Hydrogen production via photocatalytic reforming of aquieous methanol solution over copper and nickel loaded TIO2

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ABSTRACT
The purpose of this study is to develop a low cost, easy prepared, and environmentally friendly photocatalyst to produce hydrogen from aqueous methanol solution by combining catalytic reforming (metal based catalyst) and photocatalytic process (semiconductor based photocatalyst), at ambient condition under photon exposure. The effect of impregnated Cu and Ni (which are proven catalysts for thermal reforming) to TiO2 were investigated as well as the role/significance and behavior of methanol and water in photo-reforming process. As prepared Cu/TiO2 and Ni/TiO2 photocatalyst were characterized by ICP-AES, XRD, SEM, TEM, and UV-Vis DRS for better understanding of the photocatalytic reforming behavior. The optimum loadings of Cu and Ni into TiO2 surface were found to be 3% and 1% respectively. H2 generated from photoreforming of aqueous methanol solution (80% methanol v/v) over 3% Cu/TiO2 UV illumination was 4464.3 μmol.gcat 1.h-1, 5.5 times higher than unloaded TiO2 (803 μmol.gcat-1.h-1) while H2 yield over Ni/TiO2 wasfound to be 5200 μmol.gcat-1.h-1, 6.5 times higher compared to unloaded TiO2. In term of stability, Ni/TiO2 also shows superior performance compared to Cu/TiO2 and unloaded TiO2. Ni/TiO2 can still obtain final rate of 66% of its initial rate while only 42.4% was obtained for the case of Cu/TiO2, yet it is still slightly better than unloaded TiO2 (40.8%). Ni/TiO2 superiority in photocatalytic performance over Cu/TiO2 may be attributed to its higher work function which leads to higher electron trapping ability, better electron transfer from conduction band of TiO2 to metal site, and lower hydrogen overpotential. In order to investigate the role and significance of methanol and water on aqueous methanol photocatalytic reforming system, the methanol-water composition was varied during this particular study. The rates of hydrogen evolution displayed bell-shaped curves as a function of methanol volume fraction in the solution. The optimum hydrogen evolution rate was achieved in methanol volumetric ratio of 60-80%, in agreement with stoichiometric value of methanol:water mixture (1:1 molar ratio or 0.69:0.31 volumetric ratio). Both methanol and water show typical Freundlich adsorption behaviors. For solution containing 0-70% methanol, relationship between the hydrogen generation rate (v) and methanol content ([M]) is represented as v = 637.15[M]0.439. For solution containing 0-30% water, relationship between the hydrogen generation rate (v) and water content ([W]) is represented as v = 2594.1[W]0.161. This indicates that adsorption of water and methanol on the photocatalyst was a crucial part of the reaction mechanism.