論文の内容の要旨

Development of Ta or Ge-based (oxy)nitride photocatalysts for overall water splitting

(水分解を目的としたTaまたはGeを含む（オキシ）ナイトライド光触媒の開発)

氏　名　李 允玘

In this thesis is given the research results about the development and improvement of Ta or Ge-based photocatalysts for overall water splitting.

In chapter 2 is depicted the preparation of sodium tantalate thin film, $Na_2Ta_2O_6$ or $NaTaO_3$ by hydrothermal-electrochemical synthesis at low temperature (≤473 K). The preparation of sodium tantalate thin films makes the investigation of their electrochemical properties and other variety of applications possible. In chapter 3 is given the hydrothermal synthesis of fine $NaTaO_3$ powder at low temperature (373–473 K) for overall water splitting. The $NaTaO_3$ powder thus prepared exhibits high photocatalytic activity for overall water splitting attributable to the high surface area, small particle size and high crystallinity. In chapter 4 is shown the effect of 10 MPa NH_3 treatment on the activity of visible light driven Ta_3N_5 photocatalyst. The rate for H_2 evolution under visible-light irradiation ($\lambda > 420$ nm) is improved five-fold by 10 MPa NH_3 treatment at 823 K. In chapter 5 is given the effect of high-pressure NH_3 treatment on the activity of Ge_3N_4 photocatalyst for overall water splitting. A β-Ge_3N_4 powder sample treated at 823 K for 5 h under ammonia at 20 MPa exhibited a photocatalytic activity 4 times higher than that of the as-synthesized powder, attributable to a decrease in the density of anion defects in the bulk and surface.

In chapter 6 and 7 are shown the development and improvement of $(Zn_{1+x}Ge)(N_2O_x)$ as a photocatalyst for overall water splitting under visible light. A solid solution of zinc oxide and germanium nitride with a wurtzite-type structure (space group: $P6_3mc$) thus prepared exhibits band gap of ca. 2.7–2.8 eV and corresponding activity at visible wavelengths. The decrease in band gap compared to the starting materials is attributed to larger valance band dispersion resulting from the energy difference between O2p and N2p orbitals, and from the p-d repulsion between Zn3d and N2p+O2p electrons in the upper valance band. A range of cocatalysts is also examined, and $Rh_{2-x}Cr_xO_3$ is identified as the most effective cocatalyst for $(Zn_{1+x}Ge)(N_2O_x)$. Modification of the optimized $(Zn_{1.44}Ge)(N_{2.08}O_{0.38})$ sample by loading with $Rh_{2-x}Cr_xO_3$(3.0 wt% Rh, 0.2 wt% Cr) results in an effective photocatalyst for overall water decomposition with quantum efficiency of ca. 0.20 % at 420 nm.