FeNi/-Al2O3 Egg-shell Catalyst for H2 Generation by Ammonia Decomposition

Silva, Hugo José Lopes; Nielsen, Morten Godtfred; Fiordaliso, Elisabetta Maria; Damsgaard, Christian Danvad; Gundlach, Carsten; Kasama, Takeshi; Chorkendorff, Ib; Chakraborty, Debasish

Publication date: 2016

Document Version
Publisher's PDF, also known as Version of record

Link back to DTU Orbit

Citation (APA):

General rights
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain.
- You may freely distribute the URL identifying the publication in the public portal.

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.
FeNi/γ-Al₂O₃ Egg-Shell Catalyst for H₂ Generation by Ammonia Decomposition

Hugo Silva¹, Morten G. Nielsen¹, Elisabetta M. Fiordaliso², Christian D. Damsgaard¹²*, Carsten Gundlach³, Takeshi Kasama², Ib Chorkendorff¹, Debasish Chakraborty¹

¹ – CINF, Department of Physics, DTU, Building 307, 2800 kgs, Lyngby, Denmark
² – Center for Electron Nanoscopy, DTU, Building 307, 2800 kgs, Lyngby, Denmark
³ – NEMAP, Department of Physics, DTU, Building 307, 2800 kgs, Lyngby, Denmark
*

The FeNi alloyed nanoparticles are a promising alternative to expensive ruthenium-based catalysts for a real-scale application of hydrogen generation by ammonia decomposition[1-2]. In practical applications, millimeter-sized extrudates supports are used as catalysts, where the spatial distribution of the active phase should match with the type of reaction [3]. In this work, a novel synthesis route was developed for the preparation of a FeNi/Al₂O₃ egg-shell catalyst [4]. Egg-shell is a preferred profile considering the highly endothermic nature of ammonia decomposition reaction. The high viscosity of glycerol, used as a solvent, prevents the fast migration of the FeNi active phase solution towards the inner-core of Al₂O₃, giving control over the large capillary pressures during impregnation. The distribution profiles were analyzed at macroscopic scale through scanning electron microscopy mapping (SEM-EDX) and optical microscopy (Figure 1). A three-dimensional (3D) reconstruction of the spherical-shaped Al₂O₃ was achieved using x-ray micro tomography and the FeNi egg-shell spatial distribution was inspected throughout the entire volume of the support body. Transmission electron microscopy (TEM) and scanning TEM (STEM) analysis of ultrathin lamellas (< 20 nm) carved from the outer-shell region established the presence of FeNi alloy nanoparticles with a size of approximately 5 nm (Figure 2). The egg-shell catalyst showed significant higher activity in ammonia decomposition by converting 3 times more ammonia to equilibrium conversion than either egg-white or catalyst with uniform distribution.

Figure 1. (a) SEM-EDX elemental maps of Fe for the FeNi/Al₂O₃ catalysts prepared from an aqueous solution of the active phase and (b) from a glycerol solution. Optical microscopy images of the aforementioned samples, (c) impregnation with an aqueous solution, and (d) with glycerol.

Figure 2. (a) Optical microscope image of a TEM specimen prepared from an egg-shell catalyst placed on a Cu hexagonal grid. HAADF-STEM images obtained from the Fe–Ni rich outer-shell (b) and the core (c) of the γ-Al₂O₃ sphere (d) High-magnification STEM image of the egg-shell catalyst acquired from the Fe–Ni rich outer-shell.