Pure hydrogen production in a membrane reformer: Demonstration, macro-scale and atomic scale modeling

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Abstract

We demonstrate and discuss various designs for scaled down membrane steam reformer for generating pure hydrogen onboard or in a hydrogen fuel station, motivated by the expectation of using hydrogen as an energy carrier. Pure H2 separation is achieved by Pd or Pd/Ag membranes and the feeds tested include methane, ethanol and glycerol.

A novel concept for hydrogen generation by auto-thermal methane steam reforming (MSR) was demonstrated. The system incorporates two reactors exchanging and recuperating heat: an endothermic membrane reformer (catalyzed by Ni/Al_2O_3), and an exothermic oxidation reactor fed by the reforming effluents. Experiments and analysis of MSR revealed that high thermal efficiencies require temperatures higher than 550°C and that the oxidation feed should be axially distributed for improving efficiencies and mitigating hot spots. Comparable performance was achieved with other fuels. The mathematical model, considering membrane permeance inhibition, was validated without adjustable parameters and used for optimization. This design serves as proof of concept for on-board pure H_2 generators, with flexible fuel source type, feeding an adjacent fuel cell.

In a second concept, solar energy circulated by means of molten salts is used to heat the membrane reformer in a hydrogen fuel station. Laboratory scaled membrane reformer, fed with methane and steam, packed with a foam catalyst and heated externally, showed over 90% conversion and over 80% hydrogen-recovery at 525°C. Modeling this reactor suggests about 80% reduction in permeance, compared to a value measured in pure hydrogen. Ethanol steam-reforming yields mainly methane and CO_2, as well as hydrogen, as products with a high yield of H_2 under conditions of high H_2 separation.

A 2-D mathematical model that incorporates the reaction with CFD showed that concentration polarization cannot account for the observed strong permeance inhibition. To understand this phenomenon we resort to DFT-calculated adsorption energies to estimate the inhibition due to surface adsorption of possible co-adsorbates like methane, CO and water. While CO adsorption is strong, CO concentration is small, and this effect is too small to account for observations, suggesting that surface reaction on the Pd membrane should be considered to estimate coverages by C , O and other intermediates.