges showed the isoelectric point (IEP) of the CSTNs ranged between silica (IEP at pH 3.10) and titania (IEP at pH 5.29). Since the molar ratio of Ti/Si increased from 1:1 to 8:1 by degrading both colorless organic pollutant of phenol and colored substances of methylene blue (MB) under UV irradiation, the photocatalytic activity of CSTNs exhibited higher photodegradation efficiency compared with TiO₂. What is more, the experimental results also showed the CSTNs with Ti/Si molar ratio of 5:1 had the highest photocatalytic activity and showed higher photocatalytic efficiency compared with other TiO₂-SiO₂ composites reported for photodegradation of phenol and MB.

1. Introduction

Titanium dioxide (TiO₂), one of the most used photocatalysts for photodegrading inorganic and organic pollutants, has the advantages of low cost, chemical stability, low toxicity, high physical, easy availability, and excellent photoactivity [1–3]. The major drawbacks of TiO₂ arise from the wide band gap (3.2 eV for anatase) and rapid charge recombination of the electron-hole pairs, which restrict the light absorption (just ultraviolet region with wavelength < 390 nm) and suppressing the quantum efficiency [4–6]. In addition, the physical properties of the TiO₂ such as nanoparticles' agglomeration, phase transformation, morphology, and particle size will also limit the catalytic efficiency and practical applications [7–9].

In order to conquer these problems, core-shell nanoparticles with titania as shell have been studied for photocatalytic degradation due to their stability, dispersibility, and higher

surface area, reducing the recombination efficiency of electron hole and improving the quantum efficiency of photocatalyst compared to either the core or the shell materials [10–12]. The core nanoparticles include SiO₂, polystyrene (PS), ZrO₂, and Fe₂O₃ [13–15]. SiO₂, due to its stable surface chemistry, low cost, mechanical stability, and facilitation of good dispersion of TiO₂, is one of the significant core materials for synthesizing the core-shell nanoparticles [16]. Besides, SiO₂ can be easily and controllably prepared by the Stöber method [17].

Some researchers had focused their attention on photocatalytic application of the core-shell SiO₂@TiO₂ nanoparticles (CSTNs) with SiO₂ as a core and TiO₂ as a shell, and CSTNs showed better photocatalytic performance and improved photoactivity than pure TiO₂ for improved adsorption, large surface area, and suitable porous structure [12, 18–21]. However, the research that has been reported mainly focus on the morphology of the CSTNs or

how to control the uniform and spherical in shape, and just a few different weights or molar ratios of Ti/Si have been synthesized for photocatalysis with CSTNs. For example, Ullah et al. [10] synthesized core@shell $\text{SiO}_2/\text{TiO}_2$ nanoparticles with four of different weight proportions of titanium (IV) isopropoxide (TTIP) (10.4%, 20.6%, 31%, and 35%) towards degradation of crystal violet (CV). Kitsou et al. [11] produced $\text{SiO}_2/\text{TiO}_2$ core-shell nanospheres with only three weight ratios of titanium (IV) isopropoxide (TTIP): SiO_2 (1:1, 2:1, and 3:1) for NO_x degradation. Besides, few works indicated the photocatalytic applications for both colorless organic pollutants (such as phenol) and colored substances (such as methylene blue).

In this paper, the different molar ratios of the CSTNs (seven different molar ratios of Ti/Si = 1:1, 1:2, 1:5, 1:8, 2:1, 5:1, and 8:1) were presented with SiO_2 as a core and TiO_2 as a shell by hydrothermal method. The surface morphology and physical properties of the CSTNs were investigated by transmission electron microscopy (TEM), scanning electron microscopy (SEM), X-ray diffraction (XRD), Brunauer-Emmett-Teller (BET) specific surface, and surface charges. It was found that high photocatalytic efficiency was shown with low photocatalyst addition by studying the photocatalytic activity of the CSTNs for both colorless organic pollutants (such as phenol) and colored substances (such as methylene blue).

2. Experimental

2.1. Chemicals. Titanium (IV) isopropoxide 95% (TTIP, Sigma-Aldrich), tetraethyl orthosilicate 99.9% (TEOS, Xiyi Reagent Company), isopropanol, ethanol, and NH_4OH (25%) were used in the synthesis process of CSTNs. KCl, KOH, and HCl (Sinopharm Chemical Reagent Company) were used in the test of surface charges. Phenol (Sinopharm Chemical Reagent Company) and methylene blue (MB, Sinopharm Chemical Reagent Company) were employed as the substrates for photocatalytic applications.

2.2. Synthesis of SiO_2 Core Spheres. SiO_2 core spheres were prepared using the Stöber method by hydrolysis and condensation of tetraethyl orthosilicate (TEOS) [10]. Firstly, 4 mL NH_4OH and 15 mL H_2O were added to 100 mL ethanol and stirring for 30 min in a Teflon reactor. Secondly, 3.0 mL of TEOS was quickly added to the above mixture under continuous stirring for 3 h at room temperature ($25 \pm 2^\circ\text{C}$). Thirdly, the mixture was neutralized with 5 mol L^{-1} HCl and centrifuged at 4000 rpm for 10 min. Finally, the SiO_2 core spheres were washed four times with ethanol and distilled water, then dried at 70°C for at least 20 h to obtain SiO_2 core spheres.

2.3. Synthesis of Core-Shell $\text{SiO}_2/\text{TiO}_2$ Nanoparticles (CSTNs). The CSTNs with different molar ratios of Ti/Si were synthesized according to the literatures with minor modifications [10]. The process is as follows: different weight of SiO_2 core spheres were dried at 110°C for 1 h and then sonicated in 80 mL isopropanol for 1 h; then, different volumes of titanium (IV) isopropoxide (TTIP) was quickly added with vigorous stirring for 24 h (different molar ratios of Ti/Si were

1:1, 2:1, 5:1, 8:1, 1:2, 1:5, and 1:8). Subsequently, 15 mL water-alcohol mixture (5 mL H_2O :10 mL isopropanol) was slowly added (2 mL min^{-1}) to the above mixture and stirred for 3 h. The resulting precipitates were washed once with isopropanol and twice with deionized water at 8000 rpm. Then, the amorphous CSTNs were suspended in 50 mL H_2O and processed to hydrothermal treatment at 105°C for 24 h, then centrifuged at 8000 rpm for 10 min. Finally the obtained CSTNs were dried at 70°C for 20 h and calcined at 450°C for 2 h. The unsupported TiO_2 was also synthesized using 3 mL of TTIP by the same above procedures without SiO_2 in the mixture.

2.4. Characterization Methods. The size and surface morphology of the synthesized nanoparticles were examined using scanning electron microscopy (SEM) (JSM-6701F, Japan) and transmission electron microscopy (TEM) (Tecnai G², American). The crystalline structure of the samples was investigated by X-ray diffraction (XRD) using Cu-K α radiation at 40 kV and 30 mA. The specific surface area and pore size of the samples were estimated from nitrogen adsorption curves using the Brunauer-Emmett-Teller (BET) method employing ASAP 2020 (Micromeritics Instrument Corporation, USA) surface area analyzer. The pore size distribution was determined based on using the Barrett-Joyner-Halenda (BJH) model by the same instrument.

The surface charges of the samples were measured using an electrophoresis instrument (Model ZEN3690, Malvern, UK). The samples were dispersed in a 0.003 mol L^{-1} KCl electrolyte solution to give a final concentration of 0.01% (w/v). The pH of the suspension was varied by adding 0.01 mol L^{-1} HCl or 0.01 mol L^{-1} KOH to the suspension, and the isoelectric point (IEP) was taken at which the zeta potential was zero. The apparent surface coverage (ASC) of TiO_2 -coated SiO_2 nanoparticles can be calculated from the IEP data of the components by the following equation [22]:

$$\% \text{ ASC} = \frac{M_{\text{TiO}_2} (\text{IEP}_{\text{SiO}_2} - \text{IEP}_{\text{Ti/Si}})}{[M_{\text{SiO}_2} (\text{IEP}_{\text{Ti/Si}} - \text{IEP}_{\text{TiO}_2}) - M_{\text{TiO}_2} (\text{IEP}_{\text{Ti/Si}} - \text{IEP}_{\text{SiO}_2})]}, \quad (1)$$

where M_{TiO_2} and M_{SiO_2} were the molecular weights of TiO_2 and SiO_2 , respectively, and the subscript Ti/Si referred to the different molar ratios of Ti/Si nanoparticles, respectively.

2.5. Photocatalytic Activity Experiments. The photocatalytic activity of the prepared samples was evaluated by measuring the photodegradation of phenol and methylene blue (MB) in a reactor under UV light irradiation (high-pressure mercury lamp with 500 W) for 120 minutes. In a typical experiment, 75 mg of prepared photocatalysts were dispersed in 300 mL phenol or MB aqueous solution with initial concentration of 20 mg/L. The mixture was first stirred in the dark for 30 minutes to achieve adsorption-desorption equilibrium and then was carried out under UV light. 4 mL of the solution was collected from the reactor at different irradiation time intervals. The collected samples were centrifuged to separate the suspended

FIGURE 1: SEM images of (a) SiO_2 , (b) $\text{Ti/Si} = 1 : 2$, (c) $\text{Ti/Si} = 2 : 1$, (d) $\text{Ti/Si} = 1 : 5$, and (e, f) $\text{Ti/Si} = 5 : 1$.

FIGURE 2: TEM images of (a) SiO_2 , (b) TiO_2 , (c) $\text{Ti/Si} = 1 : 5$, and (d) $\text{Ti/Si} = 5 : 1$.

particles. The concentration changes of phenol were analyzed by colorimetric method of 4-amino antipyrine at 510 nm, and the MB were analyzed by recording the maximum absorbance of MB at 664 nm.

3. Results and Discussion

3.1. Morphological Characterization. The morphology of the core-shell $\text{SiO}_2@\text{TiO}_2$ nanoparticles (CSTNs) was

FIGURE 3: Particle size distributions of (a) TiO_2 , (b) SiO_2 , (c) $\text{Ti/Si} = 1 : 5$, (d) $\text{Ti/Si} = 1 : 2$, (e) $\text{Ti/Si} = 2 : 1$, and (f) $\text{Ti/Si} = 5 : 1$.

characterized by scanning electron microscopy (SEM) and transmission electron microscopy (TEM) in Figures 1 and 2. As shown in Figures 1(a) and 2(a), the uncoated SiO_2 particles showed smooth and spherical shape. The morphology of $\text{Ti/Si} = 1 : 2$ (Figure 1(b)) and $\text{Ti/Si} = 1 : 5$ (Figures 1(d) and 2(c)) had rough and textured surfaces compared with the SiO_2 core particles, which confirmed that the titania was coated on SiO_2 core particle [23]. Besides, the uniform coating of silica in $\text{Ti/Si} = 1 : 2$ and $\text{Ti/Si} = 1 : 5$ also indicated that less titania molar content was conducive to the formation of core-shell structure by

the hydrolysis-polycondensation process [24]. In contrast, the morphology of $\text{Ti/Si} = 2 : 1$ (Figure 1(c)) and $\text{Ti/Si} = 5 : 1$ (Figures 1(e) and 1(f)) had a rougher and more textured surface which can also be shown from TEM in Figures 2(c) and 2(d). Especially from the SEM and TEM morphology of $\text{Ti/Si} = 5 : 1$, the free or coreless TiO_2 nanoparticles agglomerated in addition to the deposition on the SiO_2 surface and formed an irregular core-shell structure [25].

The particle size distributions were also determined from SEM and TEM images in Figure 3 according to the literatures [26, 27]. The SiO_2 core particles had a narrow

FIGURE 4: XRD patterns of (a) TiO_2 , (b) $\text{Ti/Si} = 1 : 1$, (c) $\text{Ti/Si} = 2 : 1$, (d) $\text{Ti/Si} = 5 : 1$, (e) $\text{Ti/Si} = 8 : 1$, (f) $\text{Ti/Si} = 1 : 2$, (g) $\text{Ti/Si} = 1 : 5$, (h) $\text{Ti/Si} = 1 : 8$, and (i) SiO_2 .

FIGURE 5: N_2 adsorption-desorption isotherms of the synthesized nanoparticles.

size distribution (average size = 318 nm) (Figure 3(a)) and TiO_2 nanoparticles had a size distribution of average 16.4 nm (Figures 2(b) and 3(b)). Furthermore, the average size of the $\text{Ti/Si} = 1 : 5$ was 333 nm for the uniform TiO_2 shell coated on SiO_2 particle. The size of CSTNs increased as the content of TiO_2 increased in the Ti/Si molar ratio shown from Figure 3(b)–3(f).

3.2. X-Ray Diffractometer (XRD) Analysis. The phase compositions and crystallinity of the SiO_2 , TiO_2 , and core-shell $\text{SiO}_2@ \text{TiO}_2$ nanoparticles (CSTNs) were investigated by XRD in Figure 4. As shown in Figure 4(a), the TiO_2 with 2θ peaks of 25.2° , 37.8° , 48.0° , 53.9° , 62.6° , and 75.1° corresponded to the anatase phase of TiO_2 [28]; the average TiO_2 crystallite sizes calculated by the Scherrer equation were around 18.0 nm which basically agreed with the transmission electron microscopy (TEM) results. Figures 3(b)–3(e) indicate that the CSTNs with different Ti/Si molar

ratios (1:1, 2:1, 5:1, and 8:1) showed the same 2θ peaks of anatase TiO_2 phase but without sharp peaks due to the doped silica, which also confirmed that titania was coated on the SiO_2 particles. The wide diffraction peak at $2\theta = 23^\circ$ was ascribed to the amorphous SiO_2 in Figure 4(i), which would explain the spherical morphology and lower surface area values of SiO_2 without preferential directions for crystal growth [29–31]. Similarly, no detectable characteristic anatase TiO_2 diffraction peaks occurred in $\text{Ti/Si} = 1 : 5$ (Figure 4(g)) and $\text{Ti/Si} = 1 : 8$ (Figure 4(h)) because of more SiO_2 amorphous structure coated on it. It was reported that the crystallinity of $\text{SiO}_2\text{-TiO}_2$ composites decreased while the molar ratio of silica increased, and this behaviour was explained by the fact that the silica suppressed the growth of titania [32].

3.3. N_2 Adsorption-Desorption Isotherm Analysis. Figure 5 illustrates N_2 adsorption-desorption isotherms of the

FIGURE 6: Pore size distribution calculated from adsorption branch of the synthesized nanoparticles.

synthesized samples. As shown in Figure 5(a), the adsorption isotherm of TiO_2 nanoparticles can be categorized as type IV with hysteresis loop of type H2, which indicated the characteristic type of TiO_2 nanoparticles with mesoporous materials [33]. The isotherms of the core-shell $\text{SiO}_2@\text{TiO}_2$ nanoparticles (CSTNs) with the molar ratios of Ti/Si (1:1, 2:1, 5:1, and 8:1) exhibited the similar shape of type IV with H2 hysteresis loops (Figure 5(a)) and showed the characteristic type of mesoporous materials as the TiO_2 nanoparticles. Besides, the adsorbed volume enhanced as the molar ratios of Ti/Si increased from 1:1 to 8:1, due to the coated amorphous silica on the CSTNs as shown in X-ray diffractometer (XRD) analysis.

Figure 5(b) shows the isotherms of the SiO_2 particles and CSTNs with the Ti/Si of 2:1, 5:1, and 8:1. The isotherms of Ti/Si = 1:2 exhibited the similar shape of type IV with H2 hysteresis loops and indicated the presence of mesopores. However, the isotherms of Ti/Si = 1:5 and Ti/Si = 1:8 showed the same isotherm shape of SiO_2 particles with low adsorbed volume and type I shape without any obvious hysteresis loop, which was attributed to the less mesoporous titania coated on the CSTNs [34].

The pore size distributions were shown in Figure 6 the Brunauer-Emmett-Teller (BET) surface areas and the pore size parameters were tested (Table 1) by using the Barrett-Joyner-Halenda (BJH) method from the adsorption branch of the isotherm. As shown in Figure 6(a), the TiO_2 nanoparticles had monomodal pore size distribution with a maximum pore diameter of about 13 nm. The pore size distributions of the CSTNs were more obvious, because the Ti/Si increased from 1:1 to 8:1 (Figure 6(a)); thus, more titanium dioxide was coated. However, the CSTNs with high silica molar ratios (Ti/Si = 1:5 and Ti/Si = 1:8) did not show the pore size distributions like SiO_2 (Figure 6(b)). In general, the pore size dis-

TABLE 1: Brunauer-Emmett-Teller (BET) surface area and pore size parameters of the synthesized nanoparticles.

Sample	BET surface area (m^2g^{-1})	Pore size (nm)	Pore volume (cm^3g^{-1})
SiO_2	8.51	41.57	0.004
TiO_2	33.78	13.39	0.136
Ti/Si = 1 : 1	122.53	7.67	0.258
Ti/Si = 2 : 1	167.06	6.62	0.311
Ti/Si = 5 : 1	176.79	6.19	0.312
Ti/Si = 8 : 1	174.70	5.92	0.297
Ti/Si = 1 : 2	102.02	8.67	0.242
Ti/Si = 1 : 5	24.93	27.44	0.0946
Ti/Si = 1 : 8	14.19	45.78	0.101

tributions presented the same trend of adsorption-desorption isotherms of the CSTNs as shown in Figure 5.

Following this, we measured the specific surface area, pore size, and pore volume of the CSTNs, which are listed in Table 1. The results showed that SiO_2 nanoparticles had a specific surface area of $8.51\text{m}^2\text{g}^{-1}$ with an average pore size of 41.57 nm; the TiO_2 nanoparticles had a specific surface area of $33.78\text{m}^2\text{g}^{-1}$ with an average pore size of 13.39 nm. The CSTNs of Ti/Si = 5 : 1 had the biggest specific surface area of $176.79\text{m}^2\text{g}^{-1}$, which was more than 5 times higher than the specific surface area of TiO_2 . However, the specific surface area of the CSTNs decayed as the molar ratios of Ti/Si decreased from 1:1 to 1:8. The pore volume of the CSTNs showed the same regularity of the specific surface area, and the CSTNs of Ti/Si = 5 : 1 had the highest pore volume of $0.313\text{cm}^3\text{g}^{-1}$. It can be concluded that the CSTNs with the appropriate molar ratio of Ti/Si had

FIGURE 7: Isoelectric point (IEP) analysis with pH values.

TABLE 2: Isoelectric point analysis of (IEP) and apparent surface coverage (ASC).

Sample	IEP (pH)	ASC (%)
TiO ₂	5.29	/
Ti/Si = 1 : 1	4.60	74.3
Ti/Si = 2 : 1	4.72	79.1
Ti/Si = 5 : 1	4.78	81.4
Ti/Si = 8 : 1	4.88	85.3
Ti/Si = 1 : 2	4.45	68.2
Ti/Si = 1 : 5	3.86	41.4
Ti/Si = 1 : 8	3.76	36.5
SiO ₂	3.10	/

significant effects on the samples' specific surface area, pore size, and pore geometry [35].

3.4. Isoelectric Point (IEP) Analysis. Surface charges were measured by the isoelectric point (IEP) and apparent surface coverage (ASC) of the synthesized nanoparticles in Figure 7 and Table 2. It was reported that surface charge affected the adsorption process of organic molecules and the driving force of hole migration on the surface of photocatalyst [10, 22]. As shown in Figure 7, generally, the IEP of the core-shell SiO₂@TiO₂ nanoparticles (CSTNs) ranged between silica (IEP at pH 3.10) and titania (IEP at pH 5.29); the IEP trends of the CSTNs with the molar ratios of Ti/Si=1:1, 2:1, 5:1, and 8:1 were gradually close to TiO₂ while the IEP trends of the molar ratios of Ti/Si=1:2, 1:5, and 1:8 were gradually close to SiO₂. It can be concluded that more molar ratio of titania coating on CSTNs increased the electrophoretic mobility and shifted the IEP to the similar value of TiO₂ particles, which showed the similar trend in the literature [36]. The apparent surface coverage (ASC) of

the CSTNs showed the ASC increased from 74.3% to 85.3% as the molar ratios of Ti/Si increased from 1:1 to 8:1 (Table 2); the results were in accord with IEP of the CSTNs. Besides, the ASC results also indicated that silica particles possessed a discontinuous titania nanoparticle coating layer.

3.5. Photocatalytic Activity. The photocatalytic activity of core-shell SiO₂@TiO₂ nanoparticles (CSTNs) was showed in Figure 8 by photodegrading the substrates of phenol and methylene blue (MB) under UV light irradiation. As shown in Figure 8(a), all the nanocomposites did not show photocatalytic activity during the dark reaction, which demonstrated the nanocomposites had no adsorption efficiency towards colorless phenol under dark condition. Under the UV light irradiation, both the natural degradation and SiO₂ showed very low photocatalytic efficiency, but for the photodegradation efficiency of the CSTNs gradually increased as the molar ratio of Ti/Si increased from 1:8 to 8:1. TiO₂ showed higher photocatalytic activity than CSTNs with silica increased in the molar ratios of Ti/Si from 1:1 to 1:8, while the photocatalytic activities of the CSTNs with the molar ratios of Ti/Si=2:1, 5:1 and 8:1 were higher than the TiO₂. The Ti/Si molar ratio of 5:1 possessed the highest photodegradation efficiency of 99.4% under UV light photodegradation for 120 minutes because of the highest specific surface area of 176.79 m²g⁻¹ as shown in Table 1. Due to the higher content of TiO₂ could lead to the formation of free coreless TiO₂ nanoparticles and the subsequent aggregation in aqueous dispersions, the photoactivity of Ti/Si molar ratio of 8:1 was slightly lower than 5:1 [10–12].

We evaluated the photocatalytic activity of CSTNs for the photodegradation of MB which was showed in Figure 8(b). During the dark reaction, the nanoparticles had different adsorption efficiency towards MB and the Ti/Si molar ratio of 5:1 showed the highest adsorption efficiency, which was ascribed to the mesoporous materials with colored organic dyes adsorption [31]. Compared with

FIGURE 8: Photocatalytic activity of phenol (a) and methylene blue (b) under UV light.

the photodegradation of phenol, similar trends can be observed as shown in Figure 8(b); the CSTNs with the molar ratio of Ti/Si from 1:1 to 8:1 showed high photodegradation efficiency of more than 95% after UV irradiation for 80 minutes. The Ti/Si molar ratio of 5:1 also had the highest photocatalytic activity of 99.2% after 120 minutes of UV light photodegradation.

For CSTNs with different molar ratios of Ti/Si, the higher molar ratios of titania in Ti/Si induced agglomeration of excess TiO₂, and poor dispersibility in aqueous solution. In addition, too much titania can reduce the specific surface area, occlude the pores, and limit the diffusion of the reactant molecules. In contrast, with higher molar ratios of SiO₂ in Ti/Si, although the CSTNs showed spherical and uniform core-shell structure, the reaction sides of titania reduced and the crystallinity was low too, so the photocatalytic efficiency would be reduced [25].

Compared with reported paper about the TiO₂-SiO₂ composites for photodegradation of phenol and MB as shown in Table 3 [36–47], the CSTNs with Ti/Si molar ratio of 5:1 in this paper showed higher photocatalytic efficiency with less concentration of photocatalysis (0.25 g/L).

There were several reasons for better photocatalytic activity with the proper molar ratio of Ti/Si = 5:1 in the CSTNs. Firstly, a proper molar ratio of Ti/Si both enhanced the thermal stability and increased the surface area and surface acidity. At the meantime, higher surface area and mesoporous structure provided more adsorption sites and photocatalytic reaction centers, which can enhance the photocatalytic reaction of phenol and MB. It is noted that a suitable molar ratio of Ti/Si may lead to an increase in the surface defects, which was reported to have the ability to capture the photocatalytic carriers and increase the reactive activity of hydroxyl [48, 49].

4. Conclusions

In summary, we synthesized the core-shell SiO₂@TiO₂ nanoparticles (CSTNs) with different molar ratios of Ti/Si by hydrothermal method. The study showed that the TiO₂ and CSTNs had the anatase phase after hydrothermal method and calcination at 450°C for 2 h. Furthermore, we evaluated the morphology, structure, and other properties of the CSTNs by scanning electron microscopy (SEM), transmission electron microscopy (TEM), X-ray diffraction (XRD), Brunauer-Emmett-Teller (BET) surface area, and surface charges. The BET surface area results indicated that the different molar ratios of Ti/Si affected the specific surface area on the CSTN; for the meantime, the CSTNs with the molar ratios of Ti/Si increased from 1:1 to 8:1 had the mesoporous structures which were categorized as type IV and hysteresis loop of type H2. Importantly, the surface charge results indicated that isoelectric point (IEP) of CSTNs ranged between 3.10 (silica) and 5.29 (titania), and the apparent surface coverage (ASC) ranged from 36.5% (Ti/Si molar ratio of 1:8) to 85.3% (Ti/Si molar ratio of 8:1). The photocatalytic activity of the CSTNs showed different photodegradation efficiencies with different molar ratios of Ti/Si towards colorless phenol and colored methylene blue (MB) under UV irradiation caused by different consistency of surface area and suitable porous structure. Finally, the experimental results showed the CSTNs with the Ti/Si molar ratios of 5:1 had the highest photocatalytic activity of 99.4% towards phenol and 99.2% towards MB, which showed higher photocatalytic efficiency with the addition of 0.25 g/L photocatalysts compared with the other reported TiO₂-SiO₂ composites [36–47].

TABLE 3: A comparison of TiO_2 - SiO_2 composites for photodegradation of phenol and MB.

Photocatalyst	Synthesis method	Light source	Initial concentration of substrate (mg/L)	Concentration of photocatalysis (g/L)	Reaction time (min)	Efficiency (%)	Ref.
TiO_2 - SiO_2 photocatalyst	Sol-gel	UV light (150 W)	50 (phenol)	1	120	48	[36]
Titania-silica composites	Nonaqueous approach	UV light (8 W)	100 (phenol)	3	180	67	[37]
TiO_2 - SiO_2 catalysts	Homogeneous precipitation method	UV light (125 W)	500 (phenol)	2.5	640	100	[38]
TiO_2 - SiO_2 aerogels	Sol-gel method	UV light (15 W)	50 (phenol)	1	180	42	[39]
CSTNs	Hydrothermal method	UV light (500 W)	20 (phenol)	0.25	120	99.4	This paper
$\text{TiO}_2/\text{SiO}_2$ nanoparticles	Hydrothermal method	UV light (30 W)	10 (MB)	1	35	96.4	[40]
Titania-modified mesoporous silicates	Impregnation method	UV light (400 W)	10 (MB)	1	240	96.0	[41]
Silica-titania mixed oxides	Sol-gel method	Xenon lamp (0.68 W/m ²)	1×10^{-5} mol/L (MB)	12	180	95	[42]
TiO_2 - SiO_2 mesoporous materials	Hydrothermal method	UV light (125 W)	40 (MB)	0.3	110	65	[43]
Silica-titania photocatalysts	Hydrothermal method	UV light (125 W)	20 (MB)	5	360	90	[44]
$\text{SiO}_2/\text{TiO}_2$ nanoparticles	Sol-gel method	Xenon lamp (300 W)	10 (MB)	1	80	97.7	[45]
TiO_2 - SiO_2 mixed oxides	Sol-gel method	UV light (39 W)	5×10^{-5} mol/L (MB)	0.3	120	76.7	[46]
$\text{TiO}_2/\text{SiO}_2$ composites	Sol-gel method	Xenon lamp (300 W)	2×10^{-5} mol/L (MB)	0.3	60	98	[47]
CSTNs	Hydrothermal method	UV light (500 W)	20 (MB)	0.25	120	99.2	This paper

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that there is no conflict of interests regarding the publication of this paper.

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