

# Photodegradation of organic dyes over nickel distributed CNT/TiO<sub>2</sub> composite synthesized by a simple sol-gel method

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In this study, the multi-walled carbon nanotubes were oxidized by *m*-chlorperbenzoic acid followed by the reaction with titanium n-butoxide and nickel nitrate to prepare Ni distributed CNT/TiO<sub>2</sub> composite by a simple sol-gel method. The functional groups formed on the surface of MWCNTs were analyzed by Fourier transform infrared spectroscopy. The prepared Ni distributed CNT/TiO<sub>2</sub> composite was characterized by X-ray diffraction, scanning electron microscopy, transmission electron microscopy and energy dispersive X-ray analysis. The photodegradation of methylene blue, methylene orange and rhodamine B solution under UV irradiation was employed to test the photocatalytic activity of the Ni distributed CNT/TiO<sub>2</sub> composite. According to the results, Ni distributed CNT/TiO<sub>2</sub> composite showed very excellent photocatalytic activity to decompose MB, MO and Rh.B solutions, due to the electron absorption effect of MWCNTs and electron trapping effect of nickel.

Keywords: *MWCNTs, nickel, organic dyes, sol-gel, photocatalytic activity*

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## 1. Introduction

Semiconductor photocatalysis has attracted a lot of attention because it provides an ideal way to utilize solar energy or artificial light energy for decomposition of toxic gas and wastewater. The photocatalysis process commonly involves the following steps: (i) the semiconductors absorb photons and generate electron-hole pairs; (ii) the photoinduced electron-hole pairs are separated into electrons in the conduction band and holes in the valence band; (iii) then the electrons/holes migrate to the surface active sites of the particles; (iv) finally, the organic molecules are decomposed over the semiconductor surfaces. Among oxide photocatalysts, titanium dioxide (TiO<sub>2</sub>) has been intensively used in photocatalysis and environmental pollutant cleanup applications [1–4]. However,

electron-hole recombination is generally in direct competition with the trapping process. The rate of the trapping and subsequent photocatalytic reaction on TiO<sub>2</sub> surface can be enhanced by retarding the electron-hole recombination. One of the principal methods of improving the photocatalytic activity of TiO<sub>2</sub> is combining it with carbon nanotubes to create a CNT/TiO<sub>2</sub> system. Carbon nanotubes outperform other carbon materials due to their exceptionally high mechanical strength, high thermal conductivity, medium to high specific surface areas and high external surface area, which render them interesting candidates for the use as catalyst supports to prepare nanosized metal and metal oxide particle catalysts [5–8]. Recently, we have reported the synthesis of multi-walled carbon nanotubes (MWCNTs)/TiO<sub>2</sub> composites by a sol-gel method and determined their photocatalytic activity by decomposing different organic dyes under irradiation of ultraviolet (UV) or visible light [9–12].

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The results indicated that the photocatalytic activity of TiO<sub>2</sub> to decompose organic dyes could be improved by combining with carbon nanotubes under irradiation of UV or visible light. The MWCNTs can use its photoabsorption character to absorb a photo-induced electron (e<sup>-</sup>), which could be introduced into the conduction band (CB) of TiO<sub>2</sub> and react with O<sub>2</sub> to trigger the formation of the very reactive superoxide radical ion (O<sub>2</sub><sup>-•</sup>).

Reduction of the grain size or doping with the transition metals (Fe, V, Mn, Co, and Ni) and noble elements (Pt, Pd or Rh) have been investigated as the methods to enhance the photocatalytic performance of TiO<sub>2</sub> [13–16]. It is considered that the metal cocatalyst dispersed on the TiO<sub>2</sub> accelerates the transportation of electrons produced by the photoexcitation to the outer system, i.e. photocatalytic reaction. Although the precious metal cocatalyst, especially platinum (Pt), shows outstanding performance for photocatalytic activity [16, 17], it is fairly costly. Therefore, another important base metal cocatalyst, i.e. nickel (Ni), is proposed because of its cost-effectiveness. Tseng et al. reported the degradation of xylene vapor over Ni-doped TiO<sub>2</sub> photocatalysts prepared by polyol-mediated synthesis [18]. Ren et al. reported the improved photocatalytic properties of p-type NiO loaded porous TiO<sub>2</sub> sheets prepared via free tape-casting [19]. It is known that the metallic nickel could enhance the photocatalytic activity of TiO<sub>2</sub>.

In this study, the transition metallic Ni distributed CNT/TiO<sub>2</sub> composite was synthesized by a simple sol-gel method. Due to the fact that the MWCNTs had to be oxidized by MCPBA, the functional groups formed on the surface of MWCNTs were examined by a KBr method using Fourier transform infrared (FT-IR) spectroscopy. The prepared Ni distributed CNT/TiO<sub>2</sub> composite was characterized by X-ray diffraction (XRD), scanning electron microscopy (SEM), transmission electron microscopy (TEM) and energy dispersive X-ray (EDX) analysis. The photocatalytic properties of Ni distributed CNT/TiO<sub>2</sub> composite were investigated by decomposing three kinds of organic dyes: methylene blue (MB), methylene orange (MO) and rhodamine B (Rh.B) under UV light irradiation.

## 2. Experimental

### 2.1. Materials

Crystalline MWCNTs (purity, 95.9 wt.%; diameter: 20 nm; length: 5 μm) powder, obtained from Carbon Nano-material Technology Co., Ltd, Korea, was used as support material. *m*-chloroperbenzoic acid (MCPBA), purchased from Acros Organics, New Jersey, USA, was used as the oxidizing agent to oxidize the surface of MWCNTs. The titanium *n*-butoxide (TNB, Ti{OC(CH<sub>3</sub>)<sub>3</sub>}<sub>4</sub>, 99 %), purchased from Acros Organics, New Jersey, USA, was used as titanium alkoxide precursor to form the TiO<sub>2</sub>. Nickel nitrate (Ni(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O, 97 %), obtained from Duksan Pure Chemical Co., Ltd, Korea, was used as a nickel ion source. Benzene (99.5 %) purchased from Samchun Pure Chemical Co., Ltd, Korea was used as the organic solvent. The MB (C<sub>16</sub>H<sub>18</sub>N<sub>3</sub>S·Cl, 99.99+%), MO (C<sub>14</sub>H<sub>14</sub>N<sub>3</sub>NaO<sub>3</sub>S, 99.99+%) and Rh.B (C<sub>28</sub>H<sub>31</sub>ClN<sub>2</sub>O<sub>3</sub>, 99.99+%), purchased from Duksan Pure Chemical Co., Ltd (Korea), Daejung Chemicals & Metals Co., Ltd (Korea) and Samchun Pure Chemical Co., Ltd (Korea), respectively were used as analytical grade chemicals. Table 1 shows the structure of MB, MO and Rh.B.

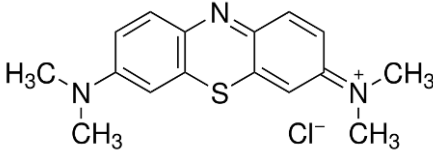
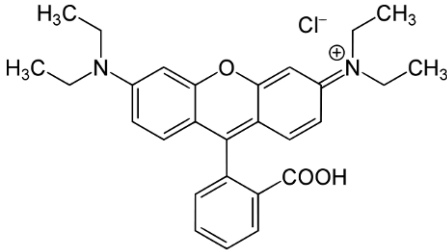
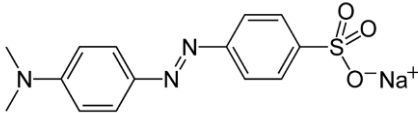
### 2.2. Synthesis

For the synthesis of the Ni distributed CNT/TiO<sub>2</sub> composite, three step procedure was used. In the first step, the MWCNTs were oxidized by MCPBA, which consequently allowed reacting with TNB in the second step to prepare CNT/TiO<sub>2</sub> composite. Then the nickel nitrate was introduced at the last step.

#### 2.2.1. Oxidization of MWCNTs

Due to high stability of the MWCNTs, they need to be treated with strong acids to introduce active functional groups on their surface. We took 1.0 g MCPBA melted in 60 mL benzene to prepare oxidizing agent. Then 0.5 g MWCNTs was put into the oxidizing agent. The mixture was magnetic stirred for 6 h at 343 K. Then the MWCNTs was dried at 373 K and spared.

Table 1. The molecular structure and  $\lambda_{max}$  of organic dyes.

| Organic dyes          | Molecular structure  | $\lambda_{max}$ |
|-----------------------|--|-----------------|
| Methylene blue (MB)   |  | 665 nm          |
| Methylene orange (MO) |  | 465 nm          |
| Rhodamine B (Rh.B)    |  | 554 nm          |

### 2.2.2. Preparation of CNT/TiO<sub>2</sub> composite

0.1 M TNB-benzene solution was prepared via dissolution of TNB in benzene solution. Subsequently, the pre-oxidized MWCNTs were put into 50 mL 0.1 M TNB-benzene solution. The mixtures were loosely covered and kept stirred at 343 K for 5 h, until a homogenous MWCNTs-contained gel was formed. The gel was heat treated at 773 K for 1 h with a heating rate of 279 K/min to obtain the CNT/TiO<sub>2</sub> composite.

### 2.2.3. Preparation of Ni distributed CNT/TiO<sub>2</sub> composite

0.1 M Ni(NO<sub>3</sub>)<sub>2</sub> solution was prepared via dissolution of Ni(NO<sub>3</sub>)<sub>2</sub> in distilled water. Subsequently, the prepared CNT/TiO<sub>2</sub> composite was put into 50 mL 0.1 M Ni(NO<sub>3</sub>)<sub>2</sub> solution. The mixtures were loosely covered and kept stirred at 343 K for 5 h. The temperature of the mixture was reduced to ambient temperature and the mixture was filtered through a filter paper. The obtained solids were dried at 373 K for 1 h and heat treated at 773 K for 1 h with a heating rate of 279 K/min. Finally the Ni distributed CNT/TiO<sub>2</sub> composite was prepared.

### 2.3. Characterization

The functional groups formed on the surface of MWCNTs were examined by a KBr method using Fourier transform infrared (FT-IR) spectroscopy (FTS 3000MX). The discs for the examinations were prepared by mixing 1 mg of powdered oxidized MWCNTs with 600 mg of KBr (for FT-IR spectroscopy) in an agitated mortar, and then pressing the resulting mixture successively under a pressure of 450 Pa for 3 minutes. The spectra of the samples were measured between 3000 and 500 cm<sup>-1</sup>. For the physical parameters measurements, XRD (Shimadzu XD-D1) results were used to identify the crystallinity with Cu K $\alpha$  radiation. SEM (JSM-5600) was used to observe the surface morphology and the structure of Ni distributed CNT/TiO<sub>2</sub> composite. TEM (JEM2000-FX) at an acceleration voltage of 200 kV was used to investigate the size and distribution among the various metals. TEM specimens were prepared by placing a few drops of the sample solution on a carbon grid. The element mapping over the desired region of the Ni distributed CNT/TiO<sub>2</sub> composite was detected by an EDX analyzer attached to the SEM.

## 2.4. Photocatalytic activity measurement

The photocatalytic activity of Ni distributed CNT/TiO<sub>2</sub> composite was analyzed by decomposition of MB, MO and Rh.B solutions under irradiation of UV light (356 nm, 8 W, 1.2 mW/cm<sup>2</sup>). In an ordinary photocatalytic test performed at room temperature, 0.05 g Ni distributed CNT/TiO<sub>2</sub> composite was added to 50 mL of  $1.0 \times 10^{-5}$  mol/L MB, MO and Rh.B solutions, respectively. Before turning on the UV lamp, the solution mixed with the composite was kept in the dark for at least 2 h, allowing the adsorption/desorption equilibrium to be reached. Then, the solution was irradiated with UV light. The first sample was taken out at the end of the dark adsorption period (just before the light was turned on), in order to determine the MB concentration in the solution, which was hereafter considered as the initial concentration ( $c_0$ ) after dark adsorption. The samples were then withdrawn regularly from the reactor in an order of 30 min, 60 min, 90 min, 120 min and 240 min, and immediately centrifuged to separate any suspended solid. The clean transparent solution was analyzed with a UV-vis spectrophotometer (Optisen POP) at the wavelength from 250 nm to 800 nm. The maximum absorbance ( $\lambda_{max}$ ) was determined at characteristic wavelengths of 665 nm, 465 nm and 665 nm, respectively for the each MB, MO and Rh.B solution degraded.

## 3. Results and discussion

### 3.1. FT-IR analysis

FT-IR was conducted on as-received MWCNTs and functionalized MWCNTs and their corresponding spectra are shown in Fig. 1. For the as-received MWCNTs, the IR spectra do not show almost any functional groups on the surface. However, after oxidation by MCPBA, the MWCNTs exhibit various kinds of functional groups on the surface. The band at 2910 cm<sup>-1</sup> and 2847 cm<sup>-1</sup> can be ascribed to C–H stretching vibration whereas the bands at 1695 cm<sup>-1</sup>, 1303 cm<sup>-1</sup> and 891.75 cm<sup>-1</sup> are ascribed to C=O, S=O and C–H stretching vibrations, respectively. The above observations suggest that the oxidation was promoted in all the treat-

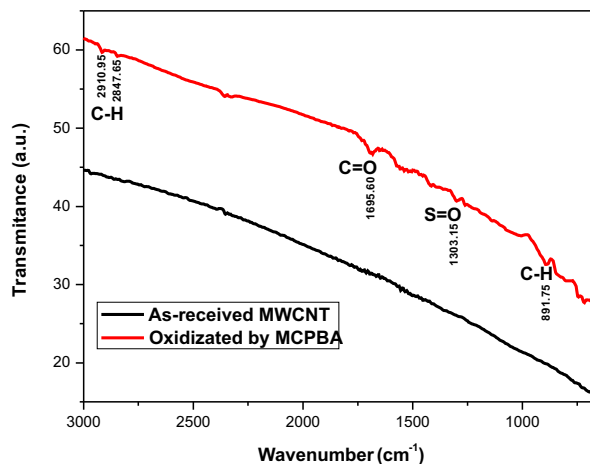


Fig. 1. FT-IR spectra of as-received MWCNTs and MWCNTs functionalized by MCPBA.

ments, and that the functional groups formed in the process, could enhance the active sites on the surface of MWCNTs. Moreover, in the spectra of as-received MWCNTs and functionalized MWCNTs, some small absorption bands around 1500 cm<sup>-1</sup> exist which can be ascribed to C=C stretching vibration. The other peaks at 2350 cm<sup>-1</sup> might be due to the ambient CO<sub>2</sub> in the spectrometer.

### 3.2. XRD analysis

The XRD patterns of CNT/TiO<sub>2</sub> composite and Ni distributed CNT/TiO<sub>2</sub> composite, heat treated at 773 K for 1 h are shown in Fig. 2. The diffraction peaks corresponding to anatase phases and nickel have been marked with 'A' and 'Ni', respectively. The crystal structure of the titanium dioxide is mainly determined by the temperature of heat treatment; the anatase phase formed below 773 K starts to transform to rutile-type structure above 873 K and changes into a single phase of rutile at 973 K–1173 K [20, 21]. In our case, all of the composites were heat treated at 773 K for 1 h. Both CNT/TiO<sub>2</sub> composite and Ni distributed CNT/TiO<sub>2</sub> composite have the peaks at 25.3, 37.8, 48.0, 53.8, 54.9 and 62.5 which can be assigned to diffraction on (101), (004), (200), (105), (211) and (204) planes of anatase (JCPDS: 21-1272). The lack of any other peaks indicates that the TNB was transferred to TiO<sub>2</sub> with anatase structure. More-

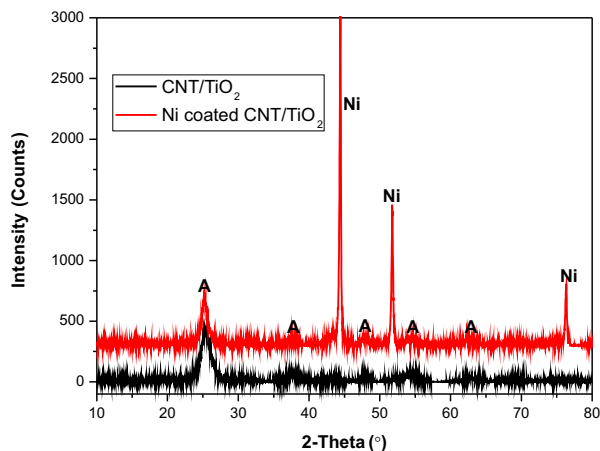


Fig. 2. XRD patterns of CNT/TiO<sub>2</sub> composite and Ni distributed CNT/TiO<sub>2</sub> composite.

over, for Ni distributed CNT/TiO<sub>2</sub> composite, apart from the peaks of anatase, some other peaks at 44.4, 51.7 and 76.3 can be assigned to diffraction on (111), (200) and (220) planes of face-centered-cubic (fcc) nickel (JCPDS: 04-0850) [22]. In addition, no nickel carbides of the cementite phase and nickel oxides were detected. In accordance with these results, one can believe that nickel ions originally distributed on CNT/TiO<sub>2</sub> composite and high purity Ni distributed CNT/TiO<sub>2</sub> composite was prepared. However, the characteristic peaks of MWCNTs can hardly be identified on the XRD patterns of the CNT/TiO<sub>2</sub> and Ni distributed CNT/TiO<sub>2</sub> composites. It can be considered that the peaks of MWCNTs were overlapped by the diffraction peaks attributed to TiO<sub>2</sub>, or the intensity of the MWCNTs peaks was significantly weaker than that of the nickel peaks.

### 3.3. SEM, TEM and EDX analysis

Fig. 3(a) and 3(b) show a SEM micrograph and a TEM image of Ni distributed CNT/TiO<sub>2</sub> composite. As can be deduced from Fig. 3(a), after the deposition of TNB and nickel, the TiO<sub>2</sub> particles and Ni particles, mixed with MWCNTs together. In Fig. 3(b), it can be seen that the TiO<sub>2</sub> aggregates are dispersed homogeneously on the surface of the MWCNTs. However, the presence of Ni particles can't be clearly observed. It can be supposed that most of the Ni particles are present on the sur-

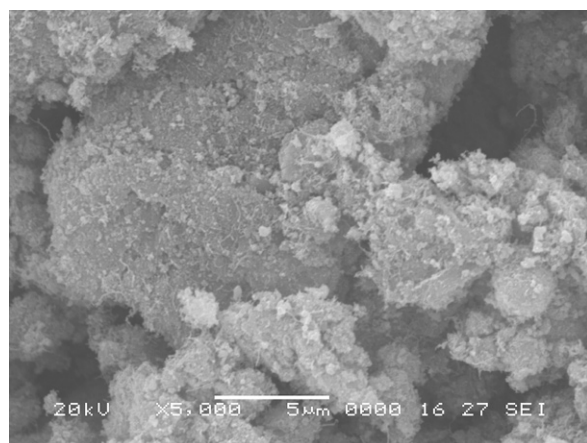
face of the TiO<sub>2</sub> because the nickel precursor was added after the complete synthesis of CNT/TiO<sub>2</sub> composite during the preparation. The morphologies of the particles and MWCNTs are clearly observed. The size of the nanoparticles varies from 10 nm to 20 nm. Most of the particles are quasi-spherical in shape. The elemental analysis by EDX proves that the resulting product contains the elements C, Ti, O and Ni (Fig. 3(c)).

### 3.4. Photocatalytic activity

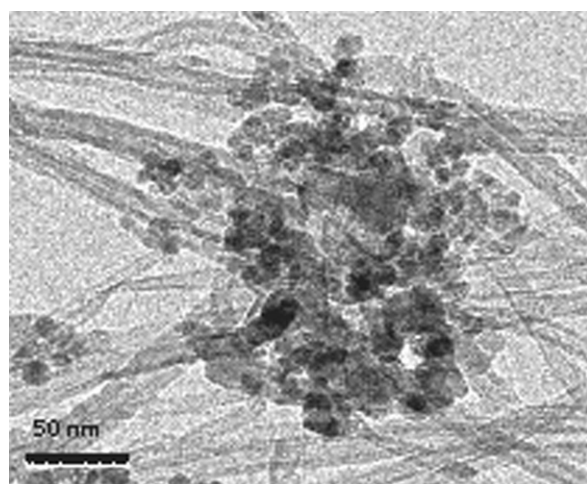
Fig. 4 shows the UV-vis absorbance spectra of MB, MO and Rh.B solutions decomposed by Ni distributed CNT/TiO<sub>2</sub> composite as a function of time. The absorbances of MB, MO and Rh.B solutions show only one maximum, observed at 665 nm, 465 nm and 554 nm, respectively, which is consistent with the data shown in Table 1. We can observe that the maximum values of absorbance for all of the organic dyes decrease with an increase in the UV irradiation time. This suggests that the color of organic dyes has been removed by Ni distributed CNT/TiO<sub>2</sub> composite with increasing the UV irradiation time. After the irradiation with the UV light for 240 min, the absorbance of the organic dyes has almost reached zero, indicating that the color of organic dyes was removed almost completely.

Fig. 5 shows the changes in the relative concentration ( $c/c_0$ ) of MB, MO and Rh.B solutions versus the time of UV irradiation for Ni distributed CNT/TiO<sub>2</sub> composite. Prior to the photocatalytic reaction, a blank test was performed without the composite. The concentration of MB, MO and Rh.B solutions without composite did not change with the time under the UV irradiation (the data are not shown). However, when Ni distributed CNT/TiO<sub>2</sub> composite was used as a photocatalyst under UV light, a significant decrease in concentration of MB, MO and Rh.B solutions was observed after an irradiation time of 120 min, namely about 50 %, 48 % and 34 %, for MB, MO and Rh.B, respectively. When the UV irradiation time was extended to 240 min, the concentration of MB, MO and Rh.B solutions were almost reduced completely, namely about 94 %, 87.5 % and 87.5 %, for MB, MO and Rh.B, respectively. Because the Ni

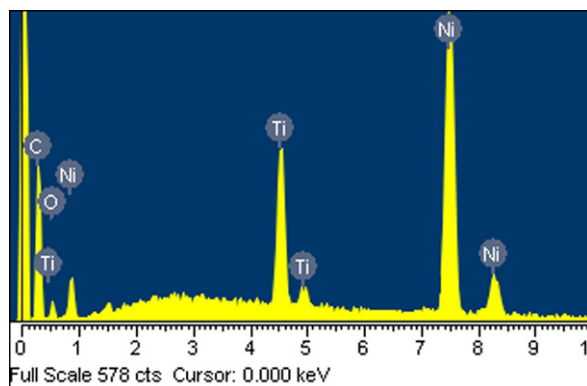




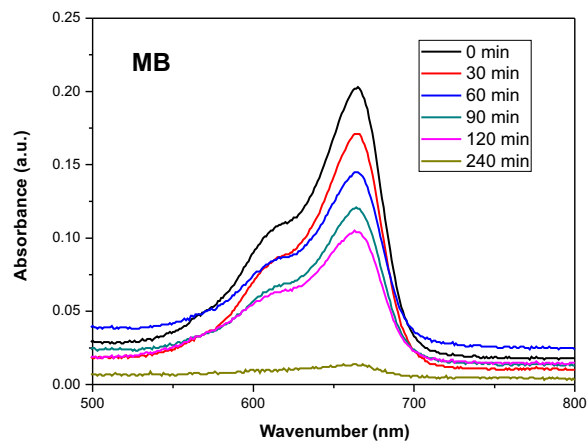
(a)



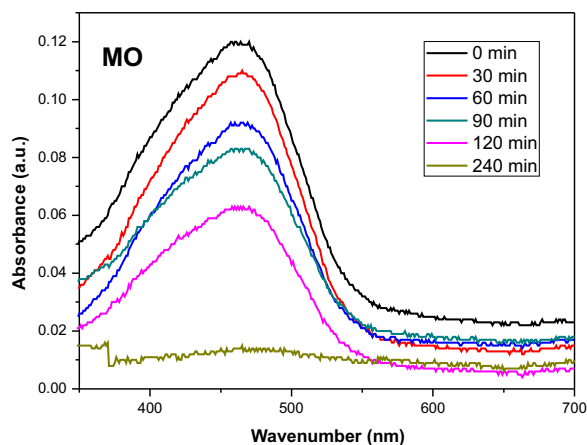
(b)



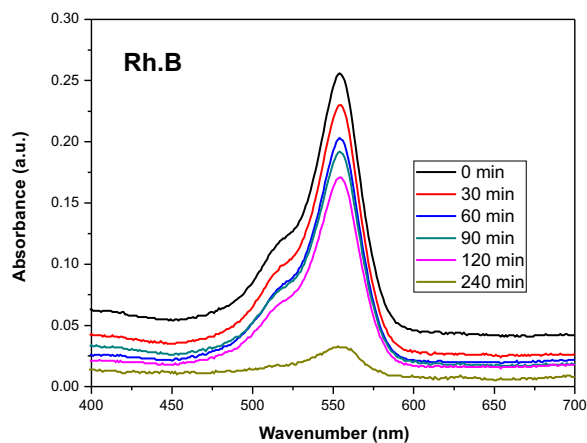
(c)



(a)



(b)



(c)

Fig. 3. SEM photo (a), TEM image (b) and EDX microanalysis (c) of Ni distributed CNT/TiO<sub>2</sub> composite.

Fig. 4. UV-vis absorbance spectra of different organic dyes photodegraded by Ni distributed CNT/TiO<sub>2</sub> composite under UV irradiation versus time.

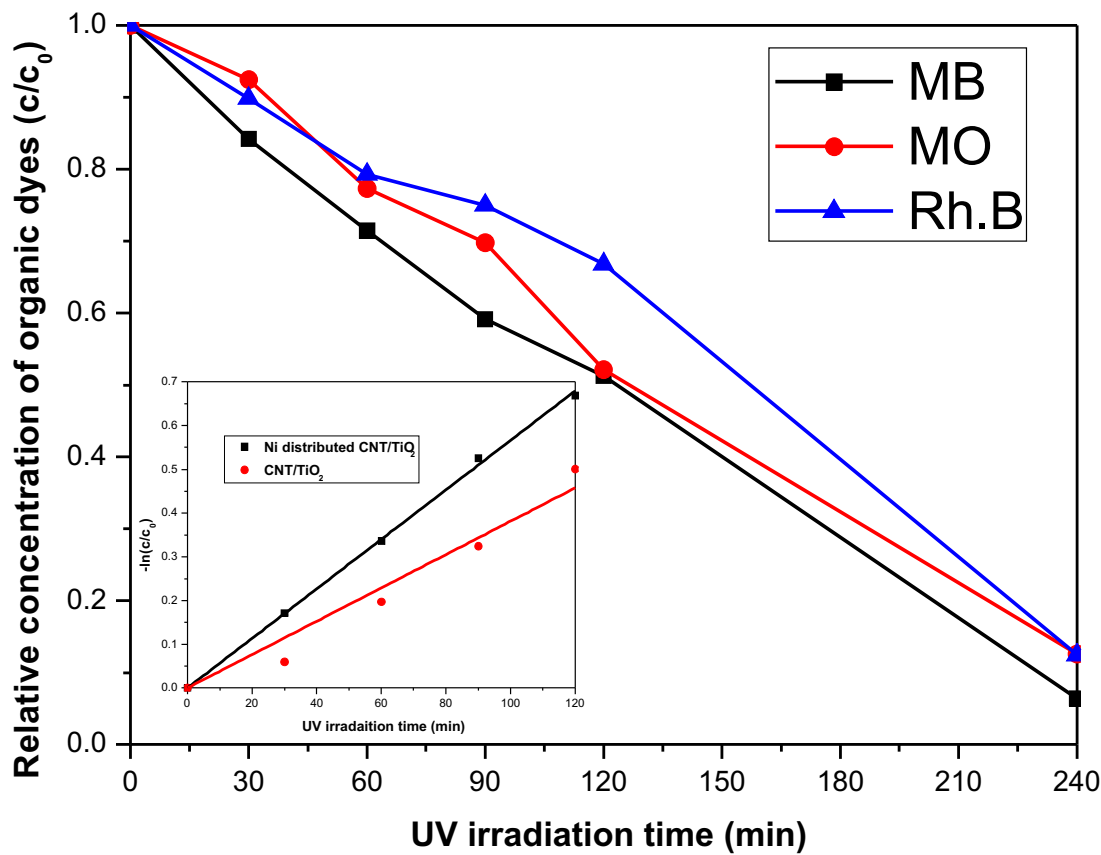


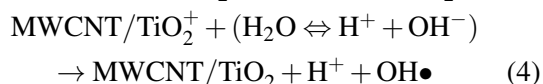
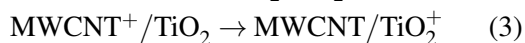
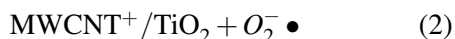
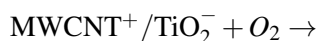
Fig. 5. Relative concentration ( $c/c_0$ ) of organic dyes solution on time of UV irradiation for Ni distributed CNT/TiO<sub>2</sub> composite.

distributed CNT/TiO<sub>2</sub> composite showed the most excellent photodegradation for MB solution, the plot of  $\ln(c/c_0)$  versus  $t$ , for MB degradation with Ni distributed CNT/TiO<sub>2</sub> composite and CNT/TiO<sub>2</sub> composite is presented in the inset in Fig. 5. The values of kinetic constant ( $k_{app}$ ) of the Ni distributed CNT/TiO<sub>2</sub> composite and CNT/TiO<sub>2</sub> composite, which are  $5.66 \times 10^{-3}$  and  $3.81 \times 10^{-3}$ , respectively, have been obtained directly from the slopes of the respective linear curves in the plots. It can be indicated that the degradation rate of MB solution degraded by Ni distributed CNT/TiO<sub>2</sub> composite is faster than that of CNT/TiO<sub>2</sub> composite, almost 1.5 times.

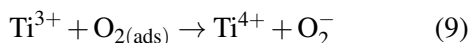
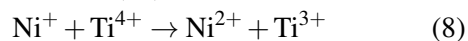
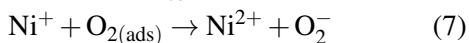
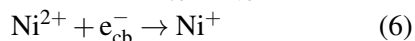
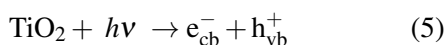
In our previous works [23–25], we indicated that the photocatalytic activity of CNT/TiO<sub>2</sub> composite was much better than that of pristine TiO<sub>2</sub>. The MWCNTs could act as an electron sensitizer and donator in the composite photocatalysts.

Considering the semiconductive properties of carbon nanotubes, MWCNTs may accept the electron ( $e^-$ ), photo-induced by UV irradiation, into the conduction band of the TiO<sub>2</sub> (Eq. (1)). It is considered that the electrons in MWCNTs are transferred into the conduction band in the TiO<sub>2</sub>. In this time, the electrons in the conduction band may react with O<sub>2</sub>, which can trigger the formation of very reactive superoxide radical ion ( $O_2^{\bullet-}$ ). Simultaneously, a positively charged hole ( $h^+$ ) might be formed with an electron transferred from the valence band in TiO<sub>2</sub> to MWCNT (Eq. (3)). The positively charged hole ( $h^+$ ) may react with the OH<sup>-</sup> derived from H<sub>2</sub>O. With this understanding, the role played by MWCNT can be illustrated by injecting electrons into TiO<sub>2</sub> conduction band under UV irradiation and triggering the formation of very reactive radicals such as superoxide radical ion  $O_2^{\bullet-}$  (Eq. (2)) and hydroxyl radical OH $\bullet$  (Eq. (4)). Consequently,

both radical groups (superoxide radical ion O<sub>2</sub><sup>-•</sup> and hydroxyl radical OH•) are responsible for the degradation of the organic compound. The suggested electron transfer between carbon and TiO<sub>2</sub> was experimentally supported by our earlier investigation.



However, for Ni distributed CNT/TiO<sub>2</sub> composite, apart from the effect of MWCNTs, the nickel particle can also act as a probable photogenerated electron trapper in TiO<sub>2</sub> (Eq. (6)), thus ensuring a good effect on the electron-hole pairs separation [26]. Subsequently, Ni<sup>+</sup> could be oxidized to Ni<sup>2+</sup> by transferring the electrons to O<sub>2</sub> absorbed on the surface of TiO<sub>2</sub> (Eq. (7)) and a neighboring surface Ti<sup>4+</sup> (Eq. (8)), which then leads to an interfacial electron transfer (Eq. (9)):



So the Ni distributed CNT/TiO<sub>2</sub> composite shows excellent photocatalytic activity for MB, MO and Rh.B solutions.

## 4. Conclusions

In this study, the Ni distributed CNT/TiO<sub>2</sub> composite was successfully prepared by a simple sol-gel method. Comparison of FT-IR spectra of as-received MWCNTs and the MWCNTs after functionalization by MCPBA showed that various functional groups were formed on the surface. The XRD results revealed the anatase structure of TiO<sub>2</sub> and nickel structure in Ni distributed CNT/TiO<sub>2</sub> composite. From the TEM image, it can be deduced

that TiO<sub>2</sub> aggregates, homogeneously dispersed on the surface of MWCNTs, and most of Ni particles are present on the surface of the TiO<sub>2</sub>. The elemental analysis by EDX proved that the Ni distributed CNT/TiO<sub>2</sub> composite contained the elements C, Ti, O and Ni. According to the decreasing absorbance of MB, MO and Rh.B solutions with an increasing UV irradiation time, it could be indicated that the concentration of MB, MO and Rh.B solutions was decreased by Ni distributed CNT/TiO<sub>2</sub> composite under UV irradiation. When the UV irradiation time extended to 240 min, the concentration of MB, MO and Rh.B solution was decreased almost completely. Because of the electron absorption effect of MWCNTs and electron trapping effect of nickel, the Ni distributed CNT/TiO<sub>2</sub> composite showed excellent photocatalytic activity to decompose MB, MO and Rh.B solutions.

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