Steam-Methane Reforming on Joule Heated Ni-Coated Metal Wires

Elmer B. Ledesma[1], Meghana Idamakanti[1], Praveen Bollini[1], Ram R. Ratnakar[2], Michael P. Harold[1]*

[1] *William A. Brookshire Department of Chemical and Biomolecular Engineering, University of Houston, Houston, TX 77204, United States of America;* [2] *Shell International Exploration and Production Inc., Houston, TX 77082, United States of America*

**Corresponding author: mpharold@central.uh.edu*

Highlights

- Uniform coating of Ni/ZrO_2 or Ni/Al_2O_3 on FeCrAl wires.
- CH_4 conversion is higher on Ni/ZrO_2 wire compared to uncoated or ZrO_2-coated wire.
- Discontinuous jump in CH_4 conversion at critical applied power.
- CH_4 conversion is higher on Joule heated wire compared to conventionally-heated wire.

1. Introduction

The global demand for hydrogen in 2021 reached all-time high at 94 million tons and is predicted to approach 130 million tons by 2030.[1] Most of the hydrogen is produced with unabated steam-methane reforming (SMR) due to its favorable cost compared to other commercial hydrogen production processes. However, unabated SMR is a significant source of greenhouse gas (GHG) emissions, emitting as much as 11 kg of CO_2 per kg H_2 produced.[2] The GHG emissions are primarily a result of the use of natural gas combustion to supply the heat needed in SMR furnaces to drive the chemistry to high conversion. To meet emerging mandates for GHG emission reduction, rapid development and deployment is needed of large-scale decarbonization measures in the hydrogen generation sector.

One potential low-carbon solution for SMR is electrifying the heating process using renewable power from sources such as wind and solar that do not emit GHG. Interesting approaches exist that utilize electricity to energize chemical reactors.[3-5] Active research efforts are reported for SMR, although there are still no current industrial-scale processes. In this study we investigate the application of Joule heating of catalyst-coated, high-resistance wires as a potential method to electrify SMR.

2. Methods

The catalysts used in this study are Ni/ZrO_2 and Ni/Al_2O_3. A wet impregnation procedure is used to synthesize the catalysts with two different Ni loadings (10 and 20 wt%). Slurries prepared with catalyst powders are uniformly deposited as a thin film onto coiled and straight FeCrAl wires. SEM-EDAX analysis reveals that the catalyst coating is uniformly distributed and the Ni particles are dispersed without significant agglomeration. Bench-scale Joule heating experiments are performed by applying direct electric current through the wire. A gas mixture containing 5.4 vol% CH_4 with excess steam (H_2O:CH_4 = 3:1 – 4:1) molar ratio is fed to a single wire reactor The gas is kept at 190°C and atmospheric pressure and flows into a stainless-steel tube housing the wire. Experiments are conducted at power settings in the range 50-90 W. After condensing unreacted steam from the reactor effluent, FTIR analysis is used to measure the effluent CO, CO_2, and CH_4 concentrations.

3. Results and Discussion

Figure 1(a) presents plots of CH_4 conversion as function of the applied power for both an uncoated and a 10 wt% Ni/ZrO_2-coated wire coil. For both wires, the feed flow rate is fixed at 130 sccm. Under otherwise identical conditions, the conversion is significantly lower for the uncoated wire as compared to coated wire. As shown in Figure 1(a), negligible conversion of methane is achieved using the uncoated wire for the range of applied power. The coated wire exhibits a very steep increase in methane conversion for power greater than 65 W. The ratio of CO to CO_2 increased from 0.77 at 58 W to 1.7 at 89 W. The results clearly demonstrate that under the employed experimental conditions, Joule heating

a Ni/ZrO_2-coated wire is significantly more effective in converting CH_4 than an uncoated wire for applied power settings greater than 65 W. Comparable results were also obtained for a 20 wt% Ni/Al_2O_3-coated coil. The conversion profile of the ZrO_2-coated wire shown in Figure 1(b) is similar to the uncoated wire. The CH_4 conversion is thus much lower for the ZrO_2-coated wire compared to the Ni/ZrO_2-coated wire. This clearly shows that the dispersed Ni is the active catalyst.

To identify and elucidate the impact of the Joule heating on SMR performance, we evaluated the reaction on a 20 wt% Ni/ZrO_2-coated wire under conventional heating. Figure 1(c) compares the fractional methane conversion versus temperature and power for the coated wire by conventional and Joule heating methods. For a range of power, the methane conversion obtained with the Joule-heated Ni/ZrO_2-coated wire is higher than the conventionally-heated Ni/ZrO_2-coated wire. The results in Figure 1(c) clearly demonstrates a marked difference in conversion profiles obtained from Joule heating and from conventional heating. Stability experiments reveal that the catalyst-coated wires exhibited sustained activity, clearly demonstrating sustained SMR for several hours without catalyst deactivation. H_2 was computed to be 2.9 mol generated per mol of CH_4 fed. This corresponds to an energy usage of 55 kWh of energy used for every Nm^3 H_2 generated and a production cost of 56 USD per kg H_2 generated. Several mechanisms have been identified for the interesting differences between Joule heating and conventional heating.

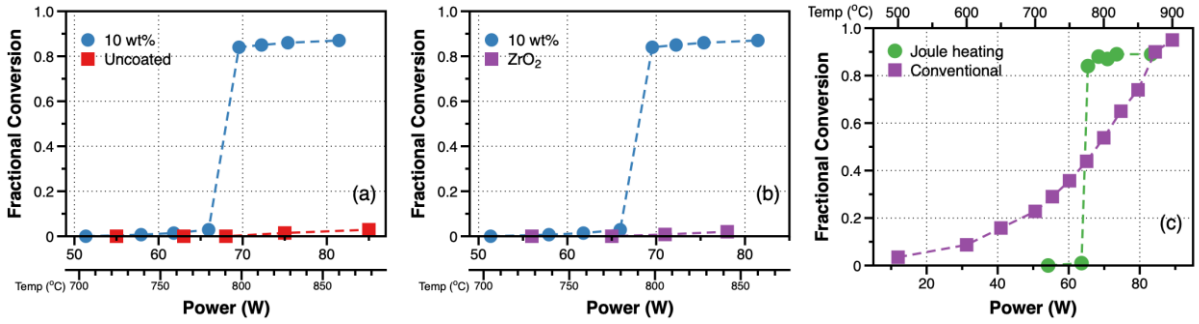

Figure 1. Comparison of fractional conversion of CH_4 obtained from the Ni/ZrO_2-coated wire coil with 10 wt% Ni loading to (a) an uncoated wire and (b) a ZrO_2-coated wire as functions of applied power. The secondary x-axis displays the corresponding estimated wire temperature. (c) Comparison between conversion profiles for the Ni/ZrO_2-coated coil with 20 wt% Ni loading obtained from Joule heating and conventional heating.

4. Conclusions

The results obtained to date provide proof-of-concept for using Joule heating of a catalyst supported on a conductive metal substrate. The catalyst-coated FeCrAl wire demonstrated significantly higher CH_4 conversion compared to an uncoated wire. Stability experiments revealed that the coated wire exhibited sustained activity, clearly demonstrating sustained steam reforming for several hours. In summary, the results provide, for the first time, experimental evidence for the potential for H_2 production using electrically heated catalytic wire reactor.

References

[1] Internal Energy Agency, Global Hydrogen Review 2022, OECD Publishing, Paris, 2022
[2] P. Sun, B. Young, A. Elgowainy, Z. Lu, M. Wang, B. Morelli, T. Hawkins, Environ. Sci. Technol. 53 (2019) 7103-7113
[3] M.E.H. Tijani, H. Zondag, Y. Van Delft, ACS Sustainable Chem. Eng. 10 (2022) 16070-16089
[4] A.I. Stankiewicz, H. Nigar, React. Chem. Eng. 5 (2020) 1005-1016
[5] M.A. Ambrosetti, Chem. Eng. Process.: Process Intensif. 182 (2022) 109187

Keywords
Electrification; decarbonization; reforming; hydrogen.