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Published in:
Book of Abstracts. DTU's Sustain Conference 2015

Publication date:
2015

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

Link back to DTU Orbit

Citation (APA):
Synthesis and Characterization of FeNi/γ-Al2O3 Egg-Shell Catalyst for H2 Generation by Ammonia Decomposition

Hugo Silva, Morten G. Nielsen, Elisabella M. Fiordaliso, Christian D. Damsgaard, Carsten Gundlach, Takeshi Kasama, Ib Chorkendorff, Debasish Chakraborty*

1: DTU Physics; 2: DTU Cen

*Corresponding author email: debc@fysik.dtu.dk

The FeNi alloyed nanoparticles are a promising alternative to expensive ruthenium-based catalysts for a real-scale application of hydrogen generation by ammonia decomposition. 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. In this work, a novel synthesis route was developed for the preparation of a FeNi/Al2O3 egg-shell catalyst. 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. 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. The egg shale 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.