

Hydrogen production via photocatalytic reforming of aqueous methanol solution over copper and nickel loaded TiO₂

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Deskripsi Lengkap: <https://lib.ui.ac.id/detail?id=20464566&lokasi=lokal>

Abstrak

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 mol.gcat⁻¹.h⁻¹, 5.5 times higher than unloaded TiO₂ (803 mol.gcat⁻¹.h⁻¹) while H₂ yield over Ni/TiO₂ was found to be 5200 mol.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.