Photocatalytic Activity of Visible-light Sensitive $InTaO_4$-based Photocatalysts

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In 2001, nitrogen-doped TiO_2 in an anatase form was reported as a visible light sensitive photocatalyst$^{1, 2)}$. It attracted much attention and has been reported one after another recently$^{3-5)}$, independently. Except for it, some other nitrogen-doped metal oxides, such as nitrogen-doped $SrTiO_3$, Ta_2O_5 and ZnO, were found out to decompose organic compounds under visible light irradiation$^{6, 7)}$. The nitrogen doping into oxides seems to be a useful method to develop the oxides sensitive to visible light. As for water-splitting photocatalysts, a series of $InMO_4$ (M=Ta, Nb, V) was investigated$^{8-11)}$. Among them, NiO_y-loaded $In_{0.9}Ni_{0.1}TaO_4$ was reported to split water under visible light irradiation without sacrificial agents, recently$^{12)}$.

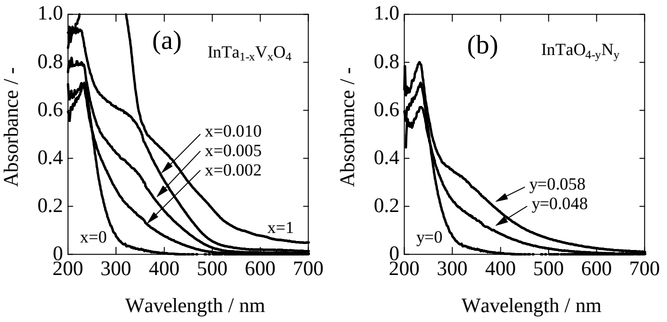
We confirmed that $InTaO_4$ could decompose organic compounds under UV light irradiation, so it was expected that nitrogen doping into $InTaO_4$ could develop it sensitive to Vis light, resulting in capable of decomposing organic compounds by irradiating with Vis light. Very recently, density of states (DOS) calculation of nitrogen-doped $InTaO_4$ was reported and new states originating from N 2p orbitals were appeared near the top of the valence band, resulting in the band gap narrowing by *ca.* 0.3 eV$^{13)}$. In addition, we found out that vanadium doping at a tantalum site in $InTaO_4$ also could absorb Vis light.

In the present study, we have fabricated nitrogen- and vanadium-doped $InTaO_4$, and evaluated their photocatalytic oxidation activity by the decomposition of gaseous 2-propanol (IPA) under the same absorbed photon number of Vis and UV lights. In addition, we discuss the features of electronic structures of $InTaO_4$, nitrogen-doped $InTaO_4$ and vanadium-doped $InTaO_4$, and the correlations of the electronic structures with UV and Vis light-induced photocatalytic activities.

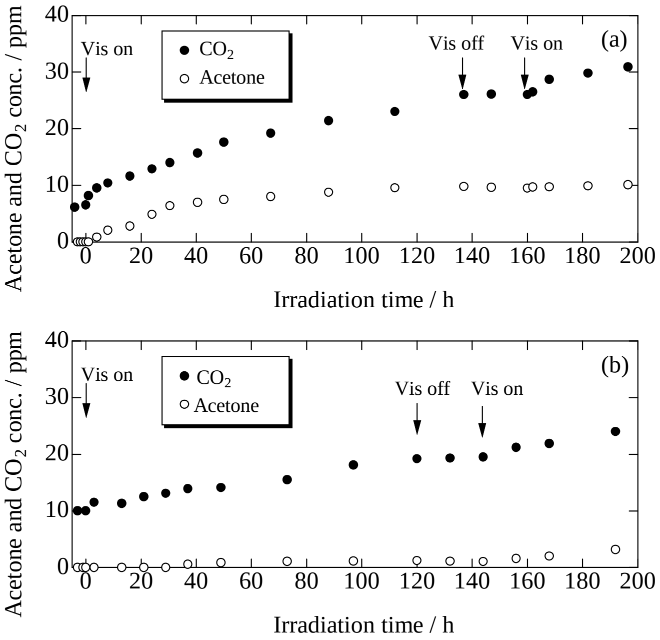
The polycrystalline powders of vanadium-doped $InTaO_4$ ($InTa_{1-x}V_xO_4$, x=0.002, 0.005, 0.01) were synthesized by a solid-state reaction method. The powders of nitrogen-doped $InTaO_4$ ($InTaO_{4-y}N_y$, y=0.048, 0.058) were synthesized by annealing $InTaO_4$ powders under NH_3.

All of the prepared powders were confirmed to have homogeneous $InTaO_4$ structure by XRD measurements. XPS measurements indicated that nitrogen was located at some of oxygen sites. Figures 1(a) and (b) show the absorbance spectra obtained by diffuse reflection of $InTa_{1-x}V_xO_4$ and $InTaO_{4-y}N_y$, respectively. Shifts of absorbance shoulders to visible light region were observed for both vanadium- and nitrogen-doped $InTaO_4$.

Figures 2(a) and (b) shows the gaseous IPA decomposition irradiated by visible light with the same absorbed photon number ($2.5*10^{14}$ $quanta.sec^{-1}.cm^{-2}$) in the presence of $InTa_{0.998}V_{0.002}O_4$ and $InTaO_{3.952}N_{0.048}$, respectively, as examples. Both acetone and CO_2 were produced when irradiating with visible light. It can be considered that these photocatalysts could completely decompose the injected IPA to the final product CO_2 under Vis light irradiation because the concentration of acetone, which is the intermediate of the IPA oxidative decomposition, reached to the maximum and then decreased with time (not shown in Figures 2(a) and (b)), indicating that the acetone was further decomposed to CO_2 by these photocatalysts. Similar experiments were conducted for the other samples irradiated by both UV and visible lights, separately, and also, similar results were obtained, although quantum yields depended on the dopants and concentrations. We calculated the DOSs of $InTa_{1-x}V_xO_4$ and $InTaO_{4-y}N_y$, and speculated that the isolated N2p above the valence band and V3d below the conduction band of $InTaO_4$ were responsible for visible light sensitivity. The details will be discussed in the conference.



Figures 1 Absorbance spectra obtained by diffuse reflection of (a) $InTa_{1-x}V_xO_4$ and (b) $InTaO_{4-y}N_y$.



Figures 2 Changes of acetone and CO_2 concentrations as a function of time irradiated by visible light in the presence of (a) $InTa_{0.998}V_{0.002}O_4$ and (b) $InTaO_{3.952}N_{0.048}$.

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