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Synthesis and Characterization of FeNi/ γ -Al₂O₃ Egg-Shell Catalyst for H₂ Generation by Ammonia Decomposition

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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 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.