



INDIAN INSTITUTE OF TECHNOLOGY GUWAHATI  
SHORT ABSTRACT OF THESIS

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**SHORT ABSTRACT**

Commercial energy demand is largely met by fossil fuels globally. Burning of fossil fuel causes huge carbon emissions, which have created environmental menace due to ever increasing human population. Therefore, alternative energy production using renewable resources are considered a key player in minimizing the dependence on fossil fuels. Hydrogen is considered as a feasible alternative to fossil fuel. "Hydrogen" contains the highest energy to weight ratio with nearly three times the energy content of gasoline and diesel. Hydrogen can be used as a fuel in polymer electrolyte membrane (PEM) fuel cells that have potential to substitute internal combustion engines to provide on-board power for portable power generators and vehicles. However, storage and distribution of hydrogen is a major problem for fuel cell based stationary or mobile power generators. 'In-situ' generation and separation of hydrogen through 'membrane reformers' using steam reforming of alcohols can be a possible solution for this problem.

Membrane reformers couple reaction and separation in a single unit. However, achieving high hydrogen purity and high hydrogen flux through membrane reformer is a major challenge. The current work provides a comprehensive study on membrane reformer.

In the newly designed membrane reformer, hydrogen separation was carried out by using palladium based membranes to achieve high purity. Pd-based membranes supported on porous SS (PSS) and ceramic tubes were synthesized using electroless deposition until a dense (non-porous) surface morphology was achieved. Membrane deposition was carried out for three combinations a) pure Pd, b) 90%Pd-10%Ag and c) 90%Pd-8%Ag-2%Au. Surface characterizations were performed with FESEM-EDX and AFM. Room temperature testing of the synthesized membrane was performed using bubble point methanol with argon as the purging gas inside the membrane dipped in water at different pressures. Further, the testing of prepared membranes were conducted with simulated

compositions of reformat to determine membrane perm-selectivity as a separator for PSS as well as ceramic supported membranes to achieve high hydrogen purity.

Subsequently, membrane separator design was evaluated with the implementation of multi-pass inside it to achieve high hydrogen flux. Gas permeation studies were carried out using 50H<sub>2</sub>:50N<sub>2</sub> (v/v) feed composition for single and multiple membranes placed at different locations inside the multi-pass membrane separator. The best arrangement was then tested using a mixture composition such as 50H<sub>2</sub>:30N<sub>2</sub>:18CO<sub>2</sub>:2CO (v/v) that simulates a synthetic reformat mixture from methanol steam reforming.

Hydrogen generation was targeted with methanol steam reforming in a fixed bed reactor with different catalysts for variable temperature (150 to 400°C), feed rate and methanol to steam compositions. Reaction testing was performed with mono-metallic (Cu, Ni, Pd, Ru, Fe) and bimetallic (Cu-Fe, Ni-Fe, Ru-Fe, Cu-Ni and Cu-Ru) catalysts supported on alumina-zinc-zirconia composite. In bimetallic catalysts, Fe as a promoter was investigated to enhance hydrogen production rate as well as to determine its feasibility in inhibiting CO generation even at high temperatures. Reaction performance was supported with material characterization tools such as XRD, TPR, TEM, FT-IR, TGA, DRIFT and Chemisorption. For the optimal catalyst, reaction mechanism was proposed and its long-term stability testing for 100 hours was carried out in order to confirm its viability for membrane reformer integration.

Finally, synthesized catalyst and prepared membrane are integrated in a single assemble, membrane reformer, and tested. The performance of in-house built membrane reformer was compared with commercially available membrane reformer unit "ME-100®" from REB Research and Consultancy, USA. The commercial unit comprise of an in-built reactor with 100 µm thick self-supported dense Pd-Ag membrane and a proprietary WGS catalyst. Experiments were performed to optimize the following parameters: a) feed composition (varied in the molar ratio of 1:1 to 1:18 methanol: water) b) temperature (between 623–723K) and c) pressure (between 3 and 5 bar). With the optimized operating conditions, the in-house developed membrane reformer was compared. Further, multiple membrane integration studies were also carried out to determine the maximum hydrogen flow rate in permeate of membrane reformer without compromising the purity of hydrogen.