Improvement of catalytic activity in styrene oxidation of carbon-coated titania by formation of porous carbon layer

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HIGHLIGHTS

► Carbon-coated titania was prepared by pyrolysis of polystyrene-coated titania.
► Polystyrene-coated titania was produced by in situ polymerization of styrene by using aqueous hydrogen peroxide.
► Porous carbon layer has been formed by treating the carbon-coated titania with KOH solution.
► The porous carbon-coated titania obtained exhibits a high catalytic activity for styrene oxidation.

ABSTRACT

Porous carbon layer has been formed by treating the carbon-coated titania (C@TiO2) with KOH solution. Carbon-coated titania (C@TiO2) was obtained by pyrolysis of polystyrene-coated titania (PS@TiO2), which was produced by in situ polymerization of styrene by using aqueous hydrogen peroxide. The presence of polystyrene and carbon on the surface of titania were confirmed by X-ray photoelectron spectroscopy (XPS) and Fourier transform infrared (FTIR) spectroscopy techniques. Carbon content was about 2.2 wt.% with thickness of carbon layer ca. 5 nm. After treating with KOH solution, PC@TiO2 with the pore size of ca. 5 nm, total pore volume of 0.05 cm3 g−1 and Brunauer–Emmett–Teller (BET) specific surface area of 46 m2 g−1 has been obtained. Catalytic activity results showed that PC@TiO2 gave a higher activity in styrene oxidation compared to bare TiO2, and C@TiO2. The highest catalytic activity was obtained by using PC@TiO2 that obtained after treating C@TiO2 with 1.0 M KOH solution with benzaldehyde and phenylacetaldehyde as the main reaction products. At the higher concentration of KOH solution, the catalytic activity decreased when crystallinity of TiO2 decreased.

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1. Introduction

Titanium dioxide or titania, with chemical formula TiO2, is an ubiquitously used material in everyday life. It has been reported that titanium dioxide is used as white pigment [1], sunscreen and UV absorber [2], electronic data storage medium [3], photocatalyst [4,5], a material in the memristor [6] and Bragg-stack style dielectric mirrors [7]. In addition to this application, titanium dioxide can also be used as a heterogeneous catalyst for synthesis of useful organic compound [8].

Many attempts have been made to improve the catalytic activity of titanium dioxide. One of the strategies is by coating the surface of titania by carbon layer. Carbon-coated titania particles can stabilize anatase structure and increase adsorption amount of organic molecules on the catalyst surface [9]. Several methods have been applied to coat the surface of TiO2 with carbon layer. Preparation of carbon-coated TiO2 by heating the mixture of polyvinyl alcohol (PVA) and commercial TiO2 (P-25) with the ratio of 10/90 under argon atmosphere at 800 °C gave a carbon layer with the proper thickness (2–3 nm). Compared to the original P-25 TiO2 catalyst, carbon-coated TiO2 have much higher adsorption capacity but lower photocatalytic activity. The performance of their photo decomposition was lower than P-25 TiO2 because of the lower dispersion of carbon-coated TiO2 in water and less penetration of UV light by the coated carbon layer reduced the photocatalytic activity because the light reaching the surface of TiO2 particles decreased [10–12]. It also has been reported that the carbon coating on the surface of TiO2 gave a beneficial effect to the catalytic activity. Carbon-coated TiO2 prepared by mixing TiO2 powders and polyethylene glycol (PEG) exhibits higher photocatalytic activity in benzene degradation than that of the pristine TiO2 because of the adsorption enrichment of benzene by carbon around the TiO2 particles.
and of the effective charge separation due to the electronic conduction of carbon [13].

As generally known, the first step in heterogeneous catalysis is the adsorption of the molecules of the reactants on the surface of TiO$_2$ catalyst. It is believed that the carbon layer improved the adsorption of substrate on the surface of catalyst [14]. First, the substrate molecule have to be adsorbed into carbon layer and then to diffuse to the surface of catalyst particle to be reacted. However, this cannot be achieved when the carbon layer is too thick and preventing the substrates to adsorb on the surface of titania. To overcome this problem, one of the strategies is to make the porous carbon layer on the surface of titania particles [12,15,16]. Thus, the substrate may be channelled through the pore system of porous carbon layer to titania surface.

In this work, we herein report a simple method for synthesis of porous carbon-coated TiO$_2$ by treating the carbon-coated titania with KOH solution. Carbon-coated titania (C@TiO$_2$) was obtained by pyrolysis of polystyrene-coated titania (PS@TiO$_2$) that produced by in situ polymerization of styrene using aqueous hydrogen peroxide. It has been reported that the graphitization of carbon was influenced by KOH treatment [17]. One expects that the carbon layer become porous after this treatment. Here, we demonstrated a method to synthesize porous coated titania. The presence of porous carbon in the surface of titania gave the beneficial in liquid phase oxidation of styrene with aqueous hydrogen peroxide.

The oxidation of styrene by using aqueous hydrogen peroxide is of considerable interest for academic research and utilization in the industry for the synthesis of important products such as benzaldehyde, styrene oxide and phenylacetaldehyde. The use of the aqueous hydrogen peroxide for oxidizing organic substrates has received much attention because of its environmental implications, water being the only chemical by-product of oxidation reactions. It is known that the styrene oxidation by hydrogen peroxide at the side chain can lead to various reaction products, depending on the catalyst and reaction conditions. Two major reactions take place during styrene oxidation depending on the nature of the catalyst and the reaction conditions. They are the oxidative C=C cleavage into benzaldehyde and epoxidation followed by isomerization into phenylacetaldehyde [18].

2. Experimental

2.1. Preparation of polystyrene-coated TiO$_2$ and carbon-coated TiO$_2$

Polystyrene-coated TiO$_2$ (PS@TiO$_2$) was prepared by using TiO$_2$ (supplied by Aldrich with over 99.9% purity), styrene and aqueous hydrogen peroxide. Typically, 30% aqueous hydrogen peroxide (1 ml) was added drop wise to 1 g of TiO$_2$ powder and then styrene was added to the mixture with the molar ratio of styrene to TiO$_2$ was 3:1. The mixture was heated under stirring at 100°C for 8 h. The product was then washed with ethanol, sonificated and centrifuged three times and dried at 100°C overnight. The polystyrene-coated TiO$_2$ obtained labeled as PS@TiO$_2$. The PS@TiO$_2$
obtained was then pyrolyzed at 450 °C for 1 h with the heating rate was 10 °C min⁻¹ in flow nitrogen to produce carbon-coated titania (C@TiO₂). In order to produce porous-carbon coated TiO₂, the C@TiO₂ (0.3 g) was treated with KOH solution (2 ml) in a various concentrations: 0.5 M; 1.0 M; 1.5 M and 2.0 M. After heating at 450 °C under a N₂ flow for 1 h, the treated sample was washed with deionized water until neutral (pH = 7), dried at 100 °C overnight and labeled as PC@TiO₂.

2.2. Characterizations

The FTIR spectra of samples were taken by the following protocol. The samples were mixed with dry KBr (1:100 ratio) in a mortar and pressed at 8 tons for 3 min to obtain the self-supporting pellets. All measurements were performed at an ambient temperature over the range 400–4000 cm⁻¹ using Perkin–Elmer Spectrum spectrometer. Diffuse reflectance UV–Vis (DR UV–Vis) spectra were recorded under ambient conditions on a Perkin Elmer Lambda 900 UV/VIS/NIR spectrometer. The spectra were monitored in the range of wavenumber 200–450 nm.

The morphology and structural investigation were performed by field emission scanning electron microscope (FESEM) and transmission electron microscope (TEM). Prior to sample scanning, the powder sample was attached to sample holder by using double-sided tape. The samples were then coated with platinum, and the image was taken on a JOEL JED-2300 analysis station.

The TEM images of bare TiO₂, carbon-coated TiO₂ before and after treating with KOH solution were taken with a JEOL JEM-2100 transmission electron microscope. Specimens were prepared by grinding the material into a fine powder, dispersing the powder in acetone under the ultrasonic bath for 30 min. The dispersion was then dropped on a carbon copper grid follow by drying at room temperature. The microscope was operated at an accelerating voltage of 200 kV. The X-ray photoelectron spectroscopy (XPS) measurements were performed on a high resolution Auger electron spectrometer AXIS ULTRA (Kratos Analytical, Shimadzu) with monochromatic Al Kα X-ray.

Thermogravimetric analysis of carbon-coated TiO₂ was carried out in TGA-SDTA 851e METTLER thermogravimetry instrument. The mass of sample was 10 mg and the reaction environment was flowing nitrogen with the heating rate was 10 °C min⁻¹. The carbon content of sample was determined from the weight loss of sample after heating from room temperature to 600 °C in air. Carbon content was determined by calculating the weight loss of sample.

Specific surface area of samples was determined by nitrogen adsorption instrument (Micromeritics ASAP 2010) utilizing the Brunauer–Emmett–Teller (BET) method [19]. The sample was degassed at 150 °C overnight prior to the measurement. Pore size distribution was calculated from the adsorption/desorption branch of isotherms by the Barrett–Joyner–Halenda (BJH) model [20].

The X-ray diffraction pattern of samples was collected on a Bruker AXS D8 Automatic Powder Diffractometer using the Cu Kα radiation with λ = 1.5406 Å at 40 kV and 40 mA, over the range 2θ = 2–90° at step 0.030 and step time 1 s (scanning speed of 1.2° min⁻¹). The particle size of the samples was calculated by using the Scherrer’s equation based on XRD data:

\[ L = \frac{K \lambda}{\beta \cos \theta} \]

where K is the shape factor, λ is the wavelength of the X-ray used, β is the full-width at half-maximum (FWHM) and θ is the angle between the incident and the scattered X-ray.
The reaction was carried out in oil bath under stirring at 80°C. The reaction mixture containing styrene (5 mmol), catalyst (50 mg), aqueous H₂O₂ (8 mmol) and acetonitrile (5 ml) as solvent were put in a round-bottom flask equipped with condenser. The reaction was carried out in oil bath under stirring at 80°C for 1 min followed by heating to 150°C at 10°C min⁻¹. The final temperature (150°C) was then kept for an additional 0.5 min. The product yield and styrene conversion were calculated according to the way of previous report [21].

3. Results and discussion

3.1. Physical properties

Fig. 1 shows schematic diagram of the preparation of PS@TiO₂, C@TiO₂ and PC@TiO₂ particles and their FESEM and TEM photographs. Hydrogen peroxide can be easily adsorbed on the TiO₂ surface due to hydrophilicity of TiO₂, allowing the formation of polystyrene on the surface of TiO₂. The polystyrene layer was then converted into the carbon layer by pyrolysis. The PC@TiO₂ is prepared after treating C@TiO₂ with KOH solution. Fig. 1a shows that TiO₂ particles had irregular shape and are in the form of aggregation. As shown in FESEM images in Fig. 1a, c and d, there is no change in the shape of PS@TiO₂, C@TiO₂ and PC@TiO₂ particles after modification. The transmission electron micrograph of bare TiO₂, carbon-coated titanium dioxide before and after treating with KOH is given in the Fig. 1. The C@TiO₂ image (Fig. 1b) shows a dark part that are reasonably supposed to be TiO₂ particles, which were surrounded by a thin layer with faint contrast, which are believed to be a carbon layer. Carbon content of sample is about 2.2 wt.% with thickness of carbon layer that covered titanium dioxide surface is about ca. 5 nm. Fig. 1d shows FESEM and TEM images of PC@TiO₂ after treated with 1.0 M KOH solution. It can be seen that the carbon layer has been changed into a porous form.

The existence of polystyrene on the TiO₂ surface was also verified by FTIR spectrometer. FTIR spectrum of the PS@TiO₂ is shown in Fig. 2c. The characteristic bands of polystyrene were clearly observed in polystyrene-coated titanium dioxide sample: C–H aromatic stretching vibration at 3058 and 3024 cm⁻¹, C–H stretching vibration at 2920 and 2847 cm⁻¹, aromatic C–C stretching vibration around 1451–1492 cm⁻¹, C–H out of plane bend at 757 cm⁻¹ and aromatic C–C out of plane band at 697 cm⁻¹. Aromatic overtones were visible in the range 1703–1943 cm⁻¹. These bands almost disappeared in the sample after pyrolysis at 450°C for 1 h (Fig. 2d), showing that most of the polystyrene was removed and converted to carbon.

The FTIR spectra of carbon-coated titanium dioxide samples (Fig. 2d) show absorption peaks at about 3400 and 1630 cm⁻¹ which are associated with the stretching vibrations of surface water molecules. Peaks around 3000 cm⁻¹ were often assigned to molecularly chemisorbed water and the existence of hydroxyl groups. Meanwhile, the peaks corresponding to the stretching vibrations of water and hydroxyl groups were broader and stronger in the carbon-coated titanium dioxide (C@TiO₂) than that of the bare TiO₂, indicating that the samples have different hydrophilicity.

Diffuse reflectance UV–Vis (DR UV–Vis) spectra of bare TiO₂, polystyrene, PS@TiO₂ and C@TiO₂ are shown in Fig. 3. It can be seen that there are two absorbance peaks at 250–260 and 260–330 nm. The absorbance peak at 250–260 nm is attributed to a charge transfer of the tetrahedral titanium site between O²⁻ and the central Ti(IV) atom, while octahedral Ti was reported appeared around 250–300 [8]. The intensity of band at 250–260 nm on polystyrene-coated titania was increased due to the polystyrene (Fig. 3c). After pyrolysis at 450°C for 1 h, DR UV–Vis spectrum of C@TiO₂ sample (Fig. 3d) showed the intensity of absorbance peak at 250–260 nm which is corresponded to peak of polystyrene, was reduced due to conversion of polystyrene to carbon on the surface of TiO₂.

The XPS measurements were used to study the chemical state of the polystyrene-coated TiO₂ (PS@TiO₂), carbon-coated TiO₂ (C@TiO₂) and carbon-coated TiO₂ after treating with 1.0 M KOH solution (PC@TiO₂). The XPS peaks show the characteristic binding energy of elements. It revealed that the C (1s) spectrum of PS@TiO₂...
(Fig. 4a) showed two components centered at 284.5 eV (C–C and C–H bonds), and at 291.4 eV which correspond to a shake-up satellite of π–π* of the aromatic ring. The two peaks were fitted to four components centered at 284.5 eV and 286.2 eV (C–C and C–H bond) while peaks centered at 290.7 eV and 292.4 eV correspond to C=C aromatics. These peaks confirm the existence of polystyrene on the sample. These results are in agreements with previous report [22].

The C (1s) peak of carbon-coated titanium dioxide (C@TiO$_2$) as shown in Fig. 4b displays one peak centered at 284.5 eV which correspond to the graphitic thin carbon layer [23]. This C (1s) peak was fitted to two peaks centered at 284.5 eV and 286.2 eV. Based on the above results, one concludes that the binding energy of polystyrene and graphitic is different. The C (1s) peak centered at 291.4 eV which correspond to a shake-up satellite of π–π* of the aromatic ring which appeared at PS@TiO$_2$ XPS spectrum was not observed in C@TiO$_2$. This result proved that polystyrene layer has been successfully converted into the carbon layer on the surface of TiO$_2$.

The C (1s) peak of carbon-coated titanium dioxide after treatment with 1.0 M KOH solution (PC@TiO$_2$) is given in Fig. 4c. This peak centered at 284.5 eV and was fitted to three peaks centered at 283.4 eV, 284.5 eV and 285.7 eV. Fig. 4d shows the wide spectra of C@TiO$_2$ after treatment with 1.0 M KOH solution. It can be seen that there is no peak at binding energy at 292–296 eV which correspond to K (2p) was observed. It can explain the absence of adsorbed aqueous hydrogen peroxide on the surface of titanium dioxide particles. It is also observed that after pyrolysis of PS@TiO$_2$ at 450 °C for 1 h, the hydrophobic character of polystyrene makes the PS@TiO$_2$ particles were floating on water (Fig. 5b). After pyrolysis at 450 °C for 1 h, the hydrophobic character of the C@TiO$_2$ particles was still maintained and made the sample float above the water’s surface (Fig. 5c). It confirmed that the carbon layer of C@TiO$_2$ and PC@TiO$_2$ are hydrophobic and hydrophilic, respectively. As shown in Fig. 5d, the PC@TiO$_2$ particles settle down to the bottom of the tube. This result indicates that water can be easily adsorbed on the PC@TiO$_2$ particles due to the porous structure of its carbon layer.

The nitrogen adsorption–desorption isotherm of C@TiO$_2$ and PC@TiO$_2$ samples are shown in Fig. 6. Specific surface area of C@TiO$_2$ is 13 m$^2$ g$^{-1}$ and after treating with KOH solution 1.0 M the specific surface area is increased to 46 m$^2$ g$^{-1}$, while the initial BET specific surface area of TiO$_2$ is 11 m$^2$ g$^{-1}$. The increment specific surface area of C@TiO$_2$ and PC@TiO$_2$ is almost a factor of 3.5, which is attributed to the contribution of porous structure of the carbon. It can be seen from Fig. 6 that the isotherms can be categorized as type IV with hysteresis loops at relative pressure ($P/P_0$) between 0.4 and 1.0, confirming its mesoporous feature. Pore size distribution calculated by the Barrett–Joyner–Halenda (BJH) method indicated that the C@TiO$_2$ was nonporous (inset of Fig. 6a) with the total pore volume 0.02 cm$^3$ g$^{-1}$ and 0.03 cm$^3$ g$^{-1}$ by the adsorption and desorption branch respectively. From the calculation of pore size distribution there are indications of pore in size of 6.06 nm, with very small pore volume. This lack of porosity was also confirmed by TEM (see Fig. 1c). The pore size distribution of PC@TiO$_2$ shows a narrow peak centered at ~5.0 nm (in the inset of Fig. 6b) indicates the existence of mesopores (2–5 nm in size). The average pore diameter was ca. 5 nm calculated from the adsorption branch of the N$_2$ sorption isotherm. The total pore volume of PC@TiO$_2$ was 0.05 cm$^3$ g$^{-1}$ and 0.095 cm$^3$ g$^{-1}$ given by the adsorption and desorption branch of the N$_2$ isotherm.

Based on the above discussion, it is clearly demonstrated that PS@TiO$_2$ can be prepared by in situ polymerization of styrene by adsorbed aqueous hydrogen peroxide on the surface of titanium dioxide particles. It is also observed that after pyrolysis of PS@TiO$_2$ at 450 °C for 1 h, the polystyrene converted to carbon layer to produce C@TiO$_2$. When C@TiO$_2$ obtained was treated with KOH solution, the porous carbon layer was formed on the surface of TiO$_2$.

### 3.2. Catalytic activity

The oxidation of styrene with aqueous H$_2$O$_2$ as an oxidizing agent for 8 h at 80 °C was used to evaluate the catalytic activity of the catalysts. Fig. 7 presents the results of oxidation of styrene over bare TiO$_2$, carbon-coated TiO$_2$ before and after treating with...
Fig. 6. \( \text{N}_2 \) adsorption/desorption isotherm of: (a) C@TiO\(_2\) and (b) PC@TiO\(_2\). The insets are pore size distribution calculated by BJH method.

Fig. 7. The yield of products in oxidation of styrene by using aqueous hydrogen peroxide in the system containing: (a) no catalyst, (b) TiO\(_2\), (c) C@TiO\(_2\) and PC@TiO\(_2\) which is obtained after C@TiO\(_2\) treated with KOH solution (d) 0.5 M, (e) 1.0 M (f) 1.5 M and (g) 2.0 M.

Fig. 8. The conversion of styrene in the styrene oxidation by using aqueous hydrogen peroxide in the system containing: (a) no catalyst, (b) TiO\(_2\), (c) C@TiO\(_2\) and PC@TiO\(_2\) which is obtained after C@TiO\(_2\) treated with KOH solution (d) 0.5 M, (e) 1.0 M (f) 1.5 M and (g) 2.0 M.

Fig. 9. The X-ray diffractograms of: (a) TiO\(_2\), (b) C@TiO\(_2\) and PC@TiO\(_2\) which is obtained after C@TiO\(_2\) treated with KOH solution (c) 0.5 M, (d) 1.0 M (e) 1.5 M and (f) 2.0 M.
KOH solution. Gas chromatography analyses indicated that benzaldehyde and phenylacetaldehyde were the main products, and styrene oxide as minor product. All of these products have been reconfirmed by GC–MS analyzed. As shown in Fig. 7, the present result indicated that the selectivity towards the formation of benzaldehyde is much higher than that of phenylacetaldehyde. This result is different from those previously reported that styrene oxidation using TS-1 gave phenylacetaldehyde as a main product [24–28].

Fig. 7 shows that the yield of styrene oxidation products without catalyst, with TiO$_2$ and C@TiO$_2$ is almost similar. C@TiO$_2$ gave slightly decreased in the formation of products because nonporous carbon layer that covered TiO$_2$ reduced the amount of styrene reaching the TiO$_2$ active site. After treating with 0.5 M KOH solution, carbon layer on C@TiO$_2$ turns into the porous structure to produce PC@TiO$_2$. This allowed more of styrene molecules which were adsorbed on PC@TiO$_2$ carbon layer to reach the TiO$_2$ active sites and gave higher catalytic activity (192 l mol of benzaldehyde and 35 l mol of phenylacetaldehyde). The highest product was obtained by using PC@TiO$_2$ (260 l mol of benzaldehyde and 41 l mol of phenylacetaldehyde). As shown in Fig. 8, the highest conversion of styrene, ca. 27%, was obtained when PC@TiO$_2$, C@TiO$_2$ after treating with 1.0 M KOH, was used as catalyst.

It has been reported that crystallinity and crystallite size of TiO$_2$ was effect the catalytic activity of TiO$_2$ [29,30]. One suggests that the concentration of KOH used to create the porosity of the carbon layer affects the crystallinity and crystallite size of TiO$_2$. Fig. 9 shows the X-ray diffractogram of TiO$_2$, C@TiO$_2$, and PC@TiO$_2$ after treated with different concentrations of KOH. All samples have shown similar peaks with the highest peak at 25.68° which was indicated as (1 0 1) plane of the crystal phase of anatase. In general, the intensity of the X-ray peaks decreases with increasing the KOH concentration suggesting that the crystallinity of the TiO$_2$ affect the catalytic activity in styrene oxidation. This is the reason why the catalytic activity of PC@TiO$_2$ treated with 1.5 and 2.0 M KOH is lower compared than that of PC@TiO$_2$ treated with 1.0 M KOH solution. It is also suggested that the optimum concentration of KOH solution for producing high porosity carbon layer and relatively high crystallinity of TiO$_2$ was 1.0 M. The distribution of crystallite sizes of TiO$_2$, C@TiO$_2$, PC@TiO$_2$ and after treated with KOH calculated by Scherrer’s formula was in the range of 30.6–33.5 nm. It can be seen that there is no significant differences of average particle size of the samples.

In the styrene oxidation reaction, the free-radical mechanism involves the interaction of hydrogen peroxide with the TiO$_2$ to form a hydroperoxy species. This species being unstable, undergoes one-electron reduction with a loss of water molecule and rearranges to give the Ti peroxo species (Ti(IV)–O/C$^\cdot$ radical). The peroxo species interacts with styrene to form a π-bonded transient species. The presence of the polar water molecules in the reaction system catalyzed the cleavage of the C–C bond of styrene to produce benzaldehyde through radical transformation. While the phenylacetaldehyde obtained from styrene oxide isomerisation [31]. Based on these considerations, a model of the possible mechanism of the oxidation of styrene with aqueous H$_2$O$_2$ as an oxidation agent over PC@TiO$_2$ is proposed (see Fig. 10).

4. Conclusion

Porous carbon coating of titania particle was possible prepared by in situ polymerization of styrene on the TiO$_2$ surface followed by pyrolysis of polystyrene formed into carbon and treatment with
KOH solution under N₂ gas flow. The existence of polystyrene and carbon were confirmed by FTIR, DR UV–Vis, TEM and XPS. Carbon content was about 2.2 wt.% with thickness of carbon layer ca. 5 nm. Catalytic results showed that the carbon layer is thick enough that hinder the substrate to reach the TiO₂ active site. Treatment of KOH solution on carbon-coated TiO₂ caused the carbon layer turns into porous carbon which allowed the substrate to reach the TiO₂ active site and gave a higher catalytic activity in styrene oxidation. The highest catalytic activity was obtained by using carbon-coated titania treated with 1.0 M KOH solution with benzaldehyde and phenylacetaldehyde as main products. At the higher concentration caused the carbon layer turns.

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