Quantitative evaluation of plasmon-induced ammonia photoelectrochemical synthesis

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Ammonia has received considerable attention as a potential energy carrier and as a fuel in addition to its use as a fertilizer because it is a non-carbon-based chemical that is easily condensed into a liquid. Localized surface plasmon resonance of metallic nanostructures has received much attention because of its light-harvesting and electric-field enhancement effects.[1] Previously, we achieved plasmon-induced ammonia synthesis via photoelectrochemical reduction of nitrogen using a strontium titanate photoelectrode loaded with gold nanoparticles (Au-NPs) and co-catalyst via plasmon-induced charge separation.[2, 3] However, the electric properties and the reaction mechanism are still unclear because the previous study only analyzed the chemical products. In this study, we report on a quantitative evaluation of ammonia photoelectrochemical synthesis on a two-electrodes system with the Au-NPs loaded strontium titanate as a plasmon photoanode and a zirconium coil as a cathode.
The photoelectrochemical nitrogen reduction device using Au-NPs/niobium-doped strontium titanate (Nb-$SrTiO_3$) plasmon photoanode was fabricated as follows. Au-NPs were fabricated on a 0.05wt% Nb-$SrTiO_3$ single crystalline substrate using a sputtering and annealing method. The nitrogen reduction device comprised reaction cells with two reaction chambers separated by Nafion 117 membrane. The Au-NPs/Nb-$SrTiO_3$ photoanode was installed in the one chamber, and a zirconium coil as a co-catalytic cathode was put in the another chamber. The both chambers were filled with an H_2SO_4 aqueous solution. The Au-NPs/Nb-$SrTiO_3$ photoanode was irradiated by a xenon lamp using the visible light, and the cathodic chamber was bubbled with nitrogen gas during the reaction. The electric properties and chemical products were monitored simultaneously.
We attempted the quantitative evaluation of ammonia photoelectrochemical synthesis, especially bias effect, stoichiometry and intermediate were investigated in detail. Also, a reaction mechanism of plasmon-induced ammonia synthesis is proposed.

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