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APXPS Study of Photocatalytic Driven Atomic Structure Transformation of Core-Shell Ni@NiCO3/NiO Photocatalyst

Content

The atomic-level understanding of the active sites and transformation mechanism under realistic conditions is a prerequisite for the rational design of a high-performance photocatalyst. A detailed study of photocatalytic driven atomic structure and electronic transformation of core@shell Ni/NiCO3/NiO photocatalysts was carried out using ambient pressure X-ray photoelectron spectroscopy under 1 mbar H2O pressure at SPECIES beamline. Ni@NiCO3/NiO, with metallic Ni0 core and mixed NiCO3 and NiO shell, is a visible light active photocatalyst used for water splitting to hydrogen.1 When subjected to a combination of water vapor and visible light, many interesting reversible structural and electronic transformations are observed. However, when exposed to light under UHV conditions, no changes could be observed in the electronic structure of the catalyst. The APXPS data shows, that in 1 mbar water vapor pressure and under visible light illumination, Ni0 is oxidized to NiOOH, (giving direct evidence that it is the metallic Ni0 that absorbs the visible light and frees up an electron. These electrons are further utilized for the reduction of hydrogen ions in the hydrogen evolution reaction (HER). The in-situ experiments were repeated several times, and the Ni0 oxidation and reduction occur reversibly under the light on/off conditions at 1 mbar. Furthermore, as a resolute of our study, we outline the role of the carbonates and their participation in the reaction mechanism, something which is rarely investigated in terms of their role in photocatalysis.
References.
[1] Talebi, P., Singh, H., Rani, E., Huttula, M. & Cao, W. Surface plasmon-driven photocatalytic activity of Ni@NiO/Nico 3 core-shell nanostructures. RSC Adv. 11, 2733–2743 (2021).

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