

Fabric Treated with Dye Modified TiO₂/Graphene Composite and its Photocatalytic and Self-Cleaning Properties

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Abstract

In this paper, a dye modified titanium dioxide/graphene composite (DTG20) was prepared using direct grey D, a dye with good water solubility, as a modifier. Then the DTG20 and its intermediates, including TiO₂ and titanium dioxide/graphene (TG), were finished on cotton fabric through the padding, coating and compounding methods, respectively. Photocatalytic properties of the treated fabrics were investigated using the MB simulated pollutant. It was found that the photodegradation efficiency of MB shown by DTG20 finished fabric treated with the compounding method can reach 90.55 %, and after four recycling experiments, the removal rate of MB can still reach 77.07 %. After being exposed to sunlight for 6 hours, the photodegradation efficiency of MB stained on the surface of DTG20 treated fabric can achieve 91.44 %; therefore, it also exhibited an excellent self-cleaning effect.

Keywords

Dye modified titanium dioxide/graphene composite, treatment method, cotton fabric, photocatalytic self-cleaning, photocatalytic degradation.

1. Introduction

TiO₂/rGO composite, with its good photodegradation ability, has been widely used in wastewater treatment [1-3]. In recent years, some metal compounds and organic molecules have been introduced into TiO₂/rGO composite to make full use of sunlight by this kind of photocatalyst [4-5], which usually have narrow bandgaps and are used as sensitizers. On the one hand, the introduction of sensitizers can enlarge light response and produce more photogenerated electrons, but on the other it can also decrease the bandgap and effectively improve the photocatalytic performance of the composite [6-7]. For example, Kalyani [8] et al. prepared a kind of PTh-rGO-TiO₂ composite by covalently functionalising reduced graphene oxide (rGO) with TiO₂ and polythiophene (PTh), because of the good hole conducting property of polythiophene. The conjugated system obtained showed a great H₂ production rate of 214.08 μmolh⁻¹, with a quantum efficiency of 14.17% at 400nm, and the composite also showed efficient degradation of methylene blue. Yu [9] et al., loaded negative reduced graphene oxide (rGO) nanosheets and positive phenylamine (PhNH₂) molecules on a TiO₂ surface to realise selective adsorption for both cationic and anionic dyes on the photocatalyst surface. Therein

graphene was used as an electron transfer medium between the catalyst (TiO₂) and photosensitizer, which transferred the photogenerated electrons from the sensitizer to the metal oxide and decreased the recombination of the photogenerated electrons, thus significantly improving the current-carrying electron mobility of the catalyst, which prolonged the charge life and enhanced the diffusion strength of the charge. Although current ternary component photocatalyst composites have been endowed with excellent photocatalytic performances, it is still difficult to fully recycle them from treated wastewater. They are usually used as a solid powder and directly poured into polluted water, thus resulting in an amount of photocatalyst remaining in the treated sewage, which influences the total purification of water [10-11]. In this article, a dye covalently functionalised titanium dioxide /graphene ternary composite (DTG20) was prepared using direct grey D as a sensitizer. Then it was used as a finishing agent to treat cotton fabric to get a functional fabric. The photocatalytic properties of fabrics treated with TiO₂, TG and DTG20 were studied using MB solution as simulation sewage, and the treating methods were compared. Wastewater purified with such fabrics not only can simplify the sewage treatment process but also can realise

the full recovery of the photocatalyst. Furthermore, the DTG20 treated fabric also has a self-cleaning effect.

2. Experimental

2.1. Materials

Flake graphite powder, 97%, Aladdin Reagent Co., Ltd. (Shanghai, China); Nano Titanium Dioxide(IV), 99%, Bailingwei Technology Co., Ltd. (Beijing, China); Direct Gray D, 97%, Yijia Industrial Co., Ltd. (Shanghai, China); Sodium dodecyl sulfate (SDS), AR, Fuchen Chemical Reagent Factory (Tianjin, China); water-based acrylic emulsion; acrylic thickener DR-73, solid content 40%, Yoshida Chemical Co., Ltd. (Shenzhen, China); Penetrant (JFC), AR, Aikeda Chemical Reagent Co., Ltd. (Chengdu, China).

2.2. Preparation of photocatalyst DTG20

2.2.1. Preparation of Titanium Dioxide / Graphene (TG) Composite

Graphene oxide (GO) was prepared by referring to the literature method [12].

Then 125 mg of GO and 60 mL of ethanol aqueous solution (V ethanol: V water = 1:2) were added into a 100mL beaker alongside a magnetic stirrer, stirred for 10 min, and then the mixture was dispersed with an ultrasonic disperser for 30 min to obtain a uniform GO dispersion. 0.5 g of TiO₂ was added to the beaker and stirred for 2 h, then ultrasonically dispersed for 30 min. The mixture was transferred to a hydrothermal kettle lined with PTEE, kept at 150°C for 12 h, and then the kettle was cooled to room temperature naturally. The solid product was centrifuged, washed with water and absolute ethanol for 3 times, and dried overnight at 60°C under a vacuum to obtain a TG composite with an rGO content of 20%, namely TG20.

2.2.2. Preparation of Dye Modified Graphene/Titanium Dioxide (D-TG)

2.7 g (0.005 mol) of direct ash D, 0.52 g (0.0075 mol) of sodium nitrite, and 10 g of water were added into a 150 mL beaker with a magnetic stirrer, which were then mixed, the mixture cooled below 5 °C, 4.5 mL of 25% sulfuric acid solution added slowly to diazotise for 3 h, and finally a dye diazonium salt was obtained.

0.1 g of the TG composite and 1g of sodium dodecyl sulfate (SDS) were added to 40 mL of deionised water and ultrasonically dispersed for 20 min to obtain TG water dispersions. The dispersion was cooled to below 10 °C, and the dye diazonium salt solution added to it. The mixture was kept below 10 °C for 1 h and then the temperature was raised to room temperature for another 8 h. The product was centrifuged at a speed of 10000 r/min for 15 min to remove the supernatant, and then washed with water, ethanol, and acetone several times until the supernatant was colourless. The solid powder obtained was dried overnight under a vacuum at 60 °C to obtain a D-TG ternary composite, namely DTG20.

2.3. Cotton fabric treatment

In this study, 100% cotton woven fabric (weight – 314 g/m², thickness – 0.25 mm, fabric density – warp: 60 yarns/cm, and

weft: 60 yarns/cm) was used and treated with a photocatalyst composite using three different methods.

Coating method: A viscous coating solution based DTG20 powder was prepared by the sonication of 0.08 g of DTG20 and 0.08 g of thickener in 10 g of water-based acrylic emulsion. Then, it was coated on the surface of the cotton fabric with a wire rod coater, dried at 100 °C for 5 min, and then cured at 150 °C for 2 min. For comparison, TiO₂ and TG20 coated cotton were prepared under the same conditions.

Padding method: Firstly, a finishing solution was prepared with concentrations of the penetrant (JFC) of 1 g/L, NaCO₃ 2 g/L, Na₃PO₄ 0.2 g/L, DTG20 2% (o.w.f) & NaCl 2 g/L, while keeping the bath ratio at 30:1. Afterwards, the pre-moistened cotton fabric was immersed in the above-prepared dispersion and maintained at 85 °C for 45 min. This was followed by padding under a pressure of 10 Kgf/cm² at a speed of 20 m/ min with 75% expression, drying at 90 °C for 3 min, and curing at 150 °C for 5 min. For comparison, TiO₂ and TG20 treated cotton were prepared under the same conditions.

Compounding method: Firstly, the cotton fabric was treated with the padding method and then by the coating process.

2.4. Characterisation

The surface morphologies of the samples were studied by means of a scanning electron microscope. (Apreo 2C, American Thermo Scientific Company).

2.5. Evaluation of photocatalytic performance

Photocatalytic activities of the treated fabrics were assessed by analysing the decrease in the concentration of methylene blue solution (10 g/L) during exposure to UV-visible and visible light. For this, a 4 cm × 4 cm finished fabric slice was added to 30 mL of solution and magnetically stirred in the dark for

60 min to achieve adsorption-desorption equilibrium. Afterwards, photocatalytic degradation experiments were carried out in a homemade photoreactor equipped with a 500W xenon lamp and reflux condenser, placed 15 cm above the solution. For the visible light experiments, a watch-glass was placed on top of the MB aqueous solution to shield from the UV light of the xenon lamp.

For both experimental setups, the efficiency of the degradation was determined by monitoring the intensity of the main absorbance peaks of MB (664 nm) using an S10 spectrophotometer. The photocatalytic degradation efficiencies were calculated as equation (1-1).

$$D\%=(C_0-C_t)/C_0 \times 100\% \quad (1-1)$$

Where: C₀ and C_t represent the concentration of the MB (pollutant) at time 0 min and at time t, respectively. In this equation, the D % shows the dye photocatalytic degradation percentage.

To investigate the reusability of the finished cotton fabrics, circulating experiments were carried out. The fabric samples were taken out from the sewage after performing a photodegradation experiment for 2 h, rinsed with water, allowed to dry naturally, and then put into new sewage again. This operation was implemented four times.

2.6. Self-cleaning effect of finished cotton fabric

Three DTG20 finished cotton fabric samples of 3 cm × 3 cm were dipped in 20 mL of MB aqueous solution with concentrations of 5 mg/L, 10 mg/L, and 20 mg/L, respectively, for 5 min, then dried at 80 °C in an oven. The samples were exposed to sunlight for 6 h (Beijing, summer, 10 o'clock to 16 o'clock). The initial color of the stained sample was compared with that after illumination in terms of their K/S values to measure the photodegradation of the MB stain using a spectrophotometer (Data color).

$$\eta=((\alpha_1-\alpha_0))/((\alpha_1-\alpha_0)) \times 100\% \quad (1-2)$$

In the equation: α_0 is the K/S value of the fabric before absorbing the MB solution, α_1 the K/S value of the fabric after the adsorption of MB, α the K/S value of the fabric after being illuminated, and η is the degradation rate of MB. The larger the η , the better the self-cleaning effect of the fabric [13-14]. All the K/S values are the average of those measured at 5 random spots at least on the sample. The experiment for every concentration was repeated for three times on the same day and during the same time.

3. Results and discussion

3.1. Effect of treating method on photocatalytic performance

3.1.1. Padding method

The photocatalytic degradation efficiency of finished cotton fabrics treated with DTG20 and their intermediates obtained through the padding method is shown in Fig.1. The finished cotton fabric treated with DTG20 showed the best MB removal effect, and after 2 hours of illumination, the MB degradation efficiency reached 80.8%, which is mainly attributed to the strong adsorption of MB by DTG20 in cotton fabrics. As a graft of direct grey D on DTG20, some polar and ionic groups, such as $-\text{OH}$, $-\text{NH}_2$ and $-\text{SO}_3\text{H}$, were introduced into the composite and endowed typical dye properties to the composite. Therefore, to improve the affinity of the composite for cotton fabric [15-17], more DTG20 was absorbed by the fabric, forming a hydrogen bond with the fabric. When treating the MB pollutant, the fabric treated with DTG20 could absorb MB molecules and degrade them. Whereas for the finished cotton fabrics treated with TG20 and TiO_2 , the removal rates of MB were only 30.02% and 27.74%, respectively, which is not much different from that of the control cotton fabric. This may be due to the poor affinity of TG20 and TiO_2 for cotton, limiting the absorption of these photocatalysts on the fabric, thus decreasing the degradation efficiency of MB.

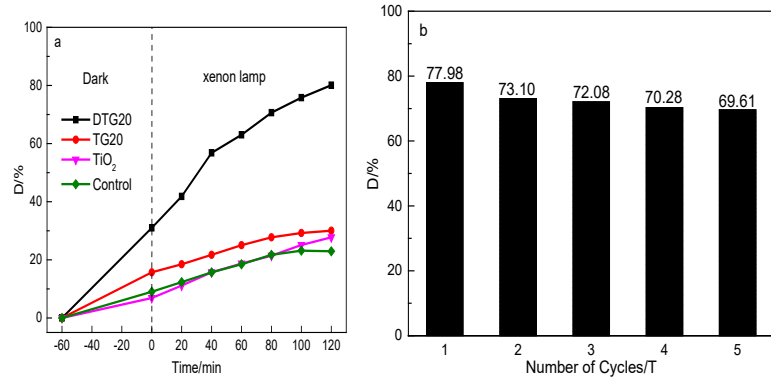


Fig. 1. Photocatalytic degradation efficiency of MB of finished fabrics treated with the padding method: (a) effect of the finishing agent; (b) Photocatalytic cycling experiments of DTG20 finished fabric

In addition, we can also see from Fig.1b that the photodegradation efficiency of MB by cotton fabric finished with DTG20 decreased as the number of cycle times increased; but after four cycles, the removal rate of MB could still reach 69.61%. The morphologies of the control cotton fabric treated with DTG20 before and after performing photodegradation experiments are shown in Fig. 2. It can be seen from Fig. 2 that the control cotton fibres were flat, smooth and showed a ribbon-like style with natural twists. The surface of the fabric treated with DTG20 was a little rough and presented small granular protrusions, indicating that the finishing agent had bonded or accumulated on the surface of the fabric [18]. After treating the MB sewage, the granular protrusions on the surface of the finished fabric became less, but the overall change was very small, indicating that the DTG20 had bonded on the fibre through hydrogen-bonding and Van der Waals forces. The reason for the decrease in the photodegradation efficiency of MB as the number of cycles increased may have been that a very small amount of DTG20 had fallen away from the fabric. Furthermore, the finished fabric absorbed a large amount of MB molecules during the degradation process, which are difficult to be degraded completely in a limited time, thus remaining on the surface of the fabric and forming a coverage layer on the surface. This will weaken the absorption and utilisation of illuminants by the photocatalyst and cause a decrease in the degradation efficiency, which trend will increase with the increasing of cycling times.

3.1.2. Coating method

Figure 3a presents the photodegradation efficiency of finished cotton fabrics treated with DTG20 and its intermediates through the coating method. It can be seen from figure 3a that the degradation efficiency of MB showed obvious diversity among the cotton fabrics treated with different catalysts. The removal rate of MB by the control fabric was only 23.04%, which was attributed to the absorption of MB by the cotton fabric. The removal rate of MB by fabrics treated with DTG20, TG20 and TiO_2 , under the irradiation of the xenon lamp reached 82.73%, 69.98% and 46.39%, respectively, and the fabric coated with DTG20 coating slurry presented the best degradation effect. This is mainly due to the various photocatalytic degradation ability of different catalysts. In addition, compared with the padding method, the finished fabrics treated with the coating method revealed better photocatalytic activities as a whole. This is a consequence of photocatalysts that have adhered to the surface of the fabric by the polyacrylic acid film. Therefore, regardless of the kind of photocatalyst, a certain amount of it has adhered to the finished fabric.

Figure 3b shows the degradation efficiency of MB for DTG20 finished fabric for different cycles of experiments. It can be seen from Fig. 3b that as the number of cycles increased, the removal rate of MB gradually decreased. After four cycles, the degradation efficiency of MB was about 72%, indicating that the

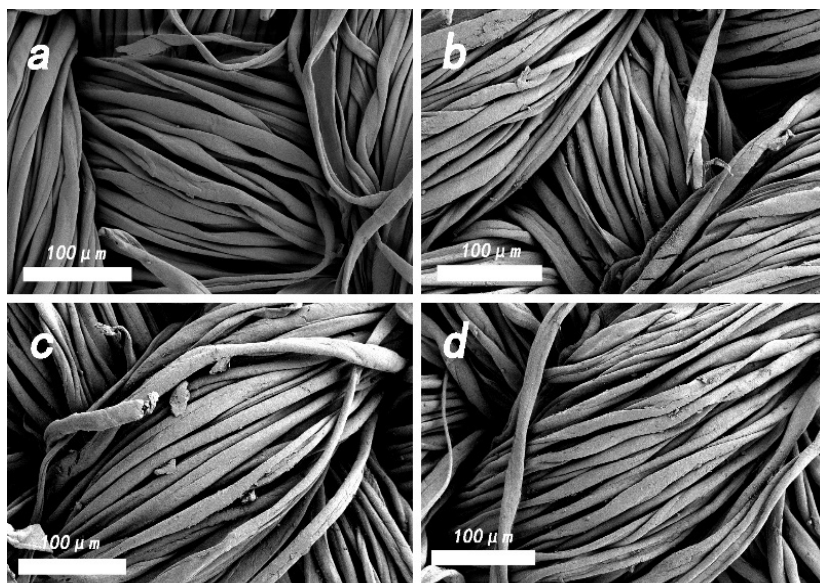


Fig. 2. SEM images of fabrics: control cotton fabric(a), DTG20 fabrics treated by padding method prior(b), and after treating MB pollutant the 1st time(c) and the 5th time(d).

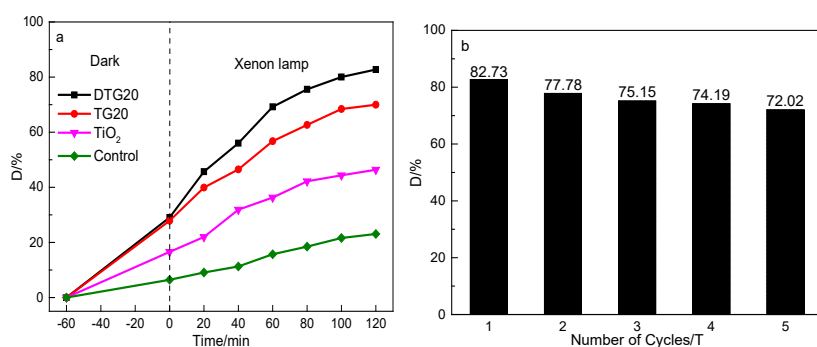


Fig. 3. Photocatalytic degradation efficiency of MB of finished fabrics treated with the coating method: (a) effect of the finishing agent ; (b) Photocatalytic cycling experiments of DTG20 finished fabric

DTG20 coated cotton fabric has a certain durability. The morphology of DTG20 coated fabric before and after performing the photocatalytic experiment is shown in Fig. 4. It was found that adhesion occurred between fibres and that a smooth film formed on the surface and among the gaps of fibres for the DTG20 treated fabric. After performing the degradation experiment, the coating film on the surface of the fibre was damaged and became rough, with some traces of abrasion appearing. As the number of cycles increased, the damage became more serious, which can explain why the degradation efficiency of MB decreased as the cycle times increased. Except for the accumulation of MB molecules on the surface of the fabric, the coating film

peeled off from the treated fabric during the degradation experiment, and the photocatalyst fell off therewith, resulting in an decrease in the degradation efficiency of MB.

3.1.3. Compounding method

In order to degrade the MB pollutant thoroughly, a compounding method was used to treat the cotton fabric using DTG20 and its intermediates. The photocatalytic degradation efficiency of MB of the finished cotton fabric treated with the compounding method is shown in Fig. 5. The degradation efficiencies of MB of finished fabrics treated with DTG20, TG20 and TiO₂ through the

compounding method were all higher than those by the coating and padding methods only, and the removal rate of MB was 90.55%, 75.27% and 51.50%, respectively. At the same time, cycling experiments were performed on the cotton fabric treated with DTG20, the results of which are shown in Fig. 5b. It can be seen from Fig. 5b that after performing the 4 cycle experiment, the removal rate of MB could still reach 77.07%.

3.2. Effects of light source on the photocatalytic performance of DTG20 treated fabrics

Different light sources were used in the treatment of MB pollutants. The photodegradation efficiency of MB treated by DTG20 finished fabrics using different treatment methods is shown in Fig. 6a, which shows that the fabric treated with the compounding method under a xenon lamp had the best MB photodegradation effect, and the removal rate of MB reached 90.55%. Moreover, the treated MB pollutant became colourless. Whereas the cotton fabrics treated by the coating and padding methods had an MB degradation efficiency of 82.73% and 77.98%, respectively, and the treated MB pollutants were still light blue, indicating that an amount of MB remained in the water. To figure out the ultraviolet light effect on the photodegradation of MB, a slice of glass was placed on the apparatus containing the pollutant, which could shield from the ultraviolet light from the xenon lamp. The photodegradation efficiency of MB under visible light is shown in Fig. 6b. It can be seen from Fig. 6b that the photodegradation efficiency of MB under visible light decreased a little as compared to that under a xenon lamp; but the treated fabrics still presented good photodegradation capability.

3.3. Photocatalytic self-cleaning property of DTG20 treated fabric

In an attempt to understand the adsorption of MB molecules on the surface of DTG20 treated fabric after performing

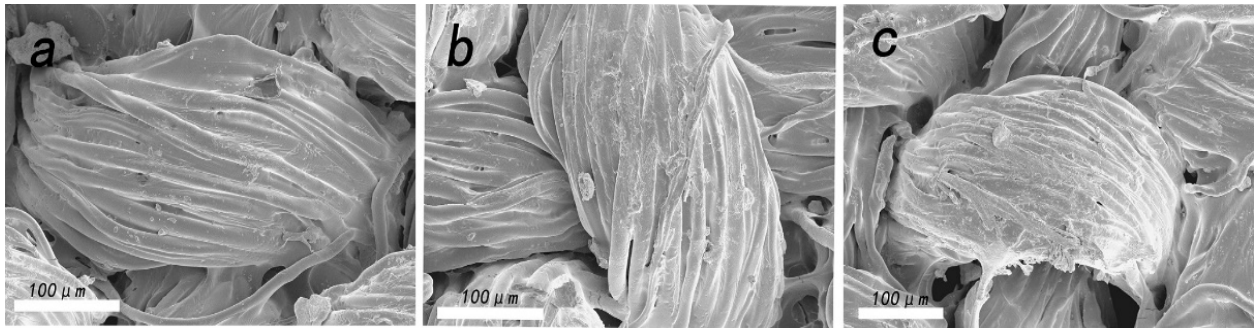


Fig. 4 SEM images of fabrics treated with DTG20 by the coating method before (a) and after performing the photocatalytic experiment for the 1st time (b) and 5th time (c)

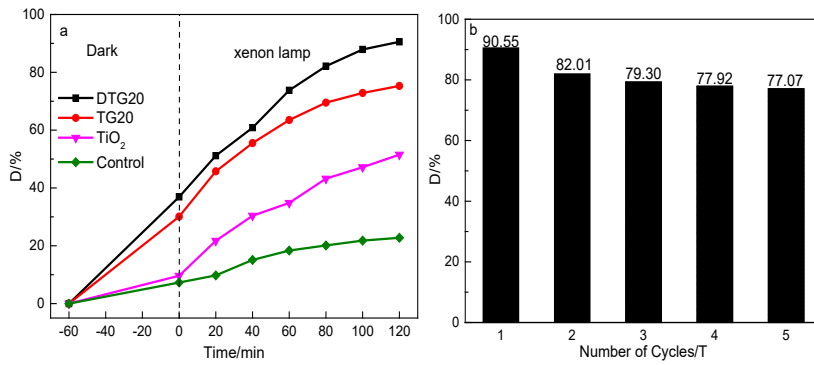


Fig. 5. Photocatalytic degradation efficiency of MB of finished fabrics treated with the compounding method: (a) effect of the finishing agent ; (b) Photocatalytic cycling experiments of DTG20 finished fabric

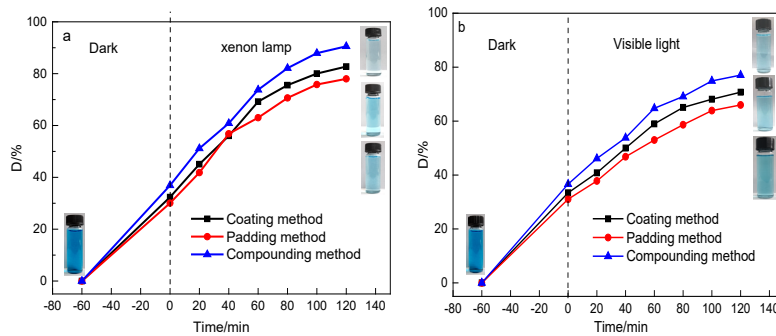


Fig. 6 Photodegradation efficiency of MB by DTG20 finished fabrics treated with different methods: (a) under xenon lamp; (b) under visible light

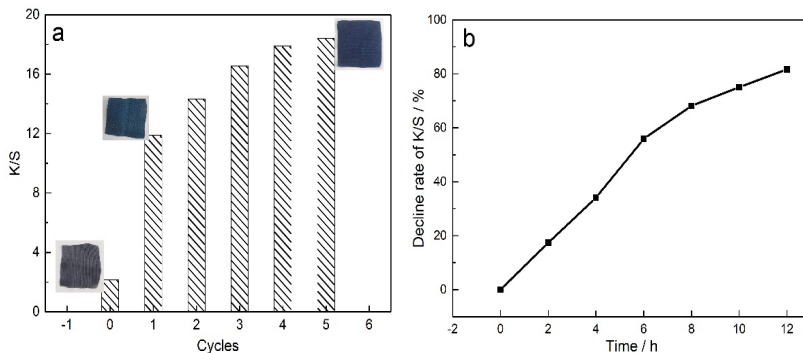


Fig. 7. K/S of DTG20 treated fabric (a) after performing photodegradation experiment (b) decline rate under xenon lamp irradiation

the photodegradation experiment, the K/S value was tested, the results of which are shown in Fig. 7a. It can be seen from Fig. 7a that as the number of cycles increased, the K/S value also increased, suggesting that the MB molecules had been adsorbed on the surface of the DTG20 treated fabric. After five cycles, the K/S value increased from 2.145 to 18.433 and the appearance of the DTG20 treated cotton fabric changed from grey to dark blue. This is also proof of a decrease in the photodegradation efficiency of MB as the number of experiment cycles increased. For further application of this fabric, the MB molecules absorbed should be removed as much as possible; hence, after performing the photodegradation experiment the treated fabric was put under a xenon lamp for illumination. The discard of MB molecules from the fabric was investigated by testing its K/S c. The graph in Fig. 7b shows that the K/S of the fabric declined a lot as the longer the illumination lasted. After 12 hours of illumination, the K/S value declined by 81.66% according to equation (1-2). It can be inferred that the DTG20 treated fabric exhibited excellent self-cleaning performance. When being exposed to sunlight, most of the remaining MB molecules on the fabric sample degraded and fell away.

To study the self-cleaning effect of DTG20 treated fabric, an actual experiment was conducted for stained fabric under natural sunlight, where the decline rate of the K/S value for the fabric was counted. The result is shown in Fig. 8. It can be seen from Fig. 8 that as the illumination time prolonged, the decline rate of K/S increased, while

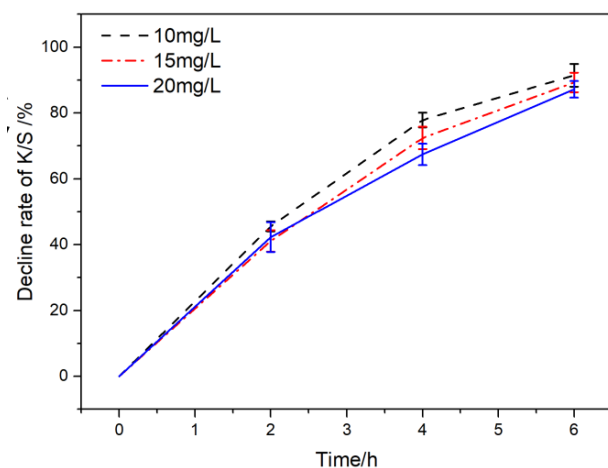


Fig. 8. Decline rate of K/S for stained DTG20 treated fabric under sunlight.

as the concentration of MB increased, the decline rate of K/S decreased a bit. When the MB concentration is 10 mg/L, the MB removal rate on the fabric can reach 91.44%. This may be associated with the amount of photocatalyst finish on the fabric and the photodegradation ability of MB being the same. When the concentration of MB was bigger, the K/S of the fabric was higher, leading to a lower value of the K/S decline rate.

4. Conclusions

In this paper, a DTG20 ternary composite material was successfully prepared and used to treat cotton fabric through the padding, coating and compounding methods, respectively. The photodegradation efficiency of MB pollutants of the treated fabrics showed that the removal rate of MB was highest for the fabric treated by the compounding method, reaching 90.55%. The cycle

experiment for DTG20 finished fabric presented good durability, and after four cycles the removal rate of MB could still reach above 75%. This was attributed to the strong affinity of the DTG20 composite to cotton fabric, bonding to the fabric by a hydrogen bond and Van der Waals forces. When being exposed to sunlight, the stained DTG20 treated fabric showed an excellent photocatalytic self-cleaning property, and the removal rate of MB could reach 91.44%.

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