Title: **Development of a Catalyst/Support for Methane Reforming**

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Grant Number: DE-FG26-04NT42182
Performance Period: October 1, 2004 to September 30, 2007

ABSTRACT

OBJECTIVES

The overall objective of this project is the development and laboratory demonstration of a unique material which combines a catalyst for reforming methane with a sorbent for the byproduct carbon dioxide. The material has the potential for markedly simplifying the process for reacting steam with methane or carbon monoxide to produce and separate hydrogen in high concentration with an attendant increase in process efficiency.

ACCOMPLISHMENTS TO DATE

The combined catalyst and sorbent under development is designed to increase the rate of reaction of steam with methane or carbon monoxide to produce hydrogen while simultaneously absorbing the carbon dioxide produced as a byproduct. Therefore, the material has the potential for greatly simplifying the process used for converting the products of coal gasification into relatively pure hydrogen, free of carbon dioxide and carbon monoxide. The catalyst and sorbent are combined into pellets which individually consist of a calcium-based core encased in a porous shell impregnated with a nickel catalyst. The concept has been demonstrated by pelletizing powdered limestone or dolomite to form the cores and then coating the cores with a mixture of powdered alumina and limestone followed by calcination to partially sinter the coating. The pellet shells are subsequently impregnated with a solution of nickel nitrate, heated to decompose the deposited nickel salt, and then treated with hydrogen to reduce nickel to its metallic state.

The utility of the material has been demonstrated in laboratory experiments in which methane or carbon monoxide or both are reacted with steam to produce hydrogen in concentrations of 95% or more. Since the reforming reaction with steam takes place between 550 and 650º where carbon dioxide is also absorbed, the pellets can be regenerated by interrupting the flow of reactants and raising the temperature to 750ºC. Alternatively, when reforming is conducted under pressure, the pellets can be regenerated by reducing the pressure.

Several problems have been identified and are currently being addressed. Problems arise from the high temperature instability of the materials which results in a decline in surface area of both the calcium-based sorbent and the alumina catalyst support as the pellets are heated to the high temperatures required for fabrication and utilization. The high surface area activated alumina

being considered as a catalyst support suffers a significant loss of surface area when the pellets are initially calcined for strengthening which ultimately causes a decline in catalyst activity. Furthermore, during usage as the pellets undergo many absorption and regeneration cycles, the absorption capacity of the calcium-based core material declines markedly.

These issues are being addressed by incorporating various additives which have been found to increase the thermal stability of the core and shell materials. One of the additives being tested is lanthanum which not only stabilizes the surface area of alumina, it also greatly enhances the strength of the pellet shells. Another additive being tested is magnesia which has been shown to stabilize the absorption capacity of calcium oxide at high temperature. A larger and more versatile reaction system has also been constructed for laboratory testing of core-in-shell pellets over a wider range of experimental conditions.

FUTURE WORK

Core-in-shell pellets will be prepared using the best formulations which have been found for the individual components. The pellets will be evaluated by reacting steam with methane and/or carbon monoxide at different temperatures and pressures with and without hydrogen sulfide present. The physical strength of the pellets will be measured, and limited life cycle testing of the most promising pellets will be conducted.

LIST OF PAPERS PUBLISHED

J. A. Satrio, B. H. Shanks, and T. D. Wheelock, "Development of a Novel Combined Catalyst and Sorbent for Hydrocarbon Reforming", *Ind. Eng. Chem. Res.*, **44**, 3901-3911 (2005)

J. A. Satrio, B. H. Shanks, and T. D. Wheelock, "A combined Catalyst and Sorbent for Enhancing Hydrogen Production from Coal or Biomass", submitted to *Energy & Fuels*, March, 2006.

LIST OF CONFERENCE PRESENTATIONS

J. A. Satrio, B. H. Shanks, and T. D. Wheelock, "A Combined Catalyst and Sorbent for Enhancing Hydrogen Production form Coal", presented at the Clearwater Coal Conference, Clearwater, Florida, April 17-21, 2005.

B. H. Shanks and T. D. Wheelock, "Development of a Catalyst/Sorbent for Methane Reforming", poster presentation at the DOE University Coal Research Contractors Review Meeting, Pittsburgh, PA, June, 2005.

STUDENTS SUPPORTED UNDER THIS GRANT

- Karl Albrecht, graduate student in chemical engineering
- Janine Keeley, undergraduate student in chemical engineering
- Tanya Harris, undergraduate student in chemical engineering