論　文　の　要　旨

題　目　　Gas-phase preparation and characterization of photocatalytic Ag-TiO_2 nanoparticulate thin films
（Ag-TiO_2 光触媒ナノ粒子薄膜の気相作製と特性評価）

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Titanium dioxide (TiO_2), as a promising photocatalyst, has been the object of extensive investigation in recent years because of its excellent property in air and water purification. The loading of Ag nanoparticles on the surface of TiO_2 nanoparticles is considered as an effective method to enhance the photocatalytic property of TiO_2 by inhibiting the recombination of electron-hole pairs and broadening the light absorbance spectrum from UV region to visible region.

In this dissertation, photocatalytic Ag-TiO_2 nanoparticulate thin films were fabricated by a combined plasma-enhanced chemical vapor deposition (PECVD) process and physical vapor deposition (PVD) process, and the properties and performance as photocatalytic materials of the films were studied.

First, Ag-TiO_2 nanoparticulate thin films were fabricated via a combined PECVD and PVD processes. The Ag-TiO_2 films were fabricated on a silicon substrate by simultaneous deposition of TiO_2 and Ag nanoparticles synthesized by the PECVD and PVD systems. The effects of the additional Ag nanoparticles on the morphology, crystal structure and photocatalytic activity of the Ag-TiO_2 nanoparticulate thin films were studied. The morphology of the prepared TiO_2 nanoparticulate thin films and Ag-TiO_2 nanoparticulate thin films were observed by scanning electron microscope (SEM) and transmission electron microscope (TEM) techniques. The crystal structure of TiO_2 nanoparticulate thin films and Ag-TiO_2 nanoparticulate thin films were measured by the X-ray diffraction (XRD). And the elemental analysis was performed by energy dispersive X-ray spectroscopy (EDX) technique. The photocatalytic performance of the fabricated pristine TiO_2 film and Ag-TiO_2 nanoparticulate thin films was tested by the photodegradation of methylene blue (MB) solution under UV light irradiation. The results indicated that the loading of the Ag nanoparticles influenced the morphology and crystal structure of the TiO_2 nanoparticles in the Ag-TiO_2 nanoparticulate thin films. The addition of Ag leads to an increase in the particle size, a decrease of phase transformation temperature from anatase to rutile and a 35% increase in photocatalytic activity. The mixture of anatase and rutile phases and separation of electrons and holes influenced by Ag are considered as the determined reasons for the increase of photocatalytic activity despite the increasing particle size.

Next, the effects of annealing temperature on the Ag-TiO_2 nanoparticulate films based on the morphology, crystal structure, and photocatalytic activity were investigated. The thin films prepared by the one-step deposition processes were annealed at different annealing temperatures. The morphology of the prepared Ag-TiO_2 nanoparticulate thin films was observed by SEM techniques. The crystal structure of the prepared Ag-TiO_2 nanoparticulate thin films was measured by the XRD. The photocatalytic performance of the fabricated pristine TiO_2 film and Ag-TiO_2 nanoparticulate thin films was tested by the photodegradation of MB solution under UV light irradiation. The particle size, film thickness crystal structure, crystallite size, phase ratio, and photocatalytic performance were found to depend on the annealing temperature. Both of particle size and crystalline size of TiO_2 increased with increasing annealing temperature while the thickness showed a decrease. As annealing temperature increased, the crystal structure experienced changes from amorphous to pure anatase to mixture of anatase and rutile to pure rutile. The highest photocatalytic activity was obtained at annealing temperature of 700 °C, when the phase structure was a mixture of anatase phase (77%) and rutile phase (23%). The photocatalytic activity of Ag-TiO_2 nanoparticulate thin films with different crystal structures decreased in the order of the mixture of anatase and rutile > anatase > rutile.

The effects of Ag concentration in the Ag-TiO_2 films on the morphology, crystal structure, light absorption capability, and photocatalytic activity were investigated. The Ag concentration in the Ag-TiO_2 films was adjusted by tuning the furnace temperature in the PVD system to generate Ag nanoparticles in different amount. The morphological analysis was carried out by the SEM and TEM techniques. The crystal structure of prepared Ag-TiO_2 nanoparticulate thin films with different Ag content was measured by the XRD. The photocatalytic performance of the fabricated pristine TiO_2 film and Ag-TiO_2 nanoparticulate thin films was tested by the photodegradation of MB solution under UV light irradiation and photodegradation of rhodamine 6G (R6G) solution under visible light irradiation. The morphology, particle size, and photocatalytic activity of the films were evidently influenced by the Ag concentration. Non-agglomerated Ag nanoparticles (below 3 nm) were found to be dispersed well on the surface of TiO_2 nanoparticles in the films. The particle size of the TiO_2 nanoparticles in the films became larger as the Ag content increased, especially above a concentration of 2.2 wt%. The number density of Ag nanoparticles increased with increasing Ag content without obvious particle agglomeration. The light absorbance of the Ag-TiO_2 nanoparticulate thin films was higher than that of pristine TiO_2 film in the visible light region and the light absorbance of Ag-TiO_2 nanoparticulate thin films increased with increasing Ag concentration in the visible light region. The evaluation of the photocatalytic activity under UV light irradiation showed that the highest photocatalytic activity of the prepared Ag-TiO_2 films was 5.5 times higher than that of the pristine TiO_2 film when Ag concentration was 2.2 wt%. The photocatalytic activity of the prepared Ag-TiO_2 films under visible light irradiation showed 2.4 times than that of the pristine TiO_2 thin film when the Ag content was 0.24 wt%.

Finally, the summary of the findings was highlighted. Suggestions for further research on Ag-TiO_2 photocatalytic films were also proposed.