

# **Economic Rationale for Safety Investment in Integrated Gasification Combined-Cycle Gas Turbine Membrane Reactor Modules**

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**JEL Classification** G11, G31, G32

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## **ABSTRACT**

A detailed Net Present Value (NPV) model has been developed to evaluate the economic viability of an Integrated Gasification Combined Cycle – Membrane Reactor (IGCC-MR) power plant intended to provide an electricity generating and pure H<sub>2</sub> (hydrogen) producing technology option with significantly lower air pollutants and CO<sub>2</sub> (carbon dioxide) emission levels, where the membrane reactor module design conforms also to basic inherent safety principles. Sources of irreducible uncertainty (market, regulatory and technological) are explicitly recognized, such as the power plant capacity factor, Pd (palladium) price, membrane life-time and CO<sub>2</sub> prices (taxes) due to future regulatory action/policies. The effect of the above uncertainty drivers on the project's/plant's value is elucidated using a Monte-Carlo simulation technique that enables the propagation of the above uncertain inputs through the NPV-model, and therefore, generate a more realistic distribution of the plant's value rather than a single-point/estimate that overlooks these uncertainties. The simulation results derived suggest that in the presence of (operational, economic and regulatory) uncertainties, inherently safe membrane reactor technology options integrated into IGCC plants could become economically viable even in the absence of any valuation being placed on human life or quality of life by considering only equipment damage and interruption of business/lost production cost. Comparatively more attractive NPV distribution profiles are obtained when concrete safety risk-reducing measures are taken into account through

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## **1. Introduction**

Integrated Gasification Combined Cycle (IGCC) combustion of coal represents a promising technology option with the potential to secure numerous energy policy goals. IGCC chemically converts coal into a synthetic gas (syngas) for combustion in a combined cycle power plant. The gas turbine and steam turbine are similar to those found in the well established technology Combined Cycle Gas turbine (CCGT), as widely used for electricity generation from natural gas. IGCC represents therefore a power generation process that integrates a gasification system with a conventional combustion turbine-based combined cycle power block. As mentioned above, the gasification system converts coal into syngas comprised predominantly of carbon monoxide and hydrogen. The combustible syngas is first treated for the removal of sulfur, nitrogen oxides and particulate matter and then used to fuel a combustion turbine to generate electricity while the exhaust heat from the turbine is used to produce steam for a second generation cycle through a system turbine as well as steam used in the gasification process. Please notice that both major components of IGCC, namely the gasification island and the combined cycle power block are associated with mature and well-tested technologies and have been broadly utilized in the petrochemical, chemical processing industries as well as natural gas-based power generation. However, their integration represents a fairly recently conceived technology option to produce commercial electricity. This new technology option has been demonstrated at only a handful of facilities around the world and its performance is currently evaluated in technical and environmental terms. In terms of technical performance, the thermodynamic efficiency of the IGCC plant can be expected to exceed that of conventional pulverized coal (PC) combustion methods. Significant

benefits can be realized due to the enhanced environmental performance of IGCC. The removal of impurities such as sulfur and mercury before syngas combustion ensures superior pollution credentials for an IGCC plant over PC alternatives (where post-combustion treatment of the exhaust flue gas takes place). Indeed, IGCC's environmental performance results in a cost-effective emissions reduction (above the 95% removal targets) of major pollutants such as nitrogen oxides, sulfur and sulfur oxides, particular matter and mercury. Furthermore, an IGCC power plant can accommodate a CO<sub>2</sub>-capture unit at a cost significantly lower compared to the PC case. On the technical performance level, efficiencies in IGCC with carbon capture units are slightly higher than PC, while on the environmental performance level the CO<sub>2</sub> (carbon dioxide) separation and capture can be accomplished more efficiently and in a cost-effective manner due to its high partial pressure (which makes IGCC with carbon capture quite attractive when sequestration and attendant costs are considered). Finally, it should be pointed out that for a country, such as the United States, with abundant coal reserves IGCC technology can help transition coal into being a modern fuel for high technology combustion purposes while preserving its long-standing benefits in terms of energy security and energy independence.

IGCC syngas contains a large proportion of hydrogen. IGCC-Membrane Reactors take this situation further. Catalytic membrane reactors permit a multitude of processes and associated application fields such as dehydrogenation, hydrogenation and oxidation reaction systems (Takht Ravanchi et al. 2009). In particular, Pd-based (Palladium-based) composite membrane reactors enable the water gas shift reaction and selective H<sub>2</sub> (hydrogen) separation to take place simultaneously in a single process unit and conversion levels to exceed the thermodynamically limited equilibrium ones through the continuous removal of H<sub>2</sub> on the products side, while ensuring high hydrogen fluxes, stable selectivity and enhanced thermal, mechanical and chemical stability. These attributes ensure that an IGCC-MR can be regarded as both a source of electricity generation and hydrogen suitable for a wide range of emerging applications (such as in fuel cells for vehicular transport). The aforementioned considerations of energy supply security considerations, growing environmental concerns and developments in the global fuel markets provide motivation for the examination of: i) the possibility of hydrogen production, a valuable energy carrier and feed to the petrochemical and chemical

processing industries, from coal which, along with natural gas, represent the favorite fuels for electric power generation, and ii) within the Integrated Gasification Combined Cycle (IGCC) context, the possibility of co-production of electricity and hydrogen (as well as other valuable chemicals). IGCC-MR technology is notable for its potential to contribute to a cleaner energy future in both electricity and fluid vehicle fuels.

From an environmental performance standpoint, one of the primary concerns related to coal combustion and gasification in coal-fired power plants is the significant amount of CO<sub>2</sub> emissions, coupled with the production of air pollutants (nitrogen and sulfur oxides) as well as toxic substances such as mercury, mentioned earlier. In light of the above remarks, the need for the development of technologies for pre-combustion CO<sub>2</sub> sequestration and economical co-production of hydrogen and electricity in coal-fired power plants is therefore well justified and quite important in order to potentially address some of the key issues that the global energy economy is facing (Veziroglu and Barbir 1998). Since carbon dioxide represents a key greenhouse gas (Amelio et al. 2007), Pd-based composite membrane reactor technology could provide the means for the simultaneous CO<sub>2</sub> capture and extra purity H<sub>2</sub> production in coal-fired power plants realized in a single process unit. Consequently, research activity increasingly now focuses on the development of new membrane technology options that would provide cost-effective strategies for further reduction of CO<sub>2</sub> emissions through enhanced pre-combustion capture and removal (due to the high quality of separation of CO<sub>2</sub> and hydrogen under favorable thermodynamic conditions attainable in the presence of an integrated membrane reactor module), while enhancing H<sub>2</sub> production and, in particular, the technical feasibility of projects involving co-production of hydrogen, valuable chemicals and electricity via coal gasification (Tarun et al. 2007). Within the aforementioned context, the IGCC-MR process system naturally represents a key option to co-produce synthesis gas, electricity, hydrogen, fuels and chemicals from coal and coal/biomass-mix in an environmentally responsible manner (Ratafia-Brown et al. 2002; Beér 2000).

## 2. Technical Background

Sulfur and mercury removal from the syngas exiting the gasifier of an IGCC plant are followed by the water-gas shift reaction (WGSR) shown below and hydrogen separation that occur simultaneously in a palladium-based membrane reactor:



It should be pointed out that the permeation of H<sub>2</sub> through metals is a complex multistep process (Shu et al. 1991). The major steps involved in the favorable H<sub>2</sub> permeation through Pd that eventually results in a high quality separation of CO<sub>2</sub> from hydrogen via a membrane reactor are: (i) adsorption and dissociation of H<sub>2</sub> molecules to H atoms at the membrane surface, (ii) diffusion of the H atoms through the bulk of the Pd layer, and (iii) re-association of H atoms and desorption of H<sub>2</sub> molecules at the membrane surface (Ward and Dao 1999; Bose 2009). Once most of the H<sub>2</sub> is removed via a Pd-based membrane, the outlet stream of the reaction side consists of mostly H<sub>2</sub>O and CO<sub>2</sub> under high pressure. Please notice that after the condensation of steam and given the fact that CO<sub>2</sub> is at a high pressure (~25 atm), a significant reduction in the compression costs associated with the operation of the sequestration units downstream is attainable (Basile et al. 2001; Basile et al. 1996; Brunetti et al. 2009; Criscuoli et al. 2000; Tosti et al. 2003; Uemiya et al. 1991).

Pd/Pd-alloy membranes supported on metal (stainless steel, Hastelloy or Inconel) were considered in the present study (Ayturk et al. 2006; Mardilovich et al. 1997; Mardilovich et al. 2002; Pomerantz and Ma 2009; Chen and Ma 2010b). Pd-alloy, supported on metal substrates and fabricated through electroless plating, exhibits many advantages such as: high permeability and selectivity, cost-effective fabrication and maintenance, ease of scale-up, practical assembly/disassembly for both small and large scale industrial applications, and long term durability (5 years, 2015 DOE target (US DOE-Hydrogen from coal, 2011)) including resilience at high temperature (400-600 °C) and pressure (20-50 atm). These are the reaction conditions required for dehydrogenation, steam reforming and high temperature WGSRs and have been described by Armor (1998)). It should be pointed out that the poisoning of the

Pd/Pd-alloy membranes by some of the feed components/impurities such as CO and H<sub>2</sub>S represents a notable challenge in the design and development of membrane reactor systems. However, Pd/alloy membranes such as Pd/Cu and Pd/Au, have shown promising results in the presence of H<sub>2</sub>S (Chen and Ma 2010b; Chen and Ma 2010a; Chen and Ma 2010a; Kulprathipanja et al. 2005; Pomerantz et al. 2010; Way et al. 2008; Chen and Ma 2010c). The effect of CO on the H<sub>2</sub> permeation is strongly dependent on reaction temperature, hence the reduction in the permeation rate may be neglected at temperatures higher than ~350°C (Hara et al. 1999; Gallucci et al. 2007; Nguyen et al. 2009).

As with any new technology, the Pd/Pd-alloy based composite membrane reactor technology has to demonstrate its technical feasibility and economic viability for a transition from pilot to large-scale applications. Bracht et al. (1997) conducted one of the earliest economic analysis involving a two step adiabatic WGS-MR system for CO<sub>2</sub> removal in an IGCC plant. The characteristics of micro-porous silica membranes were explicitly taken into account in the economic evaluation efforts. The water gas shift membrane reactor concept was found to result in higher efficiencies coupled with lower costs when compared to more conventional options (low temperature wet gas cleaning) for CO<sub>2</sub> removal. Criscuoli et al. (2001) performed an insightful process-economic analysis involving water gas shift palladium membrane reactors. Energy requirements, catalyst and palladium costs were calculated based on the feed composition of the industrial scale oxo-synthesis gas plant in Augusta (Italy). A significant reduction in capital and operating costs for the membrane reactor module was shown for Pd thicknesses of 20 µm and lower. The effect of an H<sub>2</sub> permeability falling within the range of (1-10 mol.m/[m<sup>2</sup>.s.Pa<sup>0.5</sup>]) on the membrane reactor module cost was also evaluated (Criscuoli et al. 2001). More importantly, those authors' calculations demonstrated that there was a limit value both for the permeability and Pd thickness (high permeability and low thickness) beyond which the membrane module cost does not vary noticeably with H<sub>2</sub> permeability and Pd thickness because the reaction rate becomes slower than the permeation rate, and thus, the rate controlling step. A membrane reactor module with 75 µm thick Pd layer was found to be more expensive compared to the conventional technology with high and low shift reactors and separation devices. Criscuoli et al. (2001) concluded that the preparation of defect-free Pd



membranes with a selective layer of  $\leq 20 \mu\text{m}$  would make the Pd-based membrane technology competitive with conventional technologies for  $\text{CO}_2$  separation.

Rezvani et al. (2009) performed very interesting work comparing coal-fired IGCC systems with  $\text{CO}_2$  capture using physical absorption, membrane reactors and chemical looping. Conventional physical absorption, water gas shift reactor membranes and two chemical looping combustion cycles (CLC), which employ single and double stage reactors, were considered (Rezvani et al. 2009). Particularly in the membrane reactor case, the IGCC system was configured with a water gas shift membrane reactor (WGSMR) and an oxygen transport membrane (OTM) system instead of a physical absorption unit to increase the power plant efficiency and to improve process economics. The OTM unit displayed the capacity to utilize the remaining combustibles in the gas coming from the retentate side of a WGSMR. More importantly, the membrane based technologies resulted in the lowest breakeven electricity selling price (levelized cost of energy) at a fuel cost of  $\text{€}3/\text{GJ}$  ( $\text{€}65.98/\text{MWh}$  at an 8% discounted cash flow rate).

### **3. Economic Background**

The performance and economic attractiveness of membrane reactors for  $\text{H}_2$  production from coal plants have been investigated by Dolan et al. (2010). The production of 300 tpd  $\text{H}_2$  with an 85%  $\text{H}_2$  yield using a  $40 \mu\text{m}$  thick Pd-25Ag wt% membrane was made possible with a total membrane area of  $25000 \text{ m}^2$ . The estimated capital cost of the membrane reactor module was  $\text{\$US} \sim 180$  million, much higher than the capital cost of a unit including high and low temperature shift reactors, amine-based  $\text{CO}_2$  capture and PSA-based  $\text{H}_2$  separation ( $\text{\$US} \sim 55$  million). Dolan et al. (2010) emphasized that the membrane thickness had to be lower than  $20 \mu\text{m}$ , with a plant cost of  $\text{\$US} \sim 55$  million and total membrane area of  $13000 \text{ m}^2$ , to meet the 2015 US DOE cost and flux target levels. Dijkstra et al. (2011) tested a bench scale multi-tube membrane reactor and performed a techno-economic evaluation. Interestingly, the results of the techno-economic analysis indicated that membrane water gas shift systems had better prospects for  $\text{CO}_2$  capture than membrane reforming. The most promising finding was the

combination of a membrane water-gas shift reactor with gas heated reforming resulting in higher efficiencies and CO<sub>2</sub> avoidance costs that were found to be lower than in a conventional pre-combustion CO<sub>2</sub> capture system using a Selexol unit.

All major bench-scale performance assessment studies involving Pd/Pd-alloy based composite membranes highlight certain advantages of scaling up to industrial applications. Consequently, many research groups are now increasingly focusing on process intensification concepts and methods allowing improvements in process economics, environmental performance and process safety through the design of cheaper processes, smaller equipment/plant, inherently safe process design, efficiency-focused energy management, waste/by-product minimization and risk (Stankiewicz and Moulijn 2004). Within such a context, membrane reactor technology nicely exemplifies the above possibilities since it is inherently aligned with, and amenable to, basic process intensification and inherently safe process design principles (Ayturk et al. 2009; Koc et al. 2011). It should be also pointed out that the performance of Pd-based membrane reactors has not yet been evaluated at industrial conditions. Furthermore, the lack of operating experience associated with membrane reactor technology options integrated into IGCC power plants on the commercial scale inevitably results in a dearth of real data pertinent to process safety and economics. Consequently, any safety and economic performance evaluation at this stage must be driven by reasonable, yet theoretical estimates. Methodologically, one must acknowledge irreducible uncertainties (market, regulatory, and technological) in an explicit manner. The present research study conforms to such a methodological approach.

Since pulverized coal-fired power plants started operating in the 1920s (WRI-Pulverized Coal Power, 2011), significant operating experience has been accumulated and sound safety procedures have been established. Therefore, in the case of Pd-based membrane reactor technology options integrated into coal-fired power plants the membrane reactor module needs to be considered as a new “node” and HAZOP analysis must be updated. Since the integration of Pd-based membrane reactors into coal-fired power represents a new technology option which has neither been fully tested nor yet demonstrated at a commercial scale, and given the operating and structural characteristics of membrane reactors, there is a

timely need to evaluate inherent safety design prospects and hopefully to demonstrate that inherent process safety does not undermine the economic viability of such a plant. Perhaps, in an uncertain world, inherent process safety enhances the value of an engineering design, and hence the project itself, in concrete economic terms (Mannan, 2011).

The first systematic attempt to analyze and understand the issues and challenges related to Pd-based membrane reactor safety aspects was made by Chieppetta et al. (2006). Following, a methodologically similar approach HAZOP analysis was performed by Koc et al. (2011) for WGS Pd/Pd-alloy based membrane reactors. The absence of adequate control of the reactor temperature as well as the purity of the feed (which may cause hot spots and a decline in permeance and selectivity) were identified and classified as critical for the operation of the WGS membrane reactor. Since the main advantage of the Pd/alloy-based membrane reactor technology is the high-quality H<sub>2</sub> separation driven by the H<sub>2</sub> partial pressure difference between the reaction and permeate sides, operation at high reaction side pressure was identified as a key process safety challenge. It was also shown that proper material selection, stringent process monitoring and control together with multiple pressure relief systems have to be explicitly integrated into process system design as a part of a comprehensive process intensification strategy (Koc et al. 2011). However, the primary challenge in explicitly incorporating and implementing carefully designed process safety strategies and thoughtful approaches, such as an inherently safe process design, remains the development of a transparent and sound economic justification. One would argue that historical experience and empirical evidence would advance the argument that investments in process safety make economic sense in an uncertain world where relatively low probability-major consequences events actually do happen. However, significant progress in process safety also introduced a sense of complacency by facilitating rationalizations that major catastrophic events can be averted, their consequences minimized due to “superior” existing process knowledge and availability of technological means, and as a result reinforcing resistances against sustained investments in process safety (Pasman 2000). The objective of the present work is to show that if the required process safety investment is made (as part of the initial capital expenditure) at the construction stage of an IGCC plant, then process safety investment

strategies can improve the economics of the project by preventing or minimizing the effects of accidents and possibly catastrophic events.

The paper is organized as follows: In section 4 the structure and development of a technical performance and economic assessment framework in the presence of (market, regulatory and technological) uncertainty are presented for inherently safe membrane reactor technology options integrated into IGCC plants. The present study's main results and a pertinent discussion can be found in Section 5, followed by a few concluding remarks in Section 6.

## **4. Technical Performance Assessment and Economic Analysis Framework**

### **4.1 Integration of Pd/alloy-based membrane reactors into IGCC plants: Technical performance assessment**

In this section we consider options for inherently safe Membrane Reactor technology as integrated into IGCC Power Plants. In this subsection we consider the technical performance issues. In the following subsection we shall consider the economic issues.

The goal of co-production of hydrogen and electric power using a high temperature hydrogen separation system (Case 6 – Mitretek technical paper (Gray and Tomlinson 2002)) underlies the integration option idea of Pd-based membrane reactors into a coal gasification – combined cycle power block (traditional IGCC) system. A key unit in such a process system is the gasifier and auxiliary equipment. Three companies GE, Shell and ConocoPhillips dominate the gasification business through the introduction of advanced technology options bound to play a major role in future IGCC demonstration projects (Maurstad, 2005). Operating conditions and syngas properties associated with the GE Energy gasifier such as a high operating pressure regime, H<sub>2</sub>O:CO mole ratio and low concentrations of impurities (H<sub>2</sub>S, COS, etc) make the GE gasifier quite attractive when Pd-based membrane reactors are to be

integrated into IGCC power plants. Therefore, the GE Energy gasifier has been selected for the ensuing technical and economic performance assessment in the present study.

The detailed block flow diagrams of the GEE IGCC plant with CO<sub>2</sub> capture reported in DOE/NETL report (Haslbeck et al. 2010) were compared with the target flow diagram of a Pd-based membrane reactor embedded into the IGCC plant (IGCC-MR). The detailed flow diagram of the IGCC-MR after the requisite modifications is shown in Figure 1. The gray colored units were specific for the IGCC plant with traditional packed bed reactors (IGCC-PBR) and the blue colored units are designated for the IGCC plant with a membrane reactor module embedded. In a conventional IGCC plant with CO<sub>2</sub> separation, the coal is converted to hydrogen in five main steps: the first process step involves the air separation taking place in the air separation (ASU unit), followed by the coal gasification step using oxygen feed. The third and fourth steps are associated with hydrogen production in the aforementioned water-gas-shift reactors and H<sub>2</sub>/CO<sub>2</sub> separation in the Selexol units that follow. The final step is realized by the combined cycle electricity generation block shown Figure 1. Since reaction and separation would take place in the Pd/alloy-based membrane reactor, the high and low temperature shift reactors and also the Selexol unit for H<sub>2</sub> separation were removed from the flow diagram. The membrane reactor is placed after the gas clean-up step to prevent membrane poisoning by sulfur-containing gases. However, the membrane reactor integration introduces an energy penalty at the inlet of the advanced F-class gas turbine (Haslbeck et al. 2010): the separated H<sub>2</sub> coming from the permeate side of the membrane reactor at 1 atm pressure has to be compressed to the working pressure of the gas turbine (~30 atm).

The specific performance target levels for the water-gas shift Pd/alloy-based membrane reactor were set at 98% CO conversion and 95% H<sub>2</sub> recovery. Thus, the exit stream of the membrane reactor at the retentate side would consist of mostly H<sub>2</sub>O and CO<sub>2</sub>. After condensation of the steam, the retentate stream which is comprised of mainly CO<sub>2</sub> at high pressure (~25 atm if the gasifier pressure is ~50 atm) would be ready for sequestration. In addition, separating CO<sub>2</sub> at high pressure through the membrane reactor generates substantial energy savings in terms of CO<sub>2</sub> compression costs as opposed to separating CO<sub>2</sub> at atmospheric pressure via a conventional Selexol unit.

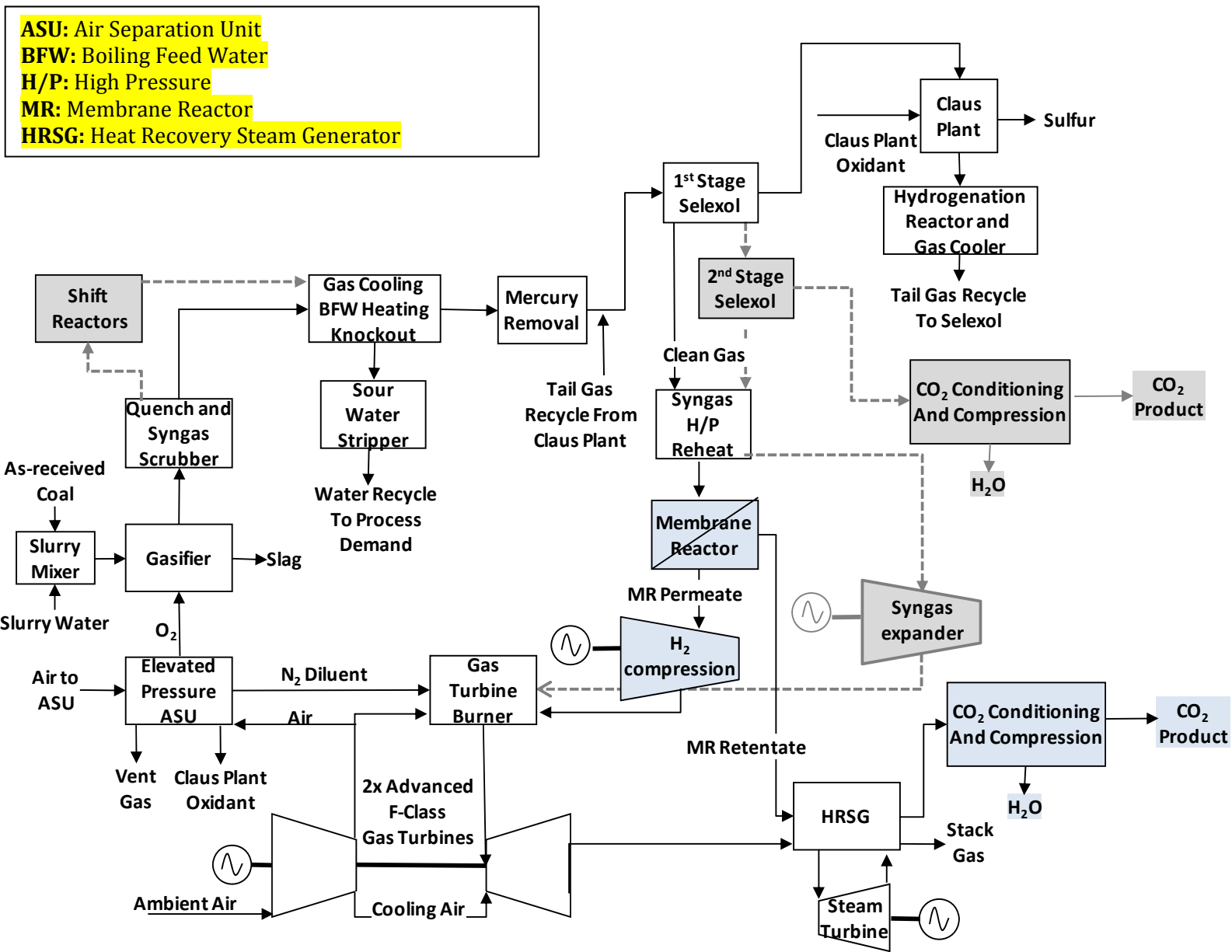


Figure 1. Schematic flow diagram of a Pd-based membrane reactor module integrated into an IGCC plant (IGCC-MR)

The feed specifications, reaction conditions and permeation properties used in the isothermal membrane reactor model are listed in Table 1. In particular, a comprehensive first principle-based two dimensional (2D) membrane reactor modeling framework has been developed to take into account the radial concentration gradients and pressure drop of the Pd-based membrane reactor. The main objective of the 2D model was to reliably address the problem of possibly overestimating H<sub>2</sub> recovery levels and the calculated membrane areas, which were less than the actual required ones as calculated through the traditional simple one-dimensional (1D) model. The main structure of the 2D membrane reactor model is presented in the Appendix.

Quite promising simulation results were found to be in satisfactory agreement with experimental findings involving a lab-scale (0.5"OD<sub>M</sub>×1"OD<sub>S</sub>×2.5"L) membrane reactor within the GHSV range of ~1000-6000 h<sup>-1</sup> (Augustine et al. 2011). The 2D membrane reactor model which runs for industrial scale conditions was evaluated under the feed specifications, reaction conditions and permeation properties given in Table 1.

**Table 1. Feed specifications, reaction conditions and permeation properties**

Total inlet flow rate to the MR [mol/s]	9575.2 (8191 w/o CO <sub>2</sub> comp.)
Feed composition [%]	CO: 23 - CO <sub>2</sub> :9 - H <sub>2</sub> :22 - H <sub>2</sub> O: 46
Reaction temperature [°C]	400
Reaction side pressure [atm]	50
Permeate side pressure [atm]	1
Permeance [m <sup>3</sup> / [m <sup>2</sup> .h.atm <sup>0.5</sup> ]	39
Bulk catalyst density [kg/m <sup>3</sup> ]	990.4

The dimensions, total feed flow rates and exit flow rates of the single membrane tube and also the whole water gas shift membrane reactor, which also conforms to the industrial scale IGCC plant load specifications, are provided in Table 1. Since the total number of Pd/Au-based membrane tubes was quite high, the membrane reactor module was assumed to consist of 8

bundle reactors in parallel, each having 1073 membrane tubes. Based on 2D membrane reactor model calculations, the Pd/Au-based membrane reactor with the aforementioned dimensions was able to achieve ~99% CO conversion and 96% H<sub>2</sub> recovery. From an inherent process safety point of view in particular, the following conditions need to be satisfied by designing and operating the Pd/alloy-based WGSMR in a safety-constrained regime, thus preventing the development and occurrence of hazards to personnel as well as process performance (loss of efficiency, deterioration of process economics, etc), as identified by Koc et al. (2011):

- The membrane reactor and feed stream have to be pre-heated to 300°C before syngas is admitted to the reactor. Pre-heating is necessary to maintain sufficient WGS reaction rates and to prevent H<sub>2</sub> embrittlement of pure Pd-based membranes.
- The extra heat of the WGS reaction (above 450°C) needs to be removed with the aid of a heat exchanger to maintain the isothermal operation of the WGSMR and to prevent potential hazards which could compromise the safety of the WGSMR.
- The main advantage of the Pd/alloy-based membrane reactor technology is the high-quality H<sub>2</sub> separation driven by the H<sub>2</sub> partial pressure difference between the reaction and permeate sides, and therefore, operation at a high reaction side pressure is necessary. Thus, the high pressure operation of the WGSMR represents a safety challenge. As proposed by Koc et al.(2011), the combination of a disk-rupture safety relief valve in series system (RRS) could be installed at the membrane reactor entrance and exit as a safety measure. In addition, sparing of the relief equipment would provide an on-line maintenance option of the RRS system through switching and also the process would not necessarily need to be shut down for replacement purposes (Center for Chemical Process Safety/AIChE 1993).

All safety measures listed above require an initial up-front capital investment in process safety associated with the integration of the membrane reactor into IGCC power plants so as to mitigate potential risks to personnel, the environment, equipment and/or process efficiency and performance.



**Table 2. Industrial scale membrane reactor specifications used for cost analysis**

<b>Single membrane tube</b>		<b>Water gas shift membrane reactor module</b>	
Pd thickness [ $\mu\text{m}$ ]	6.8	Total Pd weight [kg]	1064
Au thickness [ $\mu\text{m}$ ]	0.58	Total Au weight [kg]	145
OD of the membrane, $x_1$ [cm]	5.08	No of membrane tubes	8584
ID of the shell casing, $x_2$ [cm]	7.62	# of bundle MR in series	8
Length of the membrane, L [m]	9.525	Total $V_{\text{shell}}$ [ $\text{m}^3$ ]	372
Membrane area [ $\text{m}^2$ ]	1.52	Total area [ $\text{m}^2$ ]	13043
$V_{\text{annulus}}$ [ $\text{m}^3$ ]	0.02	Total $V_{\text{annulus}}$ [ $\text{m}^3$ ]	207
$F_{\text{total, in}}$ [mol/s]	1.12	Total $F_{\text{total, in}}$ [mol/s]	9575
$F_{\text{CO}_2, \text{exit}}$ [mol/s]	0.35	Total $F_{\text{CO}_2, \text{exit}}$ [Mtonne/year]	4.2
$F_{\text{H}_2, \text{exit}}$ [mol/s]	0.48	Total $F_{\text{H}_2, \text{exit}}$ [kg/s]	8.2
$W_{\text{catalyst}}$ [kg]	23.89	Total $W_{\text{catalyst}}$ [tonne]	205

#### 4.2 Economic analysis framework

The cost figures of the water gas shift reactor and catalyst as well as the assorted equipment were adopted from the detailed block flow diagram of the GEE IGCC with CO<sub>2</sub> capture (with traditional PBR- Plant 4) in the DOE/NTEL report (Haslbeck et al. 2010). The present value of the equipment costs were calculated by using the Marshal & Swift (M&S) equipment cost indexes (M&S cost indexes, 2010). It has to be noted that cost indexes can be used to give a

general estimate, but no index can take into account all factors, such as potential technological advancements. The M&S cost indexes permit fairly accurate estimates to be derived if the time period involved is less than 10 years (Peters and Timmerhaus 1991). In addition, the construction costs of the plants were corrected to find the equivalent cost at the present time on the basis of the Chemical Engineering plant cost indexes (M&S cost indexes, 2010). The cost indexes used in the above cost calculations can be found in

Table 3. The purchased equipment cost of the water gas shift reactor shell casing and 316L PSS supports was estimated by using the six-tenths-factor rule (Peters and Timmerhaus 1991) as given in Equation (2) The original sizes of the water gas shift reactor shell casing, catalyst amount and price were adopted from DOE/NETL report (Haslbeck et al. 2010) and along with the pertinent quote for the 316L PSS supports (from Chand Eisenman, Burlington, CT and Mott Metallurgical Corporation, Farmington, CT) are all listed in Table 4.

$$\text{Cost of equip. a} = \text{cost of equip. b} \times \left( \frac{\text{Capacity of equip. a}}{\text{Capacity of equip. b}} \right)^{0.6} \quad (2)$$

**Table 3. Cost Indexes**

M&S equipment cost indexes (M&S cost indexes, 2010)	In 2006	1302.3
	In 2009	1468.6
Chemical engineering plant cost indexes (M&S cost indexes, 2010)	In 2006	499.6
	In Oct 2009	527.9

The net power output of the IGCC-MR was fixed at 550MWe and only one stream of revenue due to electricity selling was considered (the option of H<sub>2</sub> selling has not been considered in the present analysis).

The Net Present Value (NPV) framework of analysis was used in the economic assessment of the Pd-based membrane reactor integration option into IGCC power plants. The NPV of a series of future cash flows is their present value, minus the initial investment required to obtain the future cash flows (Equation (4)). The NPV of an investment quantifies the increase in wealth that one realizes if the investment on the project is made, and mathematically represented by the following formula (Benninga 2006):

$$PV = \sum_{t=1}^N \frac{C_t}{(1 + r_t)^t} = \frac{C_1}{1 + r_1} + \frac{C_2}{(1 + r_2)^2} + \frac{C_3}{(1 + r_3)^3} + \dots \quad (3)$$

$$NPV = C_o + PV \quad (4)$$

where PV is the present value of the project,  $C_t$  is the cash flow in year  $t$ ,  $r_t$  is the real discount rate in year  $t$ ,  $N$  is the total plant lifetime in years, NPV is the net present value, and  $C_o$  is the initial capital investment (Brealey and Myers 1996). The parameters used in the NPV-model and analysis are listed in Table 4. Furthermore, the assumptions made in the NPV calculations are the following: the plant is ready to operate today (i.e. in 2011), the life time of the gasification plant is 40 years, the depreciation cost was calculated through the declining-balance method (Peters and Timmerhaus 1991), and regulatory action on carbon emissions starts in 2015 with an annual CO<sub>2</sub> growth rate of 3%. A higher discount rate of 9% for the IGCC/MR plant was used to render the high risk associated with this relatively new technology option. In particular, the real discount rate was calculated by using a nominal discount rate along with inflation estimates followed by the appropriate calculation of all future cash flows. Moreover, the IGCC plant with an embedded Pd-based membrane reactor was assumed to achieve a 98% CO<sub>2</sub> capture level. Even though there is no theoretical limit of the CO<sub>2</sub> separation level that can be attained with the Pd-based membrane reactor, 98% so as to be sure not enhance the membrane reactor economic case with an over-optimistic assumption.

**Table 4. Economic parameters**

Plant life [years]	40
Tax rate	40%
Depreciation (annual)	30%
Insurance and property tax rate	1.78%
CO <sub>2</sub> tax (\$/ tonnes CO <sub>2</sub> , starting in 2015)(Bohm et al. 2007)	25
CO <sub>2</sub> tax growth rate (Bohm 2006)	3%
CO <sub>2</sub> transportation & sequestration cost (\$/t CO <sub>2</sub> )(Bohm 2006)	5
Nominal discount rate	9%
Inflation rate	2.50%
Electricity selling price [cents/kWh]	9.97
Electricity selling growth rate[%]	0.5
Plant capacity factor	80%
Pd price [\$ /g, 10 years average]	14.2
Au price [\$ /g, 10 years average]	21.7
Membrane life time [years]	3
Shift catalyst price [\$ /kg 2006](Haslbeck et al. 2010)	6.8
316L PSS supports (0.2 µm media grade) [\$ /cm <sup>2</sup> for a 25 cm <sup>2</sup> support area] 2011	4.4

#### **4.3 Economic assessment under uncertainty: Integration of Monte Carlo methods into the NPV framework of analysis**

As mentioned earlier, the inherent uncertainty associated with key inputs of the NPV-model has to be recognized and explicitly taken into account in investment decision-making. Indeed, single values/estimates of the NPV computed on the basis of average input values (thus overlooking uncertainty) may result in compromised project investment decision-making.

Mathematically, the above is supported by the fact that using the average values of uncertain inputs in a function of random variables does not always result in the average value of the function (as a result of Jensen's inequality):

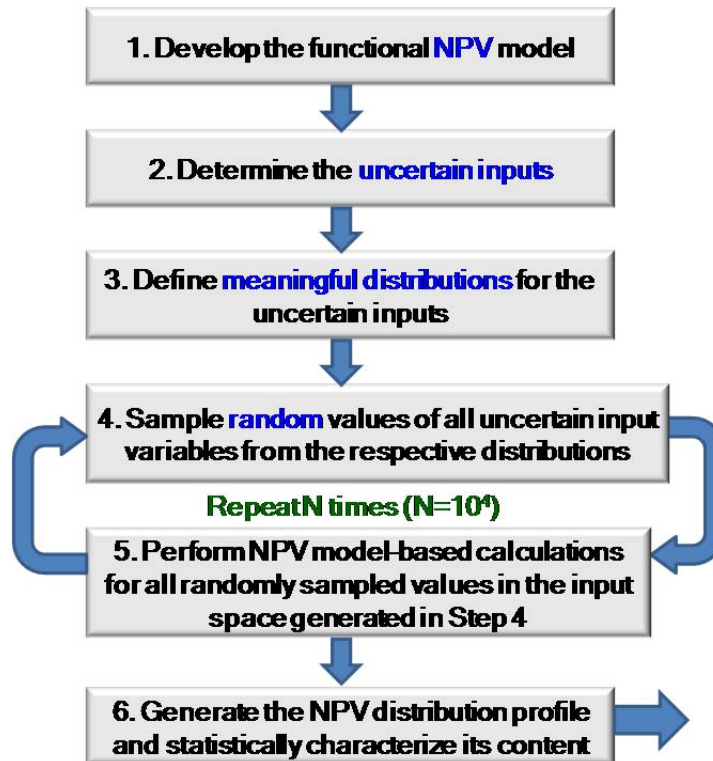
$$F(E(x)) \neq E(F(x)) \quad (5)$$

where  $x$  is the uncertain (random) variable(s) (in our case the NPV model's input variables),  $F$  is a nonlinear performance map/function (in our case the NPV), and  $E$  is the expected or the average value (Benninga 2006; Brealey and Myers 1996). Please notice that the NPV model introduces nonlinear terms such as annual income (Net power output  $\times$  Capacity Factor  $\times$  Electricity selling price), real discount rate ( $[1+\text{nominal discount rate}]/[1+\text{inflation rate}]-1$ ), etc. Equation (5) represents the mathematical statement of the *flaw of averages*, which states that the project value or performance indicator evaluated at average conditions is not the average project value or performance in the presence of irreducible uncertainty, which has to be explicitly taken into account. As pointed out earlier, this task can be practically accomplished through the integration of Monte Carlo techniques into the more traditional NPV model of economic assessment (Savage 2002).

Indeed, the study of the effect of uncertain input variables on the value of the project might be rather easily conducted by integrating standard Monte Carlo (MC) simulation methods into the above NPV model. Monte Carlo simulation uses distributions of uncertain inputs, and by propagating the uncertainty through the NPV model generates a distribution of uncertain performance or equivalently a range and frequency of various economic performance outcomes (in our case an NPV distribution profile). Thus, Monte Carlo simulation provides a very effective means of identifying, and probabilistically characterizing, the consequences and impact on project value of uncertain futures. We note with interest the application by Roques et al. (2006) of similar techniques when assessing the economic viability of new nuclear power plant construction in the face of uncertain natural gas, electricity and carbon emissions prices and with possible correlations between those prices (Roques et al. 2006).

The particular sequence of methodological steps followed is summarized in the diagram shown in Figure 2. The Monte Carlo simulation method generates thousands of possible "futures" (or "future states") by assigning random values to each input (sampled from a meaningful

distribution through which the corresponding uncertainty is quantified and probabilistically described), runs all these possible “future states” simultaneously through the NPV model (also known as uncertainty propagation through the NPV model), and finally generates the induced distribution of possible performance (project value) outcomes in the form of an NPV distribution profile while graphically summarizing all pertinent statistics (Savage 2003). The software XLSim was used for the Monte Carlo simulations conducted in the present study.



*Figure 2. The Monte Carlo simulation procedure*

The most significant uncertain inputs to the NPV model considered are the following:

1. Plant capacity factor
2. Pd price
3. Au price
4. Support price
5. Membrane lifetime
6. Initial CO<sub>2</sub> tax
7. The CO<sub>2</sub> tax starting year
8. CO<sub>2</sub> tax growth rate

9. Electricity selling price
10. Nominal discount rate
11. Inflation rate

Since all of the H<sub>2</sub> produced in the IGCC-MR case was assumed to be used for electricity production, an H<sub>2</sub> selling price was not included into the Monte Carlo simulation. Clearly, once a hydrogen economy has been established, there may be occasions when it is advantageous to sell stored hydrogen rather than to use it for electricity generation. Such options can, to a first approximation, only increase the economic case for IGCC-MR systems. Such a business model also represents a good target for future economic assessment. Please also note that the plant capacity factor plays an important role in the uncertainty analysis. The IGCC plant with an integrated Pd-based membrane reactor technology option has never been demonstrated at commercial scale. Thus, the lack of operating experience introduces significant technology risk and uncertainty concerning the plant capacity factor.

**Table 5. Probability distributions associated with the various uncertain inputs**

Parameter↓	Distribution		
	Minimum	Most Likely	Maximum
CO <sub>2</sub> tax (\$/ t CO <sub>2</sub> ), TD	0	25	75
CO <sub>2</sub> tax growth rate, TD	0	3	8
Nominal discount rate, TD	6	7	9
Plant capacity factor [%], TD	70	75	85
Inflation rate [%], RH	1979-2011 Historical data, U.S. Dep. of Lab. Bureau		
Electricity selling price [cents/kWh], RH	1979-2011 Historical data, U.S. Energy Inf. Adm.		
Pd price [\$/g], RH	1979-2011 Historical data, www.kitco.com		
Au price [\$/g], RH	1979-2011 Historical data, www.kitco.com		
Support price [\$/cm <sup>2</sup> for lab scale], TD	2.3	5	7.5
Membrane life time [years], TD	1	3	5

\*TD: triangular distribution, RH: resample historical data

Finally, it should be pointed out that uncertainty is not only associated with economic parameters, the regulatory environment and technology risks but also the safe operation of the plant which could affect its overall economic performance and profitability quite drastically in the case of an accident. Therefore, a major challenge is to explicitly incorporate and carefully implement process safety strategies and safe process system design (Pasman 2000) and furthermore demonstrating that such investments in process safety do make economic sense in an uncertain world in which low-probability/major-consequences events can occur. As noted earlier, significant progress in process safety has also introduced a sense of complacency by reinforcing rationalizations that accidents can be averted and their consequences minimized. This is reinforced by irrational resistance against sustained investments in process safety based upon a myopic perception of high costs (Pasman 2000).

Within the context of the present study, a potential hydrogen explosion or a leakage episode involving poisonous syngas components can be considered as examples of pertinent safety problems. It should be pointed out that the three most important characteristics of hydrogen being produced and separated in a process system such as the one under consideration are: 1) its diffusivity in air is very high, 2) the ignition energy of a hydrogen/oxygen mixture is very low, and 3) the associated ignition range is quite broad. Please notice that hydrogen quickly disperses into the air and, thus this feature could be thought as an inherent safety element if the initiation of an ignition episode is prevented. However, the frictional electric potential of human skin or a micro-arc from an electric switch might suffice to ignite the hydrogen/oxygen mixture (Peschka et al. 1992). Furthermore, in the case of an overpressure incident, if the required safety measures are not properly implemented, a reactor rupture may lead to an explosion. Moreover, the leakage of a poisonous gas such as CO and H<sub>2</sub>S into the environment could generate significant hazards to personnel, the population nearby and/or ecosystem functions.

Pd/alloy-based membrane reactors integrated into IGCC plants have not yet been demonstrated at a commercial scale. Any process safety and economic performance assessment must be driven by theoretical, yet reasonable, estimates. Methodologically one should



acknowledge the various uncertainties in an explicitly manner. The present research study conforms to such a methodological approach using the above framework of analysis and in a similar spirit to the one presented in work by Pasman (Pasman 2000) where costs involved in improving safety are assessed against the benefits of preventing or reducing the likelihood of occurrence of accidents. In other words, the primary aim is to examine the validity of the thesis that process safety investment strategies in an uncertain world where accidents do happen, might actually enhance overall economic performance.

## 5. Main Results and Discussion

The cost analysis of the industrial scale Pd/alloy-based composite membrane reactor was conducted by considering capital-investment costs, manufacturing costs and general expenses. The details of the fixed-capital investment and total product cost calculations for the Pd/Au-based water gas shift membrane reactor can be found in Table 7 and Table 8 , respectively. The total product cost figure listed in Table 6 for the membrane reactor module was estimated to be \$1795/ft<sup>2</sup>, thus exceeding the published DOE cost target of \$1000/ft<sup>2</sup>. However, this DOE target cost of the membrane reactor module (DOE-Fuel Cell Program, 2011) was set for fuel cell grade H<sub>2</sub> production and therefore does not include the water-gas shift reaction-related cost items (US\$ 1500/ft<sup>2</sup> for 2006 status, US\$ 1000/ft<sup>2</sup> for 2010 and <500 US\$/ft<sup>2</sup> for 2015). In light of the above consideration and under water-gas shift reaction conditions, the cost figures listed in Table 6 are in line with the US DOE cost targets when modified to account for the aforementioned operating conditions.

**Table 6. Static membrane reactor module cost summary**

Fixed-capital investment [\$/ft <sup>2</sup> ]	705
Total capital investment [\$/ft <sup>2</sup> ]	829
Total product cost [\$/ft <sup>2</sup> ]	1795

The cost of the Pd/Au-based membranes supported on 316L PSS supports was mainly dependent on the Pd cost and thickness for this particular case. A fairly thick Pd/Au layer of  $\sim 7.4$   $\mu\text{m}$  was used in the calculations and the cost could be reduced further by decreasing the Pd/Au thickness. It should be pointed out that the support cost would be much higher than the 316L PSS supports if the Pd/Alloy-based membranes were required to be used for other applications such as steam methane reforming, in which Inconel supports are preferred. More importantly, the Pd thickness contributed the highest portion of the membrane equipment cost in Table 7 when the six-tenths-factor rule was applied to the support price (Chand Eisenman and Mott Met. Cor.2011). If the support price could not be further reduced through relatively large-scale purchases, the support cost would have a stronger effect on the equipment cost than the Pd thickness. Since components of the fixed-capital investment, such as installation and engineering & supervision, are all dependent on the equipment cost associated with the Pd/Alloy-based membranes, the higher the equipment cost of the membrane bundle the higher the resulting fixed-capital investment becomes.

In addition, membrane replacement costs were included into the variable operating costs of the IGCC plant with embedded membrane reactors and calculated by dividing the equipment cost of the membrane bundle by the life-time of the Pd-based membrane (i.e. US M\$ 22.5/ 3 years). Of course it is desirable to extend the membrane lifetime, so as to reduce replacement costs. Membrane replacement-related costs were also included into the operating and maintenance costs of the IGCC-MR plant. Finally, it should be pointed out that the membrane could be repaired through re-plating in order to increase its life time or Pd could be recovered from the membrane tube, and therefore, the cost associated with these processing steps could also explicitly be taken into account in the total product cost calculation. Please notice, that even though such a detailed cost assessment pertaining to the aforementioned membrane processing steps has not been pursued yet in the pertinent body of literature due primarily to the absence of reliable data, it does represent a meaningful future research endeavor.

**Table 7. Estimation of capital investment cost**

I. Direct Costs				
A. Equipment + installation + instrumentation + piping + electrical + insulation + painting				
1. Purchased equipment				
	Reactor	HTS catalyst	Pd Membrane	
	\$28,758.4	\$1,756,783.8	Pd Cost	\$17,346,343.1
			Au Cost	\$3,622,542.1
			Support [316L SS]	\$3,094,501.4
			Subtotal	\$24,063,386.6
	Total membrane module	\$25,848,928.8		
	2. Installation (%40 of A1.)	\$10,339,571.5		
	3. Instrumentation and controls, installed (%18 of A1.)	\$4,652,807.2		
	4. Piping installed (%45 of A1.)	\$11,632,017.9		
	5. Electrical, installed (%25 of A1.)	\$6,462,232.2		
	Equipment total	\$58,935,557.6		
II. Indirect Costs				
	A. Engineering and supervision (%17.5 of direct cost)	\$10,313,722.6		
	B. Construction expense and supervision (18% of direct cost)	\$10,608,400.4		
	C. Contingency (%10 of Fixed-capital investment)	\$11,357,536.8		
	Subtotal	\$32,279,659.7		
	III. Fixed-capital investment = direct + indirect costs	\$113,575,367.9		
	IV. Working capital ( 15% of V)	\$20,042,712.0		
	V. Total capital investment = III + IV	<b>\$133,618,079.9</b>		

*Table 8. Estimation of total product cost*

I. Manufacturing cost = direct production cost + fixed charges	
A. Direct production costs	
1. Raw materials (30% of total product cost)	\$86,725,021.4
2. Operating labor (15% of total product cost)	\$43,362,510.7
3. Direct supervisory and clerical labor (17.5% of operating labor)	\$7,588,439.4
4. Utilities (15% of total product cost)	\$43,362,510.7
5. Maintenance (6% fixed-capital in.)	\$6,814,522.1
6. Operating supplies (0.75% of fixed-capital in.)	\$851,815.3
7. Laboratory charges (15% of operating labor)	\$6,504,376.6
8. Patents and royalties (3% of total product cost)	\$8,672,502.1
Subtotal	\$203,881,698.2
B. Fixed charges	
1. Depreciation (assumed 10% of fixed-capital in.)	\$11,357,536.8
2. Local taxes (4% of fixed-capital in.)	\$4,543,014.7
3. Insurance (0.7% of fixed-capital in.)	\$795,027.6
Subtotal	\$16,695,579.1
II. General expenses = administrative + distribution & selling + R&D costs	
A. Administrative costs (4% of total product cost)	\$11,563,336.2
B. Distribution & selling costs ( 11% of total product cost)	\$31,799,174.5
C. R&D costs (5% total product cost)	\$14,454,170.2
D. Financing (interest - 8% of total capital investment)	\$10,689,446.4
Subtotal	\$68,506,127.3
III. Total product cost = manufacturing cost + general expenses	<b>\$289,083,404.6</b>

The calculated values of the fixed-capital investment and operating and maintenance costs for the IGCC-MR plant are listed in Table 9. Even though a Pd/alloy-based membrane system adds greater value to the IGCC plant in the case of CO<sub>2</sub> capture assuming a carbon price/tax, both cases with and without CO<sub>2</sub> capture were considered. Please notice that the carbon price figure considered in the present study represents an average numerical value offered by the most recent and comprehensive studies that can be found in the pertinent literature (for a comprehensive account and survey the reader is referred to: Al-Juaied, M. A., Whitmore, A., 2009). In the case of no capture, investment required for CO<sub>2</sub> compression and expenses related to transportation/sequestration were not included. In addition, fixed-capital investment values listed in Table 9 do not contain any safety related pre-investment. The NPV values were calculated by using the initial investments and O&M costs in Table 9 and by taking the revenues generated annually through electricity selling (550MW) into account. The single-point projection NPV value of the IGCC-MR without CO<sub>2</sub> capture was found to be US \$ ~0.7B higher than in the CO<sub>2</sub> capture case due to the expenses related to CO<sub>2</sub> compression and sequestration.

**Table 9. Single point NPV projection results for the IGCC-MR plant**

	Without CO <sub>2</sub> capture	With CO <sub>2</sub> capture
Fixed-capital investment [B\$]	1.51	1.57
O&M Costs [M\$]	87.9	107
Single point NPV [B\$]	0.71	0.44

Monte Carlo simulations were performed to propagate the uncertainty of the 10 value drivers given in Table 5 through the NPV model and also uncertainty associated with another random variable related to plant safety that represents an additional uncertain value driver in the NPV model considered in the present study. In particular, a potential investor in this new technology and engineering project has two options: one is to make

little, or no, effort to prevent accidents, taking a chance and simply paying for the consequences should any accident happen; the other is to behave proactively by investing in plant safety up-front and thus reducing the risk of a minor incident or a major catastrophe. One is reminded that accidents are costly due to lost working days, but also due to lost production, damage to equipment and plant, investigation time as well as liability claims (Pasman 2000; Kletz 1990).

Within the above economic assessment framework, two cases were evaluated and analyzed in this study:

1. A leak incident (minor incident) with:
  - a. No initial action at start-up
  - b. Pre-investment in plant safety
2. A major leak (catastrophe) with:
  - a. No initial action at start-up
  - b. Pre-investment in plant safety

The uncertainty related to the possible occurrence of an industrial accident was represented in our NPV-model with integrated Monte Carlo through a simple Bernoulli distribution assigned to the 11<sup>th</sup> additional uncertain value driver. The modified formula for the NPV is given in Equation (6) encompassing both the initial investment in equipment to enhance plant safety as well as the total cost incurred in case the accident occurs:

$$NPV = (C_o - C_{safety}) - (pr \times D_o)DF + PV \quad (6)$$

where  $DF = \sum_{t=1}^N \frac{1}{(1+r_t)^t}$  is the so-called unit annuity present value factor (a constant for a fixed discount rate),  $C_{safety}$  is the safety pre-investment amount,  $pr$  is the probability of occurrence of the accident (associated with a Bernoulli distribution) and  $D_o$  is the expected loss or a monetary measure of damage (total cost incurred) due to the occurrence of the accident.

The amount of investment in safety is directly proportional to the magnitude of the accident: a relatively small investment would be required to reduce the risk and the associated cost of a minor leak incident whereas the amount of investment in plant safety needs to be larger in the case of a major catastrophe for the attainment of the above objectives. Representative values of the investment in safety ( $C_{safety}$ ), probability of accidents ( $pr$ ) and expected loss ( $D_o$ ) are adopted from Pasman (2000) and listed in Table 10.

**Table 10. Decision matrix for risk reduction measures**

Options of risk reduction		$C_{safety}$ [k\$]	$pr$ [year <sup>-1</sup> ]	$D_o$ [k\$]
Minor leak incident	No measure	0	10 <sup>-1</sup>	10 <sup>6</sup>
	Safety investment action	6.5×10 <sup>4</sup>	10 <sup>-4</sup>	10 <sup>3</sup>
Major leak catastrophe	No measure	0	10 <sup>-3</sup>	10 <sup>8</sup>
	Safety investment action	13×10 <sup>4</sup>	10 <sup>-6</sup>	10 <sup>6</sup>

The NPV distribution (risk-reward) profile for IGCC-MR is given in Figure 3 by considering the 10 uncertain value drivers plus the uncertainty and risk of a minor leak. Figure 3 being a graph of a cumulative distribution function allows a probabilistic/statistical characterization of the entire range of possible economic performance outcomes in the presence of uncertainty. The upper-right positive NPV zone of Figure 3 is associated with quite favorable economic outcomes due to a high plant capacity factor, low nominal discount rate, low Pd price and so on; the lower-left negative NPV zone of Figure 3 is associated with undesirable economic performance outcomes (possibly due to a low capacity factor, high nominal discount rate, high Pd price, etc). According to the NPV distribution profile in Figure 3 [a], the risk of generating a negative NPV was 62% if no measure were taken against the possibility of a minor leak. If the

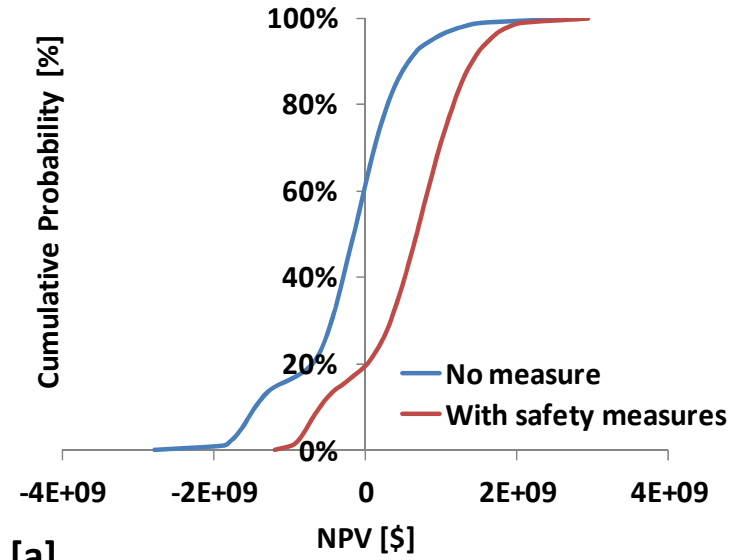
required investment were made during the construction period, the risk-reward profile could be shifted to the right and the risk of generating a negative NPV was reduced to 19%.

The risk associated with a major leak together with the other 11 uncertain value drivers was even more significant from an economic performance standpoint as shown in Figure 3 [b]. While the probability of losing money was 66% without any pre-investment in plant safety, the risk reward profile of the IGCC-MR plant was successfully shifted to the right reducing the probability of generating a negative NPV to 24% after investing in plant safety at the construction stage.

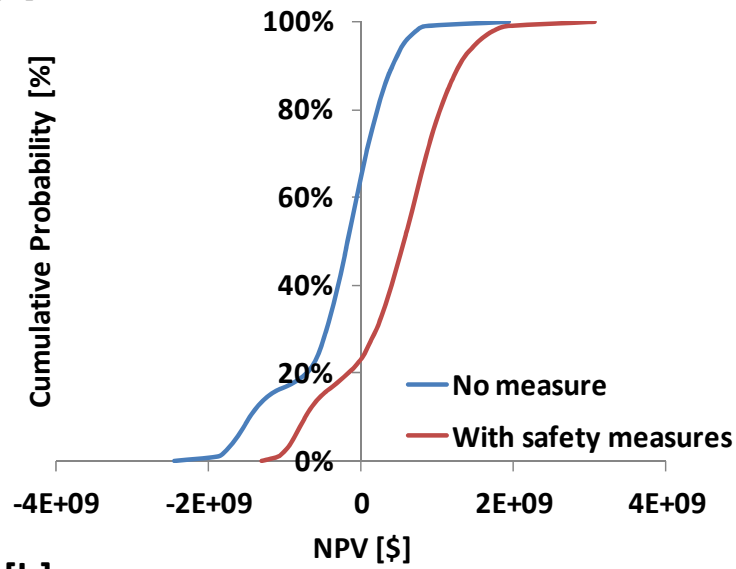
Interesting findings and results for both case studies can be found in both Figure 3 [a] and [b] as well as those tabulated in Table 11. Regardless of the size of the leak, the project has a significant chance of exhibiting a negative expected NPV values if no safety measures are taken for the membrane reactor technology in an IGCC plant with a net power output of 550MWe. Under the scenarios considered in the present study, the NPV (or cost-reward) profiles of the cases without safety measures/pre-investment are obviously less attractive than the ones in cases with safety pre- investment as evidenced in Figure 3. In particular, the associated process safety investments could economically bring the IGCC-MR technology option to a more profitable zone with representative ENPV values of US \$0.6B and \$0.4B for the cases of minor and major leak risk respectively.

The expected value of NPV (ENPV) generated through the Monte Carlo simulations in the case of pre-investment in process safety and risk reduction measures was considerably higher than the ENPV without any action against hazards given the conditions that were considered in this study. Therefore, the analysis conducted suggests that under the above conditions, an initial investment in plant safety at the initial stage of the IGCC power plant improves the project's economic value. Summarizing, the two case studies considered offer credence to the thesis that in an uncertain world where accidents do happen, market conditions change and the regulatory environment evolves, process safety investment strategies are economically rational, even before any consideration given to environmental damage or harm to human by taking into account only equipment damage and interruption of business/lost production cost on the plant level (Pasman 2000).





[a]



[b]

Figure 3. NPV distribution profile considering 10 uncertain inputs plus the risk of a [a] minor and [b] major leak

**Table 11. The expected NPV (ENPV) results for all 4 cases**

	Minor Leak Incident		Major Leak Catastrophe	
	No Measure Taken	With Pre-Investment in Process Safety	No Measure Taken	With Pre-Investment in Process Safety
Average (ENPV) [\$US]	-2.37E+08	5.79E+08	-3.11E+08	4.44E+08
Max[\$US]	2.72E+09	2.94E+09	1.93E+09	3.06E+09
Min[\$US]	-2.79E+09	-1.21E+09	-2.46E+09	-1.32E+09

## 6. Concluding Remarks

An economic performance assessment of inherently safe Pd/alloy-based membrane reactor technology option integrated into IGCC power plants in the presence of market and regulatory uncertainty as well as technology and safety risks has been performed. The membrane reactor dimensions and number of membrane tubes were determined with the aid of a two-dimensional isothermal model and used in the pertinent cost analysis. Fixed capital investment along with operating and maintenance costs for the Pd-based membrane reactor module were calculated using reaction and feed conditions compatible with the operation of a GEE coal gasifier. Furthermore, total product cost (including manufacturing costs and general expenses) for the Pd/Au-based water-gas shift membrane reactor module was estimated to be \$1795/ft<sup>2</sup> based on current economic conditions. The total product cost of \$1795/ft<sup>2</sup> for the membrane reactor module was found to be competitive with the stringent US DOE 2010 cost target level set for only fuel cell grade H<sub>2</sub> production, when appropriately modified to account for water-gas shift reaction-related expenses.

A functional NPV-based model was developed to evaluate investment opportunities for the production of electricity through coal gasification technology within the IGCC-MR context and in the presence of regulatory action on carbon emissions. Traditional single-point NPV-based economic assessment relying on average values for all value drivers (nominal discount rate, plant capacity factor, Pd price, etc.), showed that, under the conditions and uncertainties considered, the IGCC-MR technology option could become

economically viable. In particular, single-point NPV values were calculated without and with CO<sub>2</sub> taxes and found to be US\$ 0.71 and US\$ 0.44 billion, respectively.

Moreover, Monte Carlo simulation methods were integrated into the above functional NPV- model in order to explicitly take into account the main uncertainty drivers and study their effect on process economic performance. The uncertainty in key inputs to the NPV- model (plant capacity factor, nominal discount rate, inflation rate, CO<sub>2</sub> tax, CO<sub>2</sub> tax growth rate, electricity selling price, Pd price, support price and membrane lifetime) and also the uncertainty/risk in plant safety (realized as an additional random variable that follows a simple Bernoulli distribution to describe the possibility of the occurrence of an industrial accident) were explicitly considered for an IGCC-MR plant. A comparatively more attractive NPV distribution profile was obtained when concrete safety risk-reducing measures were taken into account through pre-investment in process safety.

The role of economic appraisal of planned safety measures differs from jurisdiction to jurisdiction. The United States has a long tradition of prescriptive safety regulation. In this framework the safety regulator closely specifies the measures to be taken. A project developer/operator can take comfort that they are compliant with safety regulation if they have implemented the specific given requirements. Such requirements are only weakly developed with economic considerations in mind. By contrast, following the Robens Report of 1972 the United Kingdom adopted a goal-based approach to engineering safety (Robens, 1972). A key step in this process was the Health and Safety at Work Act 1974. In this framework the safety regulator specifies the goal to be achieved, but leaves to the discretion of the facility developer (or operator) the means that will be adopted to ensure compliance. Sometimes the goal to be achieved relates to the legally grounded notion of *As Low As Reasonably Practicable* (ALARP). A key benefit of the UK approach is that, if for any reason (such as new emergent knowledge from accidents elsewhere) the set of reasonably practicable measures changes, then instantly the operator must adopt new procedures to maintain legal compliance. In the UK such compliance is ensured via the criminal code. Another benefit of the UK approach is that it leaves the chosen actions to the discretion of those most familiar with the business process – the business operator. The business operator is well used to making economically driven choices between options. Goal-based safety regulation extends this

principle into the engineering safety domain. The work reported here provides further insights for those concerned with economically-efficient prioritization of actions. The economic assessment of safety investment discussed in this work therefore has the potential to influence theory and practice concerning safety regulation. Some legal frameworks are more amenable to such approaches than others.

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## REFERENCES

Eisenman, C. and Mott Met. Corporation.[Chand Eisenman, Burlington, CT and Mott Metallurgical Corporation, Farmington, CT].

Fuel cell technologies program multi-year research, development and demonstration plan. Accessed year: 2011. <http://www1.eere.energy.gov/hydrogenandfuelcells/mypp/>

Hydrogen from coal R&D plan. Accessed year: 2011. [http://fossil.energy.gov/programs/fuels/hydrogen/Hydrogen from Coal R%26D.html](http://fossil.energy.gov/programs/fuels/hydrogen/Hydrogen_from_Coal_R%26D.html)

Pulverized coal power. Accessed year: 2011. <http://www.wri.org/publication/content/10338>

Marshall & swift cost indexes. Accessed year: 2010. <http://www.che-digital.com/che/201001?pg=90#pg90>

Adams, T. A., Barton, P. I. , 2009. A dynamic two-dimensional heterogeneous model for water gas shift reactors. *International Journal of Hydrogen Energy* 34 (21): 8877-91.

Amelio, M., Morrone, P., Gallucci, F., Basile, A. , 2007. Integrated gasification gas combined cycle plant with membrane reactors: Technological and economical analysis. *Energy Conversion and Management* 48 (10): 2680-93.

Armor, J. N. , 1998. Applications of catalytic inorganic membrane reactors to refinery products. *J.Membr.Sci.* 147 : 217,217-223.

Augustine, A. S., Ma, Y. H., Kazantzis, N. K. , 2011. High pressure palladium membrane reactor for the high temperature water–gas shift reaction. *International Journal of Hydrogen Energy* 36 (9): 5350.

Ayturk, M. E., Kazantzis, N. K., and Ma, Y. H. , 2009. Modeling and performance assessment of pd- and Pd/Au-based catalytic membrane reactors for hydrogen production. *Energy & Environmental Science* 2 : 430,430-438.

Ayturk, M. E., Mardilovich, I. P., Engwall, E. E., Ma, Y. H. , 2006. Synthesis of composite pd-porous stainless steel (PSS) membranes with a Pd/Ag intermetallic diffusion barrier. *Journal of Membrane Science* 285 (1-2): 385-94.

Basile, A., Chiappetta, G., Tosti, S., Violante, V. , 2001. Experimental and simulation of both pd and Pd/Ag for a water gas shift membrane reactor. *Separation and Purification Technology* 25 (1-3): 549-71.

- Basile, A., Drioli, E., Santelli, F., Violante, V., Capannelli, G., Vitulli, G. , 1996. A study on catalytic membrane reactors for water gas shift reaction. *Gas Separation & Purification* 10 (1): 53-61.
- Beér, J. M. , 2000. Combustion technology developments in power generation in response to environmental challenges. *Progress in Energy and Combustion Science* 26 (4-6): 301-27.
- Benninga, S. , 2006. *Principles of finance with excel*. New York: Oxford University Press.
- Bohm, M. C., Herzog, H. J., Parsons, J. E., Sekar, R. C. , 2007. Capture-ready coal plants— Options, technologies and economics. *International Journal of Greenhouse Gas Control* 1 (1): 113-20.
- Bohm, M. C. , 2006. Capture-ready power plants - options, technologies and economics. Massachusetts Institute of Technology.
- Bose, A. C. , 2009. Chapter 13. composite pd and Pd/Alloy membranes. In *Inorganic membranes for energy and environmental applications*. NY, USA: Springer Science+Business Media.
- Bracht, M., Alderliesten, P. T., Kloster, R., Pruschek, R., Haupt, G., Xue, E., Ross, J. R. H., Koukou, M. K., Papayannakos, N. , 1997. Water gas shift membrane reactor for CO<sub>2</sub> control in IGCC systems: Techno-economic feasibility study. *Energy Conversion and Management* 38 (Supplement 1): S159-64.
- Brealey, R. A., Myers, S. C. , 1996. *Principles of corporate finance*. 5th edition ed. USA: The McGraw-Hill Companies, Inc.
- Brunetti, A., Barbieri, G., Drioli, E. , 2009. Pd-based membrane reactor for syngas upgrading. *Energy & Fuels* 23 (10): 5073-6.
- Center for Chemical Process Safety/AIChE. , 1993. Chapter 14. pressure relief systems. In *Guidelines for engineering design for process safety*. Center for Chemical Process Safety/AIChE.
- Chen, C., Ma, Y. H., "The Effect of Au Content in Composite Pd/Au Membranes Prepared by Galvanic Displacement on Hydrogen Permeance and H<sub>2</sub>S Resistance " (Presentation, , 2010a).
- . , 2010b. The effect of H<sub>2</sub>S on the performance of pd and Pd/Au composite membrane. *Journal of Membrane Science* 362 (1-2): 535-44.
- . , 2010c. The effect of H<sub>2</sub>S on the performance of pd and Pd/Au composite membrane. *Journal of Membrane Science* 362 (1-2): 535-44.

- Chiappetta, G., Clarizia, G., Drioli, E. , 2006. Analysis of safety aspects in a membrane reactor. *Desalination* 193 (1-3): 267-79.
- Criscuoli, A., Basile, A., Drioli, E. , 2000. An analysis of the performance of membrane reactors for the water-gas shift reaction using gas feed mixtures. *Catalysis Today* 56 (1-3): 53-64.
- Criscuoli, A., Basile, A., Drioli, E., Loiacono, O. , 2001. An economic feasibility study for water gas shift membrane reactor. *Journal of Membrane Science* 181 (1): 21-7.
- De Falco, M., Di Paola, L., Marrelli, L., Nardella, P. , 2007. Simulation of large-scale membrane reformers by a two-dimensional model. *Chemical Engineering Journal* 128 (2-3): 115-25.
- Dijkstra, J. W., Pieterse, J. A. Z., Li, H., Boon, J., van Delft, Y. C., Raju, G., Peppink, G., van den Brink, R. W., Jansen, D. , 2011. Development of membrane reactor technology for power production with pre-combustion CO<sub>2</sub> capture. *Energy Procedia* 4 : 715-22.
- Dolan, M. D., Donelson, R., Dave, N. C. , 2010. Performance and economics of a pd-based planar WGS membrane reactor for coal gasification. *International Journal of Hydrogen Energy* 35 (20): 10994-1003.
- Fogler, H. S. , 1999. *Elements of chemical reaction engineering*. 3rd Edition ed. New Jersey: Printice-Hall International, Inc.
- Gallucci, F., De Falco, M., Tosti, S., Marrelli, L., Basile, A. , 2007. The effect of the hydrogen flux pressure and temperature dependence factors on the membrane reactor performances. *International Journal of Hydrogen Energy* 32 (16): 4052-8.
- Mitretek technical paper, Gray, D., Tomlinson, G. , 2002. *Hydrogen from coal*. MTR 2002-31.
- Hara, S., Sakaki, K., Itoh, N. , 1999. Decline in hydrogen permeation due to concentration polarization and CO hindrance in a palladium membrane reactor. *Industrial & Engineering Chemistry Research* 38 (12): 4913-8.
- DOE/NETL, Haslbeck, J. L., Kuehn, N. J., Lewis, E. G., Pinkerton, L. L., Simpson, J., Turner, M. J., Varghese, E., Woods, M. C. , 2010. *Cost and performance baseline for fossil energy plants volume 1: Bituminous coal and natural gas to electricity*. 2011.
- Hla, S. S., Park, D., Duffy, G. J., Edwards, J. H., Roberts, D. G., Ilyushechkin, A., Morpeth, L. D., Nguyen, T. , 2009. Kinetics of high-temperature water-gas shift reaction over two iron-based commercial catalysts using simulated coal-derived syngases. *Chemical Engineering Journal* 146 (1): 148-54.
- Kletz, T. A. , 1990. *Improving chemical engineering practicesnew , A look at old myths of the chemical industry*. Second Edition ed. USA: Hemisphere Publishing Corporation.

- Koc, R., Kazantzis, N. K., Ma, Y. H. , 2011. Process safety aspects in water-gas-shift (WGS) membrane reactors used for pure hydrogen production. *Journal of Loss Prevention in the Process Industries* 24 (6): 852-69.
- Kulprathipanja, A., Alptekin, G. O., Falconer, J. L., Way, J. D. , 2005. Pd and Pd-Cu membranes: Inhibition of H<sub>2</sub> permeation by H<sub>2</sub>S. *Journal of Membrane Science* 254 (1-2): 49-62.
- Mary Kay O'Connor Process Safety Center, Chemical Engineering Dep. Mannan, M. S. White paper on *challenges in implementing inherent safety principles in new and existing chemical processes*. [Accessed Year 2011]. Available from <http://psc.tamu.edu/library/center-publications/white-papers-and-position-statements>.
- Mardilovich, P. P., She, Y., and Ma, Y. H. , 1997. Defect-free palladium membranes on porous stainless-steel support. *AIChE Journal* 44 (2): 16 Apr 2004, 310 - 322.
- Mardilovich, I. P., Engwall, E., Ma, Y. H. , 2002. Dependence of hydrogen flux on the pore size and plating surface topology of asymmetric Pd-porous stainless steel membranes. *Desalination* 144 (1-3): 85-9.
- Maurstad, O. An overview of coal based integrated gasification combined cycle (IGCC) technology. [Accessed Year 2005]. Available from [http://sequestration.mit.edu/pdf/LFEE\\_2005-002\\_WP.pdf](http://sequestration.mit.edu/pdf/LFEE_2005-002_WP.pdf).
- Nguyen, T. H., Mori, S., Suzuki, M. , 2009. Hydrogen permeance and the effect of H<sub>2</sub>O and CO on the permeability of Pd<sub>0.75</sub>Ag<sub>0.25</sub> membranes under gas-driven permeation and plasma-driven permeation. *Chemical Engineering Journal* 155 (1-2): 55-61.
- Pasman, H. J. , 2000. Risk informed resource allocation policy: Safety can save costs. *Journal of Hazardous Materials* 71 : 375-394.
- Peschka, w., Wilhelm, E. A., Wilhelm, U. , 1992. *Liquid hydrogen: Fuel of the future*. Verlag: Springer.
- Peters, M. S., Timmerhaus, K. D. , 1991. *Plant design and economics for chemical engineers*. 4th edition ed. USA: McGraw Hill, Inc.
- Pomerantz, N., Ma, Y. H. , 2009. Effect of H<sub>2</sub>S on the performance and long-term stability of Pd/Cu membranes. *Industrial & Engineering Chemistry Research* 48 (8): 4030-9.
- Pomerantz, N., Meyer, N., Chen, C., Ma, Y. H. , 2010. Sulfur poisoning of high permeance Pd/Cu alloy membranes for H<sub>2</sub> separation from coal gas. *NAMS Annual Meeting, Washington, D.C. 2010*.



- Ratafia-Brown, J. A., Manfredo, L. M., Hoffmann, J. W., Ramezan, M. and Stiegel, G.J. , 2002. An environmental assessment of IGCC power systems. *Nineteenth Annual Pittsburgh Coal Conference, September 23 – 27*.
- Rezvani, S., Huang, Y., McIlveen-Wright, D., Hewitt, N., Mondol, J. D. , 2009. Comparative assessment of coal fired IGCC systems with CO<sub>2</sub> capture using physical absorption, membrane reactors and chemical looping. *Fuel* 88 (12): 2463-72.
- Robens A., Committee on Safety and Health at Work, Cmnd 5034, Her Majesty's Stationery Office 1972
- Roques, F. A., Newbery, D. M., Nuttall, W. J., de Neufville, R., Connors, S. , 2006. Nuclear power: A hedge against uncertain gas and carbon prices. *Energy Journal* 27 : 1-24.
- Savage, S. L. , 2003. *Decision making with insight*. USA: Brooks/Cole.
- , 2002. The flaw of averages. *Harvard Business Review*: 20-22.
- Shu, J., Grandjean, B. P. A., Neste, A. V., Kaliaguine, S. , 1991. Catalytic palladium-based membrane reactors: A review. *The can. J. Chem. Eng.* 69 : 1036-60.
- Stankiewicz, A. I., Moulijn, J. A. , 2004. *Re-engineering the chemical processing plant process intensification* New York : M. Dekker.
- Takht Ravanchi, M., Kaghazchi, T., Kargari, A. , 2009. Application of membrane separation processes in petrochemical industry: A review. *Desalination* 235 (1-3): 199-244.
- Tarun, C. B., Croiset, E., Douglas, P. L., Gupta, M., Chowdhury, M. H. M. , 2007. Techno-economic study of CO<sub>2</sub> capture from natural gas based hydrogen plants. *International Journal of Greenhouse Gas Control* 1 (1): 55-61.
- Tosti, S., Basile, A., Chiappetta, G., Rizzello, C., Violante, V. , 2003. Pd–Ag membrane reactors for water gas shift reaction. *Chemical Engineering Journal* 93 (1): 23-30.
- Uemiya, S., Sato, N., Ando, H., Kikuchi, E. , 1991. The water gas shift reaction assisted by a palladium membrane reactor. *Industrial & Engineering Chemistry Research* 30 (3): 585-9.
- Veziroglu, T. N., Barbir, F. , 1998. *HYDROGEN ENERGY TECHNOLOGIES. EMERGING TECHNOLOGY SERIES*. Vienna: UNIDO.
- Ward, T. L., Dao, T. , 1999. Model of hydrogen permeation behavior in palladium membranes. *Journal of Membrane Science* 153 (2): 211-31.

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Way, J. D., Lusk, M., Thoen, P. , 2008. *Sulfur-resistant composite metal membranes*. Patent US Patent 20080038567, filed 2008.

Wen, C. Y., Fan, L. T. , 1975. *Models for flow systems and chemical reactors*. New York: Dekker.

## Appendix

### The structure of a two-dimensional membrane reactor model:

The mass and momentum balance equations in the radial and axial directions were simulated to characterize membrane reactor performance at isothermal and steady state conditions. In addition, axial dispersion and radial convection was neglected, ideal selectivity of the membrane was assumed and neither vacuum nor sweep was used in the permeate side. The detailed system of partial differential equations and associated boundary conditions of membrane reactor model are reported below.

- (i) Mass balance equation for all gas phase species in the reaction side (shell)  $i=CO, H_2O, CO_2, H_2$ :

$$\frac{\partial(u.C_i)}{\partial y} - \frac{D_{i-mix}^e}{u} \left[ \frac{\partial^2(u.C_i)}{\partial x^2} + \frac{1}{x} \frac{\partial(u.C_i)}{\partial x} \right] + \rho_{Bulk,Cat} \cdot r_i = 0 \text{ (Fogler 1999)} \quad (1)$$

- (ii) Momentum balance in the reaction side (shell):

$$\frac{dP_{Total}^{Shell}}{dy} = \left( 150 + 1.75 \frac{Re}{(1-\varepsilon)} \right) \cdot \frac{G \cdot \eta_{mix} \cdot (1-\varepsilon)^2}{\rho_{mix,g} \cdot d_{particle}^2 \cdot \varepsilon^3} \quad (2)$$

$$G = \rho_{mix,g} \cdot u \quad (3)$$

$$r_{CO} = 10^{2.845 \pm 0.03} \cdot e^{\left( \frac{-111 \pm 2.63}{R_g \cdot T} \right)} \cdot P_{CO}^{1.0 \pm 0.031} \cdot P_{CO_2}^{-0.36 \pm 0.043} \cdot P_{H_2}^{1-0.09 \pm 0.007} \cdot \beta \text{ (Hla et al. 2009)} \quad (4)$$

The linear gas velocity along the length of the reactor was evaluated by using Equations (5) through (7) as shown below:

$$U_i(x, y) = u(y) \cdot C_i(x, y) = \frac{\bar{V}_{Total}(y)}{A_c} \cdot C_i(x, y) = \frac{F_i^{Shell}(x, y)}{A_c} \quad (5)$$

$$U_{Total}(x, y) = \sum_{j=1}^i U_j(x, y) \quad (6)$$

$$u(y) = \frac{\bar{V}_{Total}(y)}{A_{cr}} = \frac{U_{Total}(x, y) \cdot R \cdot T}{P_{Total}^{Shell}} \quad (7)$$

Boundary condition 1 (BC1), at the reactor inlet for  $i=CO, H_2O, CO_2, H_2, y = 0$ :

$$u \cdot C_i|_{y=0} = u^0 C_i^0 \quad (8)$$

Boundary condition 2 (BC2), at the shell casing surface for  $i=CO, H_2O, CO_2, H_2, x = x_2$ :

$$\partial(u \cdot C_i)/\partial x|_{x=x_2} = 0 \quad (9)$$

Boundary condition 3 (BC3), at the membrane surface for  $i=CO, H_2O, CO_2$  except  $H_2, x = x_1$ :

$$\partial(u \cdot C_i)/\partial x|_{x=x_1} = 0 \quad (10)$$

Boundary condition 4 (BC4), at the membrane surface for  $H_2, x = x_1$ :

$$\frac{d_{particle}}{Pe} \frac{\partial(u \cdot C_{H_2})}{\partial x} = J_{H_2} \quad (11)$$

A finite difference method was used to numerically solve the above system of partial differential equations. In particular, a forward difference method for the axial direction (y) and a central difference one for the radial direction (x) were used and the full system of equations was simulated using MATLAB.

The definitions of the dimensionless numbers used in the above membrane reactor model are as follows:

$$Peclet \ Number \ (Pe) = 8.8 \left[ 2 - \left( 1 - \frac{2 \cdot d_{particle}}{x_2 - x_1} \right)^2 \right] \text{ if } Re \geq 1000 \text{ (De Falco et al. 2007)} \quad (12)$$

$$\frac{1}{Peclet \ Number \ (Pe)} = \frac{0.4}{(Re \cdot Sc)^{0.8}} + \frac{0.09}{\{1 + 10/(Re \cdot Sc)\}} \text{ if } Re \leq 1000 \text{ (Wen and Fan 1975)} \quad (13)$$

$$Reynolds \ Number \ (Re) = \frac{d_{particle} \cdot u \cdot \rho_{mix,g}}{\eta_{mix,g}} \quad (14)$$

$$Schmidt \ Number \ (Sc) = \frac{\eta_{mix,g}}{\rho_{mix,g} \cdot D_{i-mix}^e} \quad (15)$$

$$D_{i-mix}^e = \frac{1 - f_i}{\sum_{j \neq i} f_j / D_{ij}^e} \text{ (Adams and Barton 2009)} \quad (16)$$

#### Estimation of gas mixture diffusivity and viscosity values:

All of the constants and equations used in order to estimate the diffusivity and viscosity values of the gas mixture were adopted from "The Properties of Gases & Liquids" { Reid, R.C. Copyright 1987;}.

Estimation of single gas viscosity:

$$\eta = 40.785 \frac{F_c(MW.T)^{1/2}}{V_c^{2/3} \Omega_v} \quad (17)$$

$$F_c = 1 - 0.2756\omega + 0.059035\mu_r^4 + \kappa \quad (18)$$

$$\kappa = 0.0682 + 0.2767[(17)(\text{number of } -OH \text{ groups})/MW] \quad (19)$$

$$\mu_r = 131.3 \frac{\mu}{(V_c T_c)^{1/2}} \quad (20)$$

Collision integral:

$$\Omega_v = [a_1(T^*)^{-b_1}] + c_1[\exp(-d_1 T^*)] + e_1[\exp(-f_1 T^*)] \quad (21)$$

$$T^* = 1.2593T_r \text{ and } T_r = T/T_c \quad (22)$$

$$a_1 = 1.16145, b_1=0.14874, c_1=0.52487, d_1=0.77320, e_1=2.16178, \text{ and } f_1=2.43787$$

Estimation of the viscosities of gas mixtures:

$$\eta_{mix} = \sum_{i=1}^n \frac{f_i \eta_i}{\sum_{j=1}^n f_j \phi_{ij}} \quad (23)$$

$$\phi_{ij} = \frac{[1 + (\eta_i/\eta_j)^{1/2} (MW_j/MW_i)^{1/4}]^2}{[8(1 + MW_i/MW_j)]^{1/2}} \quad (24)$$

Diffusion coefficients for binary gas systems:

$$D_{ij} = \frac{0.00266 T^{3/2}}{P \cdot MW_{ij}^{1/2} \cdot \sigma_{ij}^2 \cdot \Omega_D} \quad (25)$$

$$MW_{ij} = 2[(1/MW_i) + (1/MW_j)]^{-1} \quad (26)$$

Diffusion collision integral ( $\Omega_D$ ):

$$\Omega_D = \frac{a_2}{(T_D^*)^{b_2}} + \frac{c_2}{\exp(d_2 T_D^*)} + \frac{e_2}{\exp(f_2 T_D^*)} + \frac{g_2}{\exp(h_2 T_D^*)} \quad (27)$$

$$T_D^* = kT/\epsilon_{ij} \quad (28)$$

$$\epsilon_{ij} = (\epsilon_i \epsilon_j)^{1/2} \quad (29)$$

$$\sigma_{ij} = \frac{\sigma_i + \sigma_j}{2} \quad (30)$$

$a_2 = 1.06036, b_2=0.15610, c_2=0.19300, d_2=0.47635, e_2=1.03587, f_2=1.52996, g_2=1.76474,$   
and  $h_2=3.89411.$

For polar gases (CO and H<sub>2</sub>O):

$$\Omega_{D,p} = \Omega_D + \frac{0.19 \delta_{ij}^2}{T_D^*} \quad (31)$$

$$\delta = \frac{1940 \mu_p^2}{V_b T_b} \quad (32)$$

$$V_b = 0.285 V_c^{1.048} \quad (33)$$

$$\frac{\epsilon}{k} = 1.18 (1 + 1.3 \delta^2) T_b \quad (34)$$

$$\sigma = \left( \frac{1.585 V_b}{1 + 1.3 \delta^2} \right)^{1/3} \quad (35)$$

$$\delta_{ij} = (\delta_i \delta_j)^{1/2} \quad (36)$$

$$\frac{\epsilon_{ij}}{k} = \left( \frac{\epsilon_i \epsilon_j}{k k} \right)^{1/2} \quad (37)$$

$$\sigma_{ij} = (\sigma_i \sigma_j)^{1/2} \quad (38)$$

Effective binary diffusion coefficient:

$$D_{ij}^e = D_{ij} \frac{\epsilon}{\tau} \quad (39)$$

**Table 12. Property data for the viscosity and diffusion coefficient calculations**

Species→	CO	H <sub>2</sub> O	CO <sub>2</sub>	H <sub>2</sub>
Parameter↓				
$\sigma$	3.69	2.641	3.941	2.827
$\epsilon/k$	91.7	809.1	195.2	59.7
$T_b$	81.7	373.2	-	20.4
$T_c$	132.9	647.3	304.1	33.2
$P_c$	35	221.2	73.8	13
$V_c$	93.2	57.1	93.9	65.1
$\kappa$	0.0682	0.076	0.0682	0.0682
$\mu$	0.1	1.8	0	0
$\omega$	0.066	0.344	0.239	-0.218

## Nomenclature

- $A$  : area ( $\text{m}^2$ )
- $C$  : concentration ( $\text{mol}/\text{m}^3$ )
- $C_t$  : cash flow in year  $t$
- $C_0$  : initial capital investment
- $D$  : diffusion coefficient ( $\text{m}^2/\text{s}$ ),  $D_{i-mix}^e$  : effective diffusion coefficient of the  $i$ th species in the gas mixture,  $D_{ij}^e$  : effective binary diffusion coefficient,  $D_{ij}$  : binary diffusion coefficient
- $d_{particle}$  : particle diameter (m)
- $E_p$  : activation energy for  $\text{H}_2$  permeation (J/mol)
- $F$  : molar flow rate (mol/s)
- $f$  : mole fraction
- $G$  : mass specific gas flow rate ( $\text{kg}/[\text{m}^2.\text{s}]$ )
- ID : inside diameter (m)
- $J$  : flux (mol / [ $\text{m}^2. \text{s}$ ])
- $k$  : Boltzmann's constant,  $1.3805 \times 10^{-23}$  J/K
- $l$  : Pd thickness of the membrane ( $\mu\text{m}$ )
- MW : molecular weight
- $n$  : number of species
- $N$  : total plant life time (year)
- NPV : net present value
- OD : outside diameter (m)
- $P$  : pressure (Pa)
- $Pe$  : Peclet number

- PV : present value
- $r$  : reaction rate (mol / [kg cat. s])
- $r_t$  : real discount rate in year t (%)
- R : gas universal constant (m<sup>3</sup>.Pa/[mol.K])
- Re : Reynolds number
- $R_{H_2}$  : hydrogen recovery (%)
- Sc : Schmidt number
- $Q$  : permeability of the membrane (mol H<sub>2</sub>.μm/ [m<sup>2</sup>. s. atm<sup>0.5</sup>])
- $Q_o$  : permeability constant of the membrane (mol H<sub>2</sub>.μm/ [m<sup>2</sup>. s. atm<sup>0.5</sup>])
- T : temperature (K)
- $T_b$  : normal boiling point (@ 1 atm), (K)
- $T_c$  : critical temperature (K)
- $T_r$  : reduced temperature
- $u$  : gas velocity (m/s)
- $\bar{V}$  : volumetric flow rate (m<sup>3</sup>/s)
- $V_b$  : liquid molar volume at the normal boiling point, cm<sup>3</sup>/mol
- $V_c$  : critical volume, cm<sup>3</sup>/mol
- $X_{CO}$  : CO conversion (%)
- $x$  : radial coordinate (m)
- $y$  : spatial coordinate (m)

Greek symbols

- $\varepsilon$  : void fraction of the catalytic bed
- $\rho$  : density (kg/m<sup>3</sup>)  $\rho_{Bulk,Cat}$ : Bulk catalyst density =weight of catalyst/ annular reactor volume,  $\rho_{Bulk,Cat}$  : gas mixture density



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- $\Phi$  : permeance decline coefficient
- $\eta$  : viscosity ( $\mu\text{P}$ )
- $\Omega$  : collision integral,  $\Omega_v$ : Viscosity collision integral,  $\Omega_D$  : Diffusion collision integral,  $\Omega_{D,p}$  : Polar diffusion collision integral
- $\omega$  : acentric factor
- $\mu_r$  : dimensionless dipole moment
- $\mu$  : dipole moment (debyes)
- $\eta$  : viscosity ( $\mu\text{P}$ ),  $\eta_m$ : viscosity of the mixture ( $\mu\text{P}$ ),  $\eta_{i \text{ or } j}$ : pure component viscosity ( $\mu\text{P}$ )
- $\delta$  : polar parameter
- $\kappa$  : association factor
- $\sigma$  : characteristic length parameter ( $\text{\AA}$ );  $\sigma_i$ , for pure  $i$ ;  $\sigma_{ij}$ , for an  $i$ - $j$  interaction
- $\epsilon$  : characteristic energy parameter;  $\epsilon_i$ , for pure  $i$ ;  $\epsilon_{ij}$ , for an  $i$ - $j$  interaction
- $\tau$  : tortuosity, dimensionless

### Superscripts

- e : effective
- Shell : shell side of the membrane module where reaction is taking place (Reaction side)
- Tube : tube side of the membrane module where permeated  $\text{H}_2$  is flowing through (Permeate side)

- 0 : inlet condition

### Subscripts

- cr : cross section
- g : gas
- $i/j$  :  $i/j^{\text{th}}$  species

ef : effective

mix : mixture

p : polar

## **Abbreviations**

ASU : air separation unit

B : billion

BC : boundary condition

CLC : chemical looping combustion cycle

ENPV : expected net present value

GE : General Electric

GEE : General Electric Energy

GEE IGCC : An IGCC plant with uses GE gasifier

GHSV : gas hourly space velocity

HAZOP : hazard and operability analysis

HRSG : heat recovery steam generator

IGCC : integrated gasification combined cycle

IGCC-MR : an IGCC plant with embedded membrane reactor

IGCC-PBR : an IGCC plant with traditional shift reactors

M : million

MC : Monte Carlo

MR : membrane reactor

M&S : Marshall and Swift Cost Indexes

NPV : net present value

## **EPRG WP 1211**

OTM : oxygen transport membrane  
PC : pulverized coal  
PSA : pressure swing adsorption  
PSS : porous stainless steel  
PV : present value  
RRS : rupture disk-rupture disk-safety relief valve  
RH : resample historical data  
TD : triangular distribution  
w : with  
w/o : without  
WGSMR : water-gas shift membrane reactor