Autothermal Reforming of n-Dodecane for Fuel-Cell Applications – Nickel Based Xerogel Catalysts for Activity, Stability and Coking Studies

Venkata Phanikrishna Sharma Mangalampalli, Jale Akyurtlu*, Ates Akyurtlu

Chemical engineering, Hampton University, Hampton, Virginia -23668 (USA)

Extended Abstract

Fuel cells are one of the alternative solutions to evade the problems of air pollution and global warming due to emissions from transportation. Fuel cells work on clean hydrogen and air to produce electricity with lower or no emissions. The combination of an on-board reformer and a SOFC would enable commercial fuels such as jet fuel and diesel to be used as a hydrogen source because of high energy density of these fuels. The US military is the single largest petroleum consumer in the world and jet fuel represents 64% (volume basis) of all petroleum products consumed in the USA [1]. Hence in the present study we have chosen autothermal reforming (ATR) of n-dodecane for the production of clean hydrogen, which is a surrogate for jet fuel. ATR is chosen as the best method in the present study because it has advantages over present technologies i.e., steam reforming (SR) and partial oxidation (POx), like compact and quick starting reactors, and better sulfur tolerance and less coking. ATR is a combination of endothermic SR and the exothermic POx process; hence it is self-sustained and evades catalyst deactivation for longer operation. This process requires less energy and helps reduce the amounts of methane and coke produced while providing a high H₂ yield and a low CO yield under optimal operating conditions. Additionally, the water gas shift reaction, which proceeds simultaneously, reduces the CO content of the hydrogen-rich gas.

The present challenges in the development of ATR catalysts for higher hydrocarbons are catalyst deactivation by coking and sulfur intolerance. Even though the noble metal (Rh, Ru, Pd)-promoted catalysts are showing higher activity and stability for this reaction, catalyst cost is one of the main obstacles. Hence in our study we have tried to minimize the use of noble metals without compromising in hydrogen production. In the present experimental study, we have developed Ni-based catalysts which are resistant to carbon formation by incorporating Ce and Zr species, using a typical sol-gel method. Preparation of metal oxides by the sol-gel method results in the retention of hydroxyl-rich surfaces, which exhibit unique textural and chemical properties compared with those prepared by other conventional methods, along with high surface areas.

In the present study, with an objective to conserve noble metal usage, and, also, to improve the ATR performance, we have studied Ni-based catalysts with an alumina support along with transition metals and Ru promoter. Sol-gel method is used to prepare the catalysts. Catalysts were characterized by X-ray diffraction (XRD), temperature programmed reduction (TPR), pore size distribution, hydrogen chemisorption, XPS and BET surface area measurements. The catalysts are evaluated for ATR of n-dodecane in a microreactor setup at different reaction temperatures ranging from 650-850°C, space velocity (132000 - 660000h⁻¹), oxygen/carbon ratio (0 - 1) and steam/carbon ratio (0 - 3.14). NiCeZrAl₂O₃ catalyst is optimized for high catalytic activity, low carbon deposition, good resistance to sintering and prolonged stability compared to the equal mass of other catalysts prepared in the project. Used catalysts are evaluated for carbon formation by TEM, SEM-EDAX and TPO studies. The results from these experiments will be discussed during the presentation in terms of the effect of operation variables on the hydrogen yield and product gas composition.

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References