Exploration of 2-Stage CO_2 Reduction System

Author: Frederick Wewers, Clemson University PI: James M. Mansell, ES62

Introduction

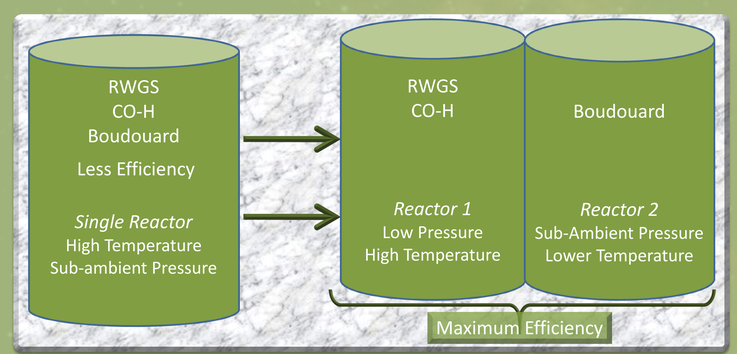
Open-loop atmosphere revitalization (AR) systems are unfavorable for long-term missions beyond low Earth orbit (LEO) due to incomplete mass recovery. These AR systems vent carbon dioxide (CO_2) overboard and require resupply of oxygen and carbon. To improve mass recovery, CO_2 reduction systems are currently being developed by NASA. The state-of-the-art Sabatier system reacts CO_2 and H_2 over a transition metal catalyst to produce methane (CH_4) and water. In contrast, the Bosch process uses a metal catalyst to react CO_2 and H_2 to produce solid elemental carbon and water.

$CO_2 + H_2 \rightarrow H_2O + CO$ (Reverse Water-Gas Shift reaction - RWGS)

$CO + H_2 \rightarrow H_2O + C(s)$ (Carbon Monoxide Hydrogenation reaction – CO-H)

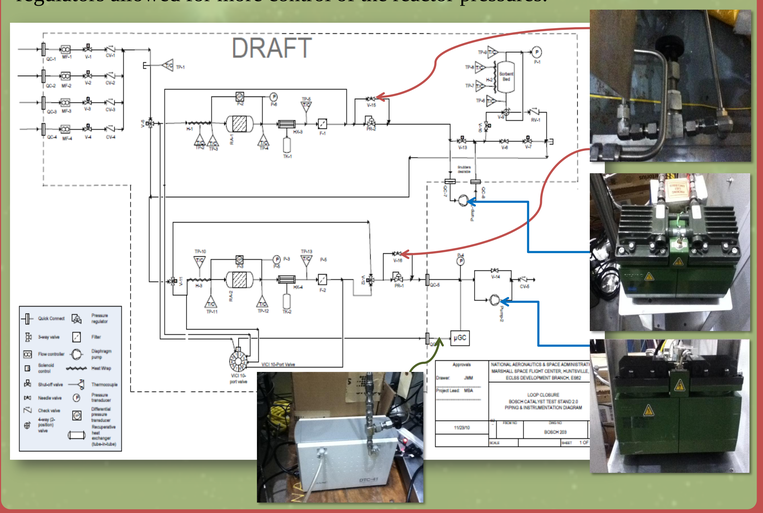
$2CO \rightarrow CO_2 + C(s)$ (Boudouard Reaction)

Taking carbon to its elemental state allows for complete recycling of both the hydrogen and oxygen in the form of water, unlike the Sabatier reaction which would have to waste hydrogen in the vented methane. However, current Bosch systems are burdened by high power consumption due to large heating and recycling requirements, and by considerable catalyst resupply needs. To increase the efficiency of the process, NASA MSFC is investigating the possibility of a two-stage reactor system to separate the process steps. This separation will allow optimization of each component reaction in the Bosch process, resulting in maximum efficiency.



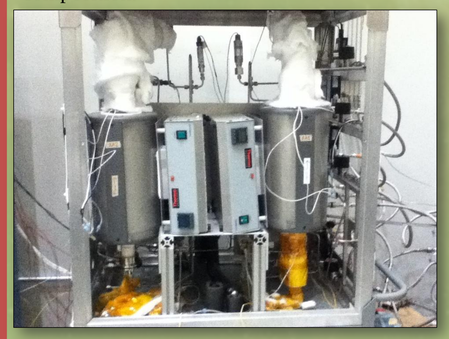
Modification

In order to investigate two stage reactions, the Series Bosch Catalyst Test Stand (Series B-CaTS) required modifications of its original design. Three new pumps were installed along with the replacement of two back-pressure regulators. The new pumps allowed for sub-ambient pressures to be reached within the system, and allowed for the micro-Gas Chromatograph (µGC) to analyze the system gases. The metering valves that were installed in place of the back-pressure regulators allowed for more control of the reactor pressures.



Methodology

The Reverse Water-Gas Shift Reaction was evaluated under various flow rates and pressures to analyze reaction rate. Seven varying flow rates of CO_2, H_2 and N_2 were flown through 80.15 g of Nickel shavings at a ratio of 5:4:1 respectively. Each of the seven flow rates were conducted at 4 different pressures using the installed pumps. A micro chromatograph (µGC) was used to analyze the system composition.



Fully Assembled Series B-CaTS Furnace and Reactors

- Total flow rates include: 100 - 700 SmLPM
- Pressures include: 3, 7.5, 10, and 15 psia
- Reactor Temperature: 650 °C

In order to isolate the RWGS reaction, only Reactor 1 was used for testing, containing the Nickel shavings. To hold the shavings in place, insulation was rolled and placed inside the reactor, shown below.

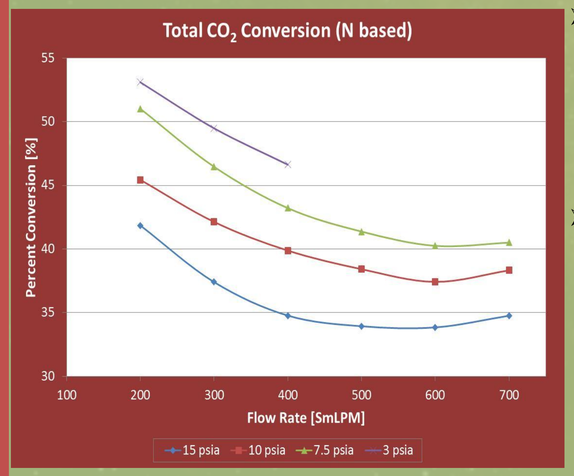
Packed Reactor with Nickel Shaving and Insulation

Single Reactor used for Testing

Results and Discussion

RWGS Testing

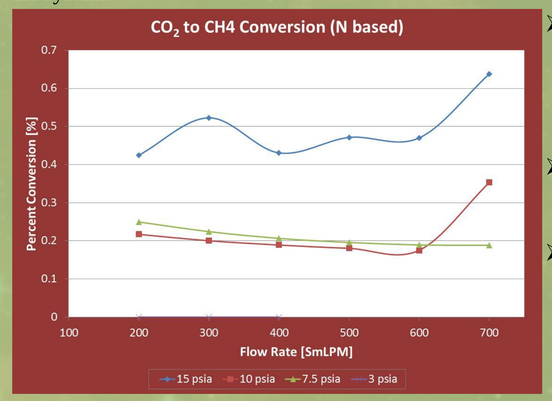
Seven different flow rates were directed through a single reactor containing Nickel Shavings at four pressures and 650 °C. A µGC was able to take samples from the pre-stream of Reactor 1 and the exit stream. From the µGC data, the conversion of the CO_2 to CO and CH_4 were calculated.



- This graph displays Total CO_2 Conversion based on the Nitrogen balance of the reactor.
- As predicted in theoretical equilibrium calculations, lower pressures resulted in higher reaction coefficients.

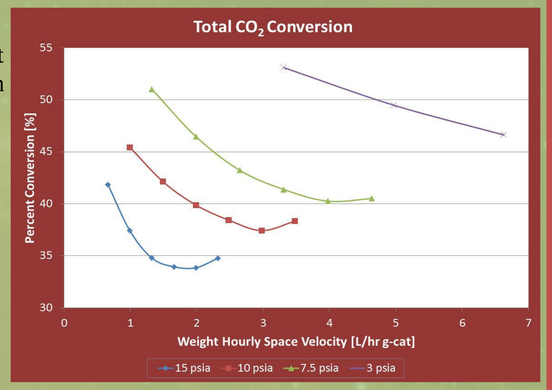
Sub-Ambient Pressures

Because of these higher reaction rates at lower pressures, it is beneficial to operate the reactions at sub-ambient pressures. Along with higher reaction rates and selectivity, a safety feature is present, as a leak in the system would result in air leaking in. Positive pressures would result in poisonous gases leaking out of the system.



- RWGS reaction selectivity increases as pressure decreases.
- Corresponds with equilibrium calculations.
- Shows the desire to operate tests at sub-ambient pressures.

- Allows for the amount of catalyst needed for a given situation to be calculated.
- Displays clearer how as pressure decreases, larger volumes of CO_2 can be converted per gram of catalyst.



Conclusions

The Reverse Water-Gas Shift reaction explored with Nickel Shavings and various flow rates proved to be more selective as the system pressure decreased. In addition, the reaction rate increased with decreasing pressure. This corresponds to the desire to operate the test stand at sub-ambient pressures, so as to increase efficiency, and build in a safety net incase of a leak.

Future Work

- Design and modify the pumping system for Series B-CaTS test stand to reach maximum efficiency.
- Replace metering valves with new Back-Pressure Regulators for easier manipulation of pressures.
- Begin full testing of the 2-stage process with Nickel Shavings and Steel Wool catalysts.
- Explore ideal temperature and pressure environment for Boudouard Reactor 2

References

[1] Abney, Morgan B., and Mansell, J. Matthew, "Evaluation of Bosch-Based Systems Using Non-Traditional Catalysts at Reduced Temperatures," NASA MSFC ES62, 2011.
[2] Holmes, R. F., Keller, E. E., and King, C. D., "A Carbon Dioxide Reduction Unit Using Bosch Reaction And Expendable Catalyst Cartridges," NASA CR-1682, November 1970.