High Temperature Electrochemical Removal of Hydrogen Sulfide from Coal Gasification Process Streams

CONTRACT INFORMATION

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ABSTRACT

Objective: An advanced process for the separation of hydrogen sulfide (H_2S) from coal gasification streams through an electrochemical membrane is being perfected using funds from this grant. H_2S is removed from a syn-gas stream, split into hydrogen, which enriches the exiting syn-gas, and sulfur, which is condensed downstream from an inert sweep gas stream. The process allows for continuous removal of H_2S without cooling the gas stream while allowing negligible pressure loss through the separator. Moreover, the process is economically attractive due to the elimination of the need for a Claus process for sulfur recovery. To this extent the project presents a novel concept for improving utilization of coal for more efficient power generation.

Research Results: Lithiated nickel (porosity-85%) was identified as a strong candidate for both the cathode and anode due to its excellent conductivity and stability. Yttria-stabilized zirconia membranes (porosity ~65%) proved very effective, creating a barrier against H_2 diffusion as well as providing a low resistance path for ion migration and diffusion. Both were successfully utilized in full-cell experimental runs.

Experimental full-cell runs for sour coal gas streams (2500 ppm and 4200 ppm H_2S) at a run temperature of 580 °C for flow rates between 100-700 cc/min have yielded over 90% H_2S removal with corresponding current efficiencies above 90%. The stable phases of the cathode in raw coal gas

containing > 60 ppmv H_2S at 650 °C are $Ni_{3-x}S_2$ and Ni_3S_2. However, these constituents form an eutectic at 640 °C which dramatically decreases cathode surface area. Run temperatures at 580 °C alleviated this problem.

Modeling of the H_2S removal system was performed. The model, which accounted for Nernstian effects, concentration and activation overpotential, and ohmic losses, agreed well with experimental results, specifically predicted cell potentials and current efficiencies.

Significance to the Fossil Energy Program: The Fossil Energy Advanced Research Program requires high temperature separations to remove environmental contaminants from post-combustion fuel gases as well as pre-combustion process gases. This project is aimed at the latter; the removal of hydrogen sulfide from coal gas at gasifier temperatures. This development would enable a simplification of the entire gasification operation by permitting a continuous one-step removal of hydrogen sulfide and production of elemental sulfur. High temperature processing and the elimination of a Claus plant for sulfur regeneration would result in energy and monetary savings.

Future Activities: Future goals will focus on optimizing the cell materials. H_2S removal experiments with stainless steel cell housings which have been aluminized will be attempted. Also, the manufacture of a cobalt cathode and subsequent incorporation into full cell runs will be studied. Full cell experiments consists of determining H_2S removal and current efficiencies at various inlet H_2S concentrations and gas flow rates.

Further work will also focus on the H_2S removal model, which theoretically describes the preferred reduction of H_2S, the transport of S^{2-}, and the competing transport of CO_2. The model should identify the maximum current efficiency for H_2S removal and cross-cell potentials, depending on variables such as flow rate, temperature, and current application. The effect of gas crossover will be incorporated into the model.

ARTICLES AND PRESENTATIONS

J.S. Robinson and J. Winnick, "High Temperature Electrochemical Separation of H_2S from Coal Gasification Streams", Proceedings of the AIChE, 1994.

J.S. Robinson and J. Winnick, "High Temperature Electrochemical Separation of H_2S from Coal Gasification Streams", AIChE Meeting, Atlanta, GA, 1994.

Jeffrey S. Robinson and Jack Winnick, "Removal of Hydrogen Sulfide from Coal Gasification Streams Using an Electrochemical Membrane Separation Process", Proceedings of the AIChE, 1996.