II.C.4 Enhanced Hydrogen Production Integrated with CO_2 Separation in a Single-Stage Reactor

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Objectives

- Demonstrate success of a technology to effectively and economically produce a pure hydrogen stream by coal gasification with integrated capture of CO_2 emissions.
- Demonstrate a high reactivity, mesoporous calcium oxide for *in situ* carbon dioxide separation.
- Test the regenerability of the sorbent over multiple calcination-carbonation cycles.

Introduction

Currently, there is renewed interest in the production of hydrogen where its separation is integrated with the water-gas-shift (WGS) reaction in a single-stage reactor. The project is an effort to demonstrate the success of a technology to effectively and economically produce a pure hydrogen stream by coal gasification with integrated capture of carbon dioxide emissions, for their subsequent sequestration. A high reactivity, mesoporous calcium oxide will be demonstrated for *in situ* carbon dioxide separation. The regenerability of the sorbent over multiple calcination-carbonation cycles will be tested.

Approach

The first step will involve identification of the temperature range selective towards carbonation by temperature-programmed reaction of the calcium oxide sorbent in a thermal gravimetric analyzer (TGA). Differential experiments will be performed for estimation of kinetic parameters of the carbonation reaction. This will be followed by life-cycle testing of the sorbent by *in situ* carbonation and vacuum/steam calcination (regeneration). Subsequently, integral (breakthrough) experiments will be carried out to measure the effectiveness of commercial WGS catalysts along with the calcium oxide sorbent for coupled WGS and carbonation reactions in the selected high temperature range. Preliminary evaluation of the catalytic activity of calcium carbonate for WGS reaction alone by integral experiments will be followed by investigation of the catalytic activity of calcium for coupled WGS and carbonation reactions. Finally, the selectivity of CaO for H_2S/CO_2 by simultaneous heterogeneous gas-solid reactions under entrained flow conditions will be explored.

Accomplishments

- Completed thermodynamic analysis of kinetics of carbonation of CaO under simulated fuel gas conditions.
- Initiated catalyst life-cycle testing.
- Completed construction and shakedown of reactor system.
- Procured and set up vacuum calcination studies/system components (MSB-TGA, gas analyzers, etc). Completed the design and fabrication of the new reactor.
- Completed the new reactor shakedown/sorbent testing/online gas analysis and completed the regeneration of CaS to $CaCO_3$ to provide a sulfur removal unit upstream of the carbonation/hydrogen generation reactor.
- Completed the carbonation of CaO under multiple fuel gas concentrations and initiated catalyst screening.
- Completed multiple cycle tests of carbonation and calcination under simulated process conditions in the TGA. Completed hydrogen generation enhancements.

Future Directions

- Investigate the catalytic activity of calcium for coupled WGS and carbonation reactions.
- Determine selectivity of CaO for H_2S/CO_2 by simultaneous heterogeneous gas-solid reactions under entrained flow conditions.