

Application of TiO₂ Photocatalyst Deposited on SiC for Degradation of Organic Compounds Diluted in Water

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AZojomo (ISSN 1833-122X) Volume 3 December 2007

Abstract

TiO₂-SiC photocatalyst was prepared by a calcination of TiC-SiC nano particle precursor synthesized by the carbothermic reduction method. On TiO₂-SiC photocatalyst the formation of the mixture of anatase and rutile phases of TiO₂ was observed after the calcination at 873 K, while no crystallized TiO₂ phase was formed on TiO₂-SiO₂ photocatalyst prepared by a conventional sol-gel method and calcined at 873 K. TiO₂-SiC photocatalyst decomposed 2-propanol into acetone, CO₂, and H₂O when UV-light was irradiated and finally acetone was degraded into CO₂, and H₂O. TiO₂-SiC exhibited the higher photocatalytic reactivity than TiO₂-SiO₂ photocatalyst. Amount of water adsorption on TiO₂-SiC photocatalyst was much smaller than TiO₂-SiO₂ photocatalyst. Well-crystallized TiO₂ deposited on SiC and hydrophobic surface of SiC are important factors for enhancement of photocatalytic degradation of organic compounds diluted in water.

Keywords

TiO₂ Photocatalyst, SiC Support, Degradation of Pollutants

Introduction

The design of highly efficient photocatalytic systems which work for the reduction of global atmospheric pollution and the purification of polluted water is attractive and one of the most desirable yet challenging goals in the research of environmentally-friendly catalysts [1-8]. TiO₂ photocatalysts have been attracting much attention in recent years, because TiO₂ photocatalyst can completely mineralize toxic and nonbiodegradable organics to CO₂, H₂O, and inorganic constituents [3]. Many research have been undertaken to use TiO₂ photocatalyst for purification of polluted water. During the process of purification of polluted water, photocatalyst has to be separated from the treated water. To separate the photocatalyst from water, photocatalyst should be supported on bulk materials. As photocatalyst support, SiO₂ and zeolite has been widely used [1, 3-7]. On the other hand, silicon carbide (SiC) has physical bulk properties such as high thermostability, high mechanical strength, and high electrical conductivity and has been used as catalyst support [8-12]. In addition, SiC is easily molded into a filter and has been already used for exhaust emission filter of automobile [10]. Although it may be a useful support for photocatalysts used in liquid phase, there

have been no reports on the properties of TiO₂ photocatalyst deposited on SiC. In the present study, we have dealt with the preparation and characterization of TiO₂ photocatalysts deposited on SiC support using the TiC-SiC nano particle precursor synthesized by the carbothermic reduction method [13] and demonstrated the successful utilization of this TiO₂-SiC catalyst for the photocatalytic degradation of 2-propanol diluted in water.

Experimental Procedure

To synthesize TiC-SiC precursor, titanium tetraisopropoxide (TPOT), tetraethyl orthosilicate (TEOS), and phenolic resin were mixed at room temperature. After the continuous stirring of the mixtures to the gelation, the gels obtained were pyrolyzed at 1273 K under a nitrogen atmosphere. This material was preheated at 1873 K under Ar flow to yield the TiC-SiC precursor by the carbothermic reduction [13]. The obtained TiC-SiC powders (Ti/Si=0.4 in molar ratio) were then calcined in air at 873 K to synthesize TiO₂-SiC photocatalyst. TiO₂-SiO₂ photocatalyst (Ti/Si=0.4 in molar ratio) were prepared by the sol-gel method using TEOS, TTIP and ethanol and calcined in air at 873 K. The X-ray Absorption Near Edge Structure (XANES) spectra were measured at the BL-7C facility of the Photon Factory at the National Laboratory for High-Energy Physics, Tsukuba. X-ray diffraction patterns of the samples were measured by Rigaku RINT2500 diffractometer with CuK α radiation. X-ray photoelectron spectroscopy (XPS) was recorded with JEOL microprobe system using the Mg K α line. The photocatalyst (50 mg) was transferred to a quartz cell with an aqueous solution of 2-propanol (2.6×10^{-3} mol dm⁻³, 25 ml). Prior to UV-light irradiation, the suspension was stirred in a flow of O₂ for 1 h under dark conditions. The sample was then irradiated at 295 K using UV light ($\lambda > 250$ nm) from a 100 W high-pressure Hg lamp with continuous stirring under O₂ atmosphere in the system. The products were analyzed by gas chromatography. Water adsorption isotherms of the catalysts was measured at 293 K using a conventional vacuum system.

Results and Discussion

Figure 1 shows the XRD patterns of TiC-SiC precursor and TiO₂-SiC photocatalyst prepared from TiC-SiC precursor and TiO₂-SiO₂ photocatalyst prepared by the sol-gel method and calcined at 873 K. In the TiC-SiC sample, very sharp XRD peaks assignable to TiC and SiC were observed, indicating well-crystallized TiC-SiC precursor was obtained. After calcination at 873 K, anatase and rutile phases from the crystalline TiO₂ were observed on TiO₂-SiC photocatalyst. However, TiO₂-SiO₂ photocatalyst calcined at 873 K exhibited no XRD peaks due to the crystallized phases, indicating that the TiO₂ species exist in an amorphous phase or as ultrafine particles. TiO₂-SiO₂ photocatalyst calcined at 1173-1473 K exhibited the XRD peaks of anatase but the crystallinity was lower than that of TiO₂-SiC photocatalyst calcined at the same temperature and the surface area decreased significantly at the high calcination temperature, surface area changed from 169 m²g⁻¹ at 873 K to 0.6 m²g⁻¹ at 1473 K.

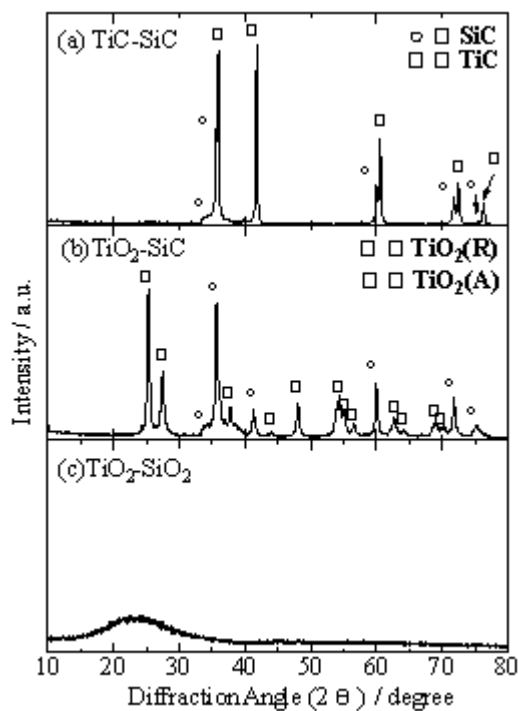


Figure 1. XRD patterns of TiC-SiC precursor (a), TiO₂-SiC photocatalyst (b), and TiO₂-SiO₂ photocatalyst (c).

The XANES spectra at Ti K-edge of TiO₂-SiC photocatalyst and TiO₂-SiO₂ photocatalyst are shown in Figure 2. The XANES spectra at the Ti K-edge show several well-defined pre-edge peaks which are related to the local structure surrounding the Ti atom. The relative intensities of these pre-edge peaks provide useful information on the coordination number of the Ti atom [1]. TiO₂-SiC photocatalyst exhibited three small well-defined pre-edge peaks which can be assigned to a presence of a mixture of anatase and rutile with high crystallinity. However, TiO₂-SiO₂ photocatalyst exhibit an intense single pre-edge peak, indicating that part of titanium oxide species have a tetragonal coordination in the SiO₂ matrix [5].

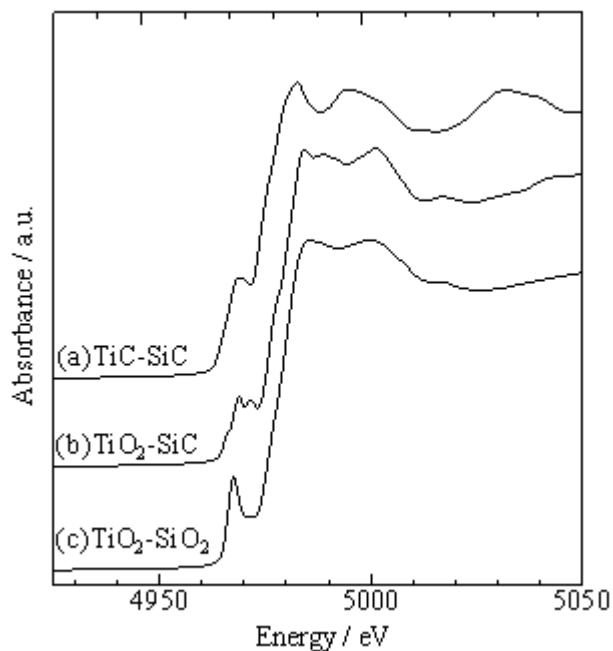


Figure 2. Ti K-edge XANES spectra of TiC-SiC precursor (a), TiO₂-SiC photocatalyst (b), and TiO₂-SiO₂ photocatalyst.

Figure 3 shows the Ti 2p and Si 2p XPS spectra of TiO₂-SiC photocatalyst and TiO₂-SiO₂ photocatalyst. The position of Ti 2p XPS band indicated that TiO₂ existed on the surface of both photocatalysts. Comparing TiO₂-SiC with TiO₂-SiO₂, peak intensity corresponding to Ti 2p_{3/2} and Ti 2p_{1/2} on TiO₂-SiC was higher than that on TiO₂-SiO₂. This result indicates that the content of TiO₂ on TiO₂-SiC surface is higher than that on TiO₂-SiO₂ surface and most part of TiO₂ species of TiO₂-SiO₂ exists inside of SiO₂ matrix. Si 2p spectrum revealed that SiC on the surface of TiO₂-SiC was partially oxidized into SiO₂ by the calcination of TiC-SiC.

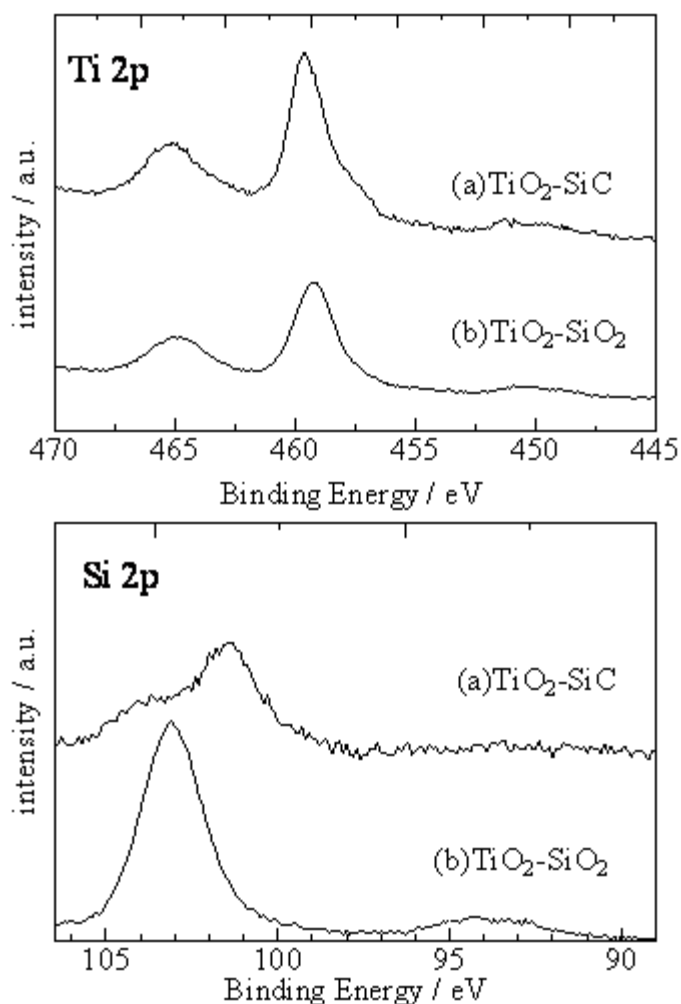


Figure 3. Ti 2p and Si 2p XPS spectra of TiO₂-SiC photocatalyst (a) and TiO₂-SiO₂ photocatalyst (b).

To investigate the photocatalytic reactivity of TiO₂-SiC photocatalyst, degradation of 2-propanol diluted in water was carried out under UV-light irradiation. Figure 4 shows the reaction time profiles of the liquid-phase photocatalytic reaction on the TiC-SiC precursor, TiO₂-SiC photocatalyst, and TiO₂-SiO₂ photocatalyst. When UV-light turned on, photocatalyst decomposed 2-propanol into acetone, CO₂, and H₂O, and finally, acetone was also decomposed into CO₂ and H₂O. On the other hand, TiC-SiC precursor did not decompose to 2-propanol. TiO₂-SiC photocatalyst decomposed 2-propanol faster than TiO₂-SiO₂ photocatalyst. This indicates that SiC is efficient support for nano-sized TiO₂ photocatalyst which can be utilized for degradation of organic compounds in water.

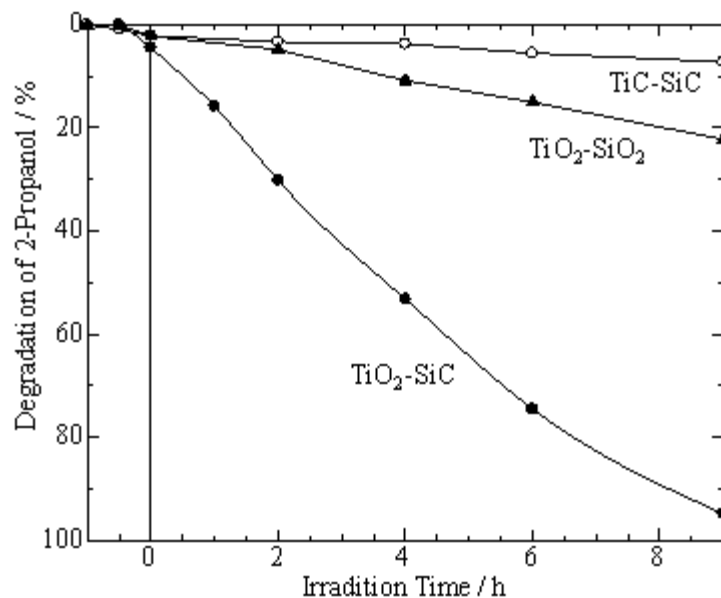


Figure 4. Photocatalytic degradation of 2-propanol diluted in water on TiC-SiC precursor, TiO₂-SiC photocatalyst, and TiO₂-SiO₂ photocatalyst.

Figure 5 shows the photocatalytic reaction rate normalized by weight of TiO₂ included within catalysts after 6 h irradiation on TiO₂-SiC photocatalyst, TiO₂-SiO₂ photocatalyst, and the amounts of water adsorption on TiO₂-SiC photocatalyst and TiO₂-SiO₂ photocatalyst are also shown. TiO₂-SiC photocatalyst exhibited the higher photocatalytic reactivity than TiO₂-SiO₂ photocatalyst and the amount of water adsorption on TiO₂-SiC photocatalyst is much smaller than on TiO₂-SiO₂ photocatalyst. The hydrophobic property of TiO₂-SiC photocatalyst may be caused by the hydrophobic property of SiC surface. This indicates that well-crystallized TiO₂ deposited on SiC has an efficient photocatalytic reactivity and hydrophobic surface of SiC support is also very important factor to realize the efficient photocatalytic reactivity for the liquid phase reaction.

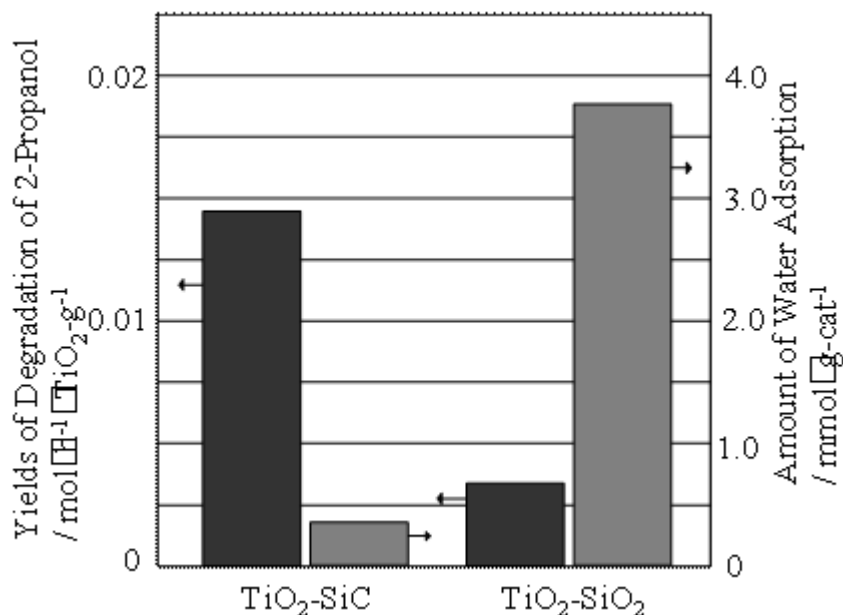


Figure 5. Photocatalytic reactivity for 2-propanol and amount of water adsorption on TiO₂-SiC photocatalyst and TiO₂-SiO₂ photocatalyst.

Conclusions

TiO₂ photocatalysts deposited on SiC (TiO₂-SiC) prepared by the calcination of the TiC-SiC precursor showed high photocatalytic reactivity for the degradation of 2-propanol diluted in water. TiO₂-SiC photocatalyst showed the higher photocatalytic reactivity than the TiO₂-SiO₂ photocatalyst prepared by the conventional sol-gel method. The formation of well-crystallized TiO₂ on SiC and the hydrophobic surface of SiC support were found to be related to the efficient photocatalytic reactivity of TiO₂-SiC photocatalyst. Since SiC is mechanically strong enough to be used as a filter for water purification, SiC is a good candidate as support for TiO₂ photocatalysts used in liquid phase reactions.

Acknowledgements

The present work is supported by the Grant-in-Aid for Scientific Research (KAKENHI) in Priority Area "Molecular Nano Dynamics" from Ministry of Education, Culture, Sports, Science and Technology (No. 17034036, No. 17360388). This work is partly performed under the project of collaborative research at the Joining and Welding Research Institute (JWRI) of Osaka University. The X-ray adsorption experiments were performed at the Photon Factory of KEK (2004G295).

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This paper was also published in "Advances in Technology of Materials and Materials
Processing Journal, 9[1] (2007) 59-62".

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