

TOPIC 3

Biogas-reforming catalysts Deactivation mechanisms of Ni-Fe and Ru-exsolution during prolonged exposure to H₂S and DMS contaminants

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Methane reforming is a recommended step for using biogas in solid oxide fuel cells. The main reactions are dry methane reforming (methane and carbon dioxide are converted to hydrogen and carbon monoxide in equimolar fraction) and steam methane reforming (methane and water reacts to form three dihydrogen molecules and one carbon monoxide molecule). In addition, a number of reactions occur in parallel and shift the reformed gas composition. Among these, the reversible water gas shift reaction that can lead to an enrichment in hydrogen through the reaction between carbon monoxide and water.

Methane is typically reformed on transition metal catalysts loaded on ceramic supports. Ruthenium showed the best properties for the activation of C-O bond, and demonstrated intrinsic stability and coking resistance. However, its scarcity, price, and difficulty to uniform dispersion on a support are important drawbacks for its use at large scale. Hence nickel-based catalysts are often preferred as they are more abundant and affordable, even though they are more sensitive to deactivation through coking and particle growth.

In this work, a ruthenium-exsolution type catalyst and a nickel-iron catalyst were studied for the dry and steam reforming of methane. The effect of the gas velocity on methane conversion and the reforming mechanism led to the selection of optimal gas velocities in both conditions. Afterwards, hydrogen sulfide and dimethyl sulfide traces (0.5-20 ppm) were added to the gas feed and the deactivation mechanisms studied. By studying the reaction mechanism, it is demonstrated that these contaminants have specific effects on the different active sites. Finally, after full deactivation of the catalysts, temperature programmed oxidation-reduction cycles were studied as regenerative procedures.