PROCESS INTENSIFICATION DURING POWER GENERATION VIA MEMBRANE-BASED REACTIVE SEPARATIONS

Ashkan Garshasbi¹, Doug Parsley², Richard J. Ciora², Jr., Paul KT Liu², and Theodore T. Tsotsis¹*

¹Mork Family Department of Chemical Engineering and Materials Science, University of Southern California, HED 216, Los Angeles, CA 90089-1211, (USA)
²Media and Process Technology, Inc., Pittsburgh, PA 15328, (USA)

Abstract

A novel technology, termed the "one-box" process, has been proposed in order to economically produce pure hydrogen from coal/biomass gasification for use during power generation. The heart of the process is a catalytic membrane reactor (MR) making use of carbon molecular sieve membranes (CMSM). The MR also incorporates a sulfur-resistant Co/Mo/Al₂O₃ catalyst. Experimental studies have been carried out in the laboratory to determine the catalytic reaction kinetics, membrane characteristics, and the MR performance. These are coupled with extensive field-testing of commercial-scale CMSM modules in the real coal/biomass gasification environment, which are conducted at the US National Carbon Capture Center (NCCC) coal gasification facility. Testing of the "one-box" process is also currently under way in the same NCCC facility, and the findings will be presented during the meeting.

Keywords

Membrane Reactor, Clean Coal Processing Technologies, Hydrogen Production, Process Intensification

Introduction

Producing hydrogen from syngas from coal and/or biomass gasification is a significant first step in the environmentally-benign power generation from these important raw resources for several nations including the USA [1]. Integrated gas combined cycle (IGCC) power plants, in particular, show promise for environmentallybenign power generation from coal and/or biomass. The current IGCC process involves first reacting coal and/or biomass with steam and/or oxygen in a gasifier to produce syngas. The syngas is then, typically, cooled down in order to remove various contaminants such as H₂S, NH₃, tars, organic vapors, and other impurities; it is then reheated to be further reacted with steam in a water gas shift (WGS) reactor to maximize its hydrogen content. The WGS reaction is exothermic, and its equilibrium conversion decreases with temperature. Therefore, typically, two reactors are deployed, one operating at high temperatures (HTS) and the other at a lower temperature (LTS), in order to overcome simultaneously both equilibrium and kinetic limitations, and to thus increase CO conversion at practical space velocities [2]. The total process, as it is now envisioned, is complex and energy-intensive, and thus not very attractive for application in the context of power generation with carbon capture and sequestration (CCS).

To supplant the conventional WGS reactor process, this team has proposed a novel technology, termed the "one-box" process [3]. The heart of the process is a catalytic membrane reactor (MR) making use of carbon molecular sieve membranes (CMSM). The membrane reactor also incorporates a sulfur-resistant Co/Mo/Al₂O₃ catalyst. Experimental studies are being carried out in the laboratory to determine the catalytic reaction kinetics,

^{*} tsotsis@usc.edu

membrane characteristics, and the MR performance. These are coupled with extensive field-testing of commercialscale CMSM modules in the real coal/biomass gasification environment, which are conducted at the US NCCC coal gasification facility [4]. Testing of the "one-box" process is also currently under way in the same facility, and findings will be presented during the meeting. High-temperature MR have the potential to enhance process intensification and to increase energy savings and/or product yield. But though the potential benefits of such reactors are substantial, commercialization still remains elusive. A major technical barrier is the lack of robust membranes and full-scale modules which are suitable for use at the high-temperature and high-pressure conditions required. A key advance from this research is the development of such membranes, which are robust for the proposed IGCC application [4].

Results

The feasibility of the "one-box" process was previously proven in the laboratory for pressures up to 5 atm [3]. The focus of our recent efforts was to expand the testing for higher pressures, which are more relevant to the coal gasification environment. Figure 1, for example, shows experimental CO conversions for the 'one-box' system for feed pressures of ~ 14 atm, and compares such conversions with those measured with a packed-bed reactor (PBR). The MR shows superior performance to that of the PBR for the whole range of conditions studied.



Figure 1. Conversion of MR and PBR,

Towards technology commercialization, full-scale multi-tubular CMSM modules have also been constructed and characterized comprehensively, and their effectiveness has been demonstrated. Our successful preparation of these membrane modules overcomes some of the technical barriers which have hampered to date the commercial implementation of inorganic membranes in reactive separations. Field-tests of these membranes have been conducted at the US NCCC under conditions suitable for the "one-box" process. During continuous use in raw coalderived and/or biomass-derived syngas, the CMSM successfully rejected H₂S, NH₃, tar-like species, etc. present in the syngas without any evidence of fouling. Figure 2, for example, shows the H₂ content (dry-basis) in

the permeate and reject streams obtained during the membrane module testing at NCCC.



Figure 2. Hydrogen concentration (dry-basis) in the permeate and reject sides during the testing of the "fullscale" CMSM module feed with air blown coal-fed gasifier off-gas at 250~ °C and ~14.8 bar.

Conclusions

A novel technology, termed the "one-box" process has been proposed in order to economically produce pure hydrogen from coal/biomass gasification for use during power generation. It utilizes a catalytic MR making use of CMSM and sulfur-resistant Co/Mo/Al₂O₃ catalysts. Experimental studies have been carried out in the laboratory to determine the catalytic reaction kinetics, membrane characteristics, and the MR performance. These are coupled with extensive field-testing of commercialscale CMSM modules in the real coal/biomass gasification environment at the NCCC. Testing of the "one-box" process is also currently under way in the same facility, and findings will be presented during the meeting.

Acknowledgments

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