Title: **Water-gas Shift with Integrated Hydrogen Separation Process**

Annual Report

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Abstract

Optimization of the water-gas shift (WGS) reaction system for hydrogen production for fuel cells is of particular interest to the energy industry. To this end, it is desirable to couple the WGS reaction to hydrogen separation using a semi-permeable membrane, with both processes carried out at high temperatures to improve reaction kinetics and permeation. Reduced equilibrium conversion of the WGS reaction at high temperatures is overcome by product H_2 removal via the membrane.

This project involves fundamental research and development of novel cerium oxide-based catalysts for the water-gas-shift reaction and the integration of these catalysts with Pd-alloy H_2 -separation membranes supplying high purity hydrogen for fuel cell use. Conditions matching the requirements of coal gasifier-exit gas streams will be examined in the project.

The first-year screening studies of WGS catalysts identified Cu-ceria as the most promising high-temperature shift catalyst for integration with H_2-selective membranes. Formulations containing iron oxide were found to deactivate in the presence of CO_2, and were thus eliminated from further consideration. Cu-containing ceria catalysts, on the other hand, showed high stability in CO_2-rich gases. This type gas will be present over much of the catalyst, as the membrane removes the hydrogen produced from the shift reaction. Several catalyst formulations were prepared, characterized and tested in the first year of study. Details from the catalyst development and testing work were given in our first annual technical report.

Hydrogen permeation through Pd and Pd-alloy foils was investigated in a small membrane reactor constructed during the first year of the project. The effect of temperature on the hydrogen flux through pure Pd, $Pd_{60}Cu_{40}$ and $Pd_{75}Ag_{25}$ alloy membranes, each 25 μm thick, was evaluated in the temperature range from 250°C to 500°C at upstream pressure of 4.4 atm and permeate hydrogen pressure of 1 atm. Flux decay was observed for the Pd-Cu membrane above 500 ^{o}C. From 350-450 ^{o}C, an average hydrogen flux value of 0.2 mol H_2/m^2/s was measured over this Pd-alloy membrane. These results are in good agreement with literature data.

In this year's report, we discuss reaction rate measurements, optimization of catalyst kinetics by proper choice of dopant oxide (lanthana) in ceria, long-term stability studies, and H_2 permeation data collected with unsupported flat, 10 μm-thick Pd-Cu membranes over a wide temperature window and in various gas mixtures. The high-temperature shift catalyst composition was further improved, by proper selection of dopant type and amount. The formulation 10 at%Cu-Ce(30 at%La)Ox was the best; this was selected for further kinetic studies. WGS reaction rates were measured in a simulated coal-gas mixture. The stability of catalyst performance was examined in 40-hr long tests. A series of hydrogen permeation tests were conducted in a small flat-membrane reactor using the 10 μm-thick Pd-Cu membranes. Small inhibitory effects of CO and CO_2 were found at temperatures above 350 ^{o}C, while H_2O vapor had no effect on hydrogen permeation. No carbon deposition took place during many hours of membrane operation. The reaction extent on the blank (catalyst-free) membrane was also negligible. A larger flat- membrane reactor will be used next year with the catalyst washcoated on screens close coupled with the Pd-Cu membrane.

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Introduction

The water-gas shift (WGS) reaction, $CO + H_2O \leftrightarrow CO_2 + H_2$, is used industrially for the production of hydrogen for ammonia synthesis and for adjusting the CO/H_2 ratio for the subsequent synthesis of methanol. Recently, there has been a renewed interest in WGS because of its potential use in supplying hydrogen for fuel cell power generation. Fuel cells are currently undergoing rapid development for both stationary and transportation applications.

Industrially, WGS is carried out at two temperature regimes; the high-temperature shift reactors use a Fe_2O_3-Cr_2O_3 catalyst, which can effectively reduce CO from several percentage points to the equilibrium CO value dictated by the operating temperature; further reduction of CO takes place at low temperatures over a more active catalyst based on Cu-ZnO. This catalyst is very sensitive to temperature excursions, operating in the narrow temperature window of 200-250°C, requires careful activation (in H_2 gas), and is readily deactivated by exposure to air or by water condensation. Thus, this type catalyst is unsuitable for use with low-temperature PEM fuel cells, under development for automotive applications.

Optimization of the WGS system for hydrogen production for fuel cells is of particular interest to the energy industry. High-temperature shift may be used to increase the hydrogen content of coal gas produced by gasification or of reformate gas produced by autothermal reforming of fuel oils. Conversion of CO over the high-temperature shift (HTS) catalyst is limited by the equilibrium of the shift reaction. To overcome the equilibrium limitation, a highly selective membrane may be used to remove hydrogen from the product gas. Thus, a HTS membrane reactor is envisioned for pure hydrogen generation to feed a number of fuel cells, including the solid oxide (SOFC) and the PEM fuel cell types. There are several challenges in developing a HTS membrane reactor that is small and cost-effective. The membrane should comprise a minimal amount of Pd, and the catalyst should be both robust and active in the presence of large amounts of CO_2, since H_2 will be continuously removed by the membrane. Finally, the membrane permeance must match the hydrogen flux dictated by the catalyst activity.

In an economical analysis of palladium membrane reactors for the WGS reaction[1], costs of different membrane systems were compared with costs of conventional systems (high temperature shift followed by low temperature one and separation) for a fixed pure hydrogen recovery. The results showed that to make membrane reactors possible as alternatives to traditional reactor systems, the Pd thickness should be less than or equal to 20μm, or some new material or alloy more permeable to hydrogen and less expensive than palladium should be found.

This project involves fundamental research and development of novel shift catalysts and the integration of these catalysts with H_2-separation membranes supplying high purity hydrogen for fuel cell power generation. The approach taken here is to treat the WGS catalyst and membrane as a single, tightly coupled system designed to operate efficiently over a wide temperature window.

The first-year screening studies of WGS catalysts identified Cu-ceria as the most promising high-temperature shift catalyst for integration with H_2-selective membranes[2]. Formulations containing iron oxide were found to deactivate in the presence of CO_2, and were thus eliminated from further consideration. This is in agreement with a recent report from SUNY-Buffalo [3]. Cu-containing ceria catalysts, on the other hand, showed high stability in CO_2-rich gases. This type gas will be present over much of the catalyst, as the membrane removes the hydrogen produced from the shift reaction. Several catalyst formulations were prepared, characterized and tested in the first year of study. Details from the catalyst development and testing work were given in our first annual technical report [2].

Hydrogen permeation through membrane foils was investigated in a small membrane reactor constructed during the first year of the project. The effect of temperature on the hydrogen flux through pure Pd, $Pd_{60}Cu_{40}$ and $Pd_{75}Ag_{25}$ alloy membranes, each 25 nm thick, was evaluated in the temperature range from 250^oC to 500^oC at upstream pressure of 4.4 atm and permeate hydrogen pressure of 1 atm. Flux decay was observed for the Pd-Cu membrane above 500 ^oC. From 350-450 ^oC, an average hydrogen flux value of 0.2 mol $H_2/m^2/s$ was measured over this Pd-alloy membrane[2]. These results are in good agreement with literature data [4].

In this year's report, we discuss reaction rate measurements, optimization of catalyst kinetics by proper choice of dopant oxide (lanthana) in ceria, long-term stability studies, and H_2 permeation data collected with unsupported flat Pd-Cu membranes over a wide temperature window and in various gas mixtures.

Experimental

1. Catalyst preparation

The Cu- ceria catalysts were prepared by the urea gelation co-precipitation (UGC) method, which produces more homogeneous mixed oxides with finer particle size than conventional co-precipitation [5]. Details about this synthesis procedure were given in last year's report [2]. All catalysts were calcined in air at 650 ^oC for 4h.

New catalyst formulations were prepared to examine the effect of dopant type and amount on reaction kinetics and the long-term catalyst activity. La or Zr was used as dopant in ceria in small amounts (from 10 to 30 at %). This was added to improve the surface area and thermal stability of ceria. Copper oxide was a minor component in ceria. The amount of copper was fixed at 10 at %, based on the screening tests conducted last year [2]. All catalyst formulations are written with the atomic percentages of the additive indicated. As an example, the formulation $10\%Cu-Ce(30\%La)O_x$ means:

$Cu/(Cu+Ce+La) = 0.10$, $Ce/(Cu+Ce+La) = 0.90 \times 0.70$, and $La/(Cu+Ce+La) = 0.90 \times 0.30$.

A leached sample was prepared by immersing a $10\%Cu-Ce(10\%La)O_x$ sample in 70% HNO_3 solution for 8 h, filtering and washing it twice with de-ionized water, drying it at about 100ºC overnight and calcining it at 400ºC for 4 h.

2. Catalyst characterization

The elemental composition of each sample was analyzed by inductively coupled plasma (ICP) spectroscopy.

The BET surface area of each sample was measured by single- point nitrogen adsorption/ desorption cycles in a Micromeritics Pulse Chemisorb 2705 instrument, using a $30\%N_2$/He gas mixture. The results are shown in Table 1. The addition of La or Zr at 30 at% in ceria has a similar effect on the surface area of the samples. However, the higher-lanthanum content material has lower surface area (86 m2/g) than the 10% La-doped ceria (112 m2/g) indicating a small dopant effect on this property after calcination at 650 ^{o}C.

Temperature programmed reduction (TPR) tests with H_2 were run in the same instrument according to the following procedure: about 1 gram sample was heated to 350°C (10°C/min) in 50ml/min (STP) $20\%O_2$/He and kept at this temperature for 30 min to fully oxidize the sample. Heating was then stopped and when the temperature dropped to 200°C, purge gas, 50 ml/min N_2 (99.999%), was switched in. The temperature-programmed reduction began after the sample had been cooled to room temperature. The reduction gas was $20\%H_2/N_2$ (50 ml/min) and the heating rate was 5°C/min.

Temperature programmed oxidation (TPO) tests were run according to the following procedure: the sample was heated to 350°C (10°C/min) in He (99.999%, 50ml/min) and kept at 350°C for 30 min to remove any CO_2 absorbed. After the sample cooled down to room temperature, heating in a $20\%O_2$/He (50ml/min) gas mixture took place to 650°C. The heating rate was 5°C/min. A mass spectrometer (MKS-model RS-1) was used for detection of CO_2.

3. Activity tests in a packed-bed microreactor

Rate measurements were performed at atmospheric pressure. Catalyst samples (< 150 µm size) were diluted with quartz salt and loaded on a quartz frit at the center of a flow quartz-tube microreactor, which was heated inside an electric furnace. All samples were used without activation. Water was injected into the flowing gas stream by a calibrated syringe pump and was vaporized in the heated gas feed line before entering the reactor. A condenser filled with ice was installed at the reactor exit to collect water. The exit gas was analyzed by a Hewlett Packard 5890A gas chromatograph, equipped with a thermal conductivity detector and a ¼-inch dia. x 6 ft long Carbosphere column for CO and CO_2 separation. The CO conversion was calculated from the concentrations of CO and CO_2 detected. There was no methane formation even when H_2 and CO_2 were included in the feed gas mixture.

The activation energies were calculated from the reaction rates at CO conversions < 20%.

4. Membrane Apparatus and Testing Procedure

A stainless steel membrane holder with copper gaskets was constructed and used for hydrogen permeability tests. Bleed and permeate flow rates were measured by a bubble flow meter and had a deviation of ± 3-5%. The temperature was measured by a thermocouple at the outer edge of the membrane holder. The flow rate of each gas was controlled by a mass flow meter. When water was used, it was injected into the flowing gas stream by a calibrated syringe pump and vaporized in the heated gas feed line before entering the furnace.

Reduced thickness, free-standing Pd-Cu membranes were prepared by wet etching of 25- μm thick foils according to methods disclosed in US Patent 6,103,028 [6]. Each Pd_{60}-Cu_{40} membrane (diameter 9/16 inch, disk area 1.6 cm^2) was punched out, washed with acetone using ultrasound and cleaned with fine blotting paper. Fresh membranes were heated to 400^oC at a hydrogen upstream pressure of 4.4 atm (50 psig) and hydrogen downstream pressure 1 atm (0 psig). After the permeation in pure hydrogen was stable, hydrogen permeation was measured at the desired temperatures and pressures in either pure H_2 or a gas mixture. After about 20 min of stabilization, the H_2 flux was measured over a period of 30-40 min at each temperature to ensure steady-state operation.

To examine how fast the WGS reaction would be on the blank Pd-Cu membrane (without catalyst), a test was run with a feed gas of 20%CO-60%H_2O-20%CO_2 at 450^oC in the same system with the permeation path blocked. The total gas flow was 100 ml/min and the conversion of CO was about 1.7%. Since the flowrate of each mixture was 800 ml/min, and at 450^oC the reverse WGS reaction is slower than WGS reaction, both the WGS and reverse WGS reaction would be too slow to change the composition of mixture.

Results and Discussion

1. Effect of Dopant

Steady-state conversions of CO in WGS over the 10%Cu-Ce(dopant)O_x samples with dopants 10%La, 30%La and 30%Zr were measured in the microreactor in a gas mixture of 11%CO-23%H_2O-11%CO_2-17%H_2-He, with a catalyst amount of 0.05 g and at a contact time of 0.0101g*s/cc (STP). The results are shown in Figure 1. The 30%La sample has the highest activity among the three samples under these conditions. On the basis of the surface areas of these samples which are all similar (see Table 1), we conclude that the effect is chemical, and not scalable with the surface area.

Figure 2 shows the long-term stability of 10%Cu-Ce(30%La)O_x and 10%Cu-Ce(30%Zr)O_x in the same gas mixture at 450^oC for 40 h. Again, a higher conversion of CO was measured over the La-doped sample than the Zr-doped catalyst throughout the test period. Both catalysts deactivated fast at the beginning, but their activities became stable after several hours on-line. Carbon deposition was not found on either of the two used samples by temperature programmed oxidation (TPO). Deactivation may thus be a result of structural modifications of the catalysts in the reaction gas mixture. The used catalysts had lost between 20-35% of their initial surface areas shown in Table 1.

On the basis of these findings, the 10%Cu-Ce(30%La)O_x formulation was selected for further study in this project.

2. WGS reaction rate measurements

Figure 3 shows WGS reaction rates measured over the 10%Cu-Ce(30%La)O_x and the copper-free Ce(30%La)O_x catalysts in the simulated coal-gas mixture 11%CO-23%H_2O-11%CO_2-17%H_2-He. The activation energies of the reaction over these catalysts are 69 kJ/mol and 91 kJ/mol, respectively, at

this condition. This large difference is due to the modification of the surface of ceria by the addition of copper. As discussed in last year's report [2], copper greatly increases the reducibility of the surface oxygen of ceria. This modified site is the active site for the WGS reaction. The reaction pathway on copper-free ceria is different, and of larger activation energy.

At high temperatures, such as 450^oC, the fresh $10\%Cu-Ce(30\%La)O_x$ sample deactivates fast. To get a stable conversion and reaction rate, and an accurate measurement of the activation energy, we measured the reaction rate over the $10\%Cu-Ce(30\%La)O_x$ sample after it had been used in the reaction gas mixture at 450^oC for 20 h. (The $Ce(30\%La)O_x$ sample does not deactivate that fast.)

Figure 4 shows WGS reaction rates measured over the $10\%Cu-Ce(30\%La)O_x$ and $Ce(30\%La)O_x$ in a gas mixture free of H_2 and CO_2; $11\%CO-23\%H_2O-He$. The activation energies are 54 kJ/mol and 75 kJ/mol, respectively, i.e. considerably lower than the values measured in the simulated syngas in Figure 3. Therefore, there is inhibition of the reaction by at least one of the two reaction products (H_2 or CO_2). Under the conditions of Figure 4, both samples deactivated much more slowly than before. For the fresh $10\%Cu-Ce(30\%La)O_x$ sample, only the data from 250 to 350^oC were used to calculate the activation energy because the CO conversions at 400^oC and 450^oC were outside the kinetics regime (i. e. >20%).

We further examined the properties of the Cu-ceria catalysts after leaching some of the copper out by the use of nitric acid. According to ICP analysis, the HNO_3-leached $10\%Cu-Ce(10\%La)O_x$ sample lost ~55% Cu, ~10% La, and almost no Ce (~1%). The leached sample has approximately the same composition as a freshly prepared $5\%Cu-Ce(10\%La)O_x$. That is the reason we compared the activity of the leached sample with the 5%Cu-ceria sample prepared by UGC. The WGS rates over the leached sample, the original $10\%Cu-Ce(10\%La)O_x$ and a fresh $5\%Cu-Ce(10\%La)O_x$ are compared in Table 2 and Figure 5. The three samples have almost the same activation energy, which indicates that they share a common reaction mechanism. The reaction rate over the 10%Cu catalyst is about twice that over the 5%Cu sample, and the reaction rate over the leached sample is about half that over the 5%Cu sample.

The following hypothesis is made to explain the above results: It is known from previous work [7] that at copper loadings of less than 15 at%, copper exists in the form of very fine copper oxide clusters, highly dispersed in ceria; these are associated with the active sites. Since the 10%Cu-ceria sample has twice as much copper as the 5%Cu-sample, the former has twice the number of active sites and thus twice the reaction rate of the latter. After the 10%Cu sample is leached with HNO_3, although half of the Cu is still left in the catalyst, about half of those sites have now changed from clusters to inactive (or less active) isolated cupric ions, embedded in the ceria lattice [7]; this would explain the loss of overall activity shown in Figure 5 and Table 2. However, it would leave the activation energy unaffected, which is what we observed.

To further test the above mechanistic arguments, all three samples were subjected to H_2-TPR. The TPR profiles in Figure 6 show that the leached sample is less reducible than the fresh 5%Cu and 10%Cu-ceria samples. This correlates well with the lower reaction rates measured over the leached catalyst.

4. Hydrogen permeation tests

Hydrogen permeation through 10μm-thick $Pd_{60}Cu_{40}$ alloy membranes was investigated in pure hydrogen in the membrane reactor system. To study the effects of CO, CO_2 and H_2O, hydrogen permeation was also measured in the following gas mixtures: 53%CO-H_2, 53%CO_2-H_2, 53%H_2O-H_2 and 10%CO-23%H_2O-10%CO_2-10%He-47%H_2, and, for comparison, also in 53%He-H_2 (or 53%Ar-H_2) at the same flow rate and temperature conditions.

Figure 7 shows the hydrogen flux (hydrogen permeation) through the 10μm-thick membrane in the temperature range from $80^{o}C$ to $450^{o}C$ at upstream pressure 50 psig (4.4 atm absolute) and permeate hydrogen pressure 0 psig (1 atm absolute). Our data of hydrogen permeation through the 25μm-$Pd_{60}Cu_{40}$ membranes (see last year's report [2]) and data in the literature[4](US patent 3,439,474) are also shown in this figure for comparison. The superiority of the thin membrane is clearly depicted in this figure.

Figure 8 shows the effect of CO and CO_2 on H_2 permeation through the 10μm-thick membrane in the temperature range 350-$450^{o}C$. In the 53%CO-H_2 and the 53%CO_2-H_2 gas mixtures, the permeation was less than in the 53%He-H_2 mixture or the 53%Ar-H_2 gas mixture. This shows that both CO and CO_2 decrease H_2 permeation. The lower the temperature, the more pronounced the inhibition. Because the diffusion coefficient of the Ar-H_2 system is close to that of CO-H_2 or CO_2-H_2 gas mixture, the effect of CO and CO_2 is not due to mass transfer limitation. Overall, the inhibitory effects are minor above $400\ ^{o}C$.

Figure 9 shows that H_2O vapor has a negligible effect on hydrogen permeation through the Pd-Cu membrane. Finally, the overall effect of the simulated coal-gas mixture on the permeation was evaluated. The results are shown in Figure 10 in a mixture of 10%CO-23%H_2O-10%CO_2-10%He-47%H_2; minor inhibition of hydrogen permeation is observed. From this study, we concluded that operation of the coupled catalyst/membrane system at $450\ ^{o}C$ would be almost free of inhibitory effects on hydrogen permeation.

A larger (10cm x 5cm oval) flat Pd-Cu membrane reactor will be used next with the catalyst close coupled with the membrane. A schematic of this is shown in Figure 11. The decision was made to prepare catalyst-loaded screens to place above the Pd- membrane in this reactor. Washcoating the screens in a slurry of the micronized catalyst will be used. Global reaction rate measurements, parametric studies, and modeling of the WGS membrane reactor are planned for next year.

Conclusions/Future Work Plan

During the second-year of the project, the high-temperature shift catalyst composition was further improved, by proper selection of dopant type and amount. The formulation 10%Cu-Ce(30%La)Ox was the best; this was selected for further kinetic studies. WGS reaction rates were measured in a simulated coal-gas mixture, and the long-term stability of catalyst performance established. A series of hydrogen permeation tests were conducted in a small flat-membrane reactor using 10 μm thick Pd-Cu membranes (produced from thinning of 25 μm- thick foils). Small inhibitory effects of CO and CO_2 were found at temperatures above 350 ^{o}C, while H_2O vapor had no effect on hydrogen permeation through the Pd-Cu membrane. No carbon deposition took place during many hours of membrane

operation. The reaction extent on the blank (catalyst-free) membrane was also negligible. A larger flat-membrane reactor will be used next year. Washcoating of catalysts on screens to be used in this membrane reactor is currently underway. Next year, global reaction rate measurements and reactor modeling of this system will take place.

Literature Cited

[1] A. Criscuoli, A. Basile, E. Drioli and O. Loiacono, An economic feasibility study for water gas shift membrane reactor, *J. Membrane Science* 181 (2001) 21-27

[2] M. Flytzani-Stephanopoulos et al. Annual Report to DOE, *Water Gas Shift with Integrated Hydrogen Separation Process,* December 2001.

[3] Carl R. F. Lund, *Water gas shift kinetics over iron oxide at membrane reactor conditions,* Final Report to DOE, FE-FG2699-FT40590, August 2002.

[4] U.S. Patent 3,439,474

[5] Y. Li, Q. Fu, M. Flytzani-Stephanopoulos, *Appl. Catal. B*, 27 (2000) 179-191

[6] U.S. Patent 6,103,028

[7] W. Liu and M. Flytzani-Stephanopoulos, *J. Catal.* 153 (1995) 304

Table 1. Specific surface area of WGS catalysts[#]

Sample[*]	BET surface area (m^2/g)
10%Cu-Ce(30%La)O_X	80
10%Cu-Ce(10%La)O_X	112
10%Cu-Ce(30%Zr)O_X	86
Ce(30%La)O_X	85
5%Cu-Ce(10%La)O_X	107

all catalysts were prepared by the UGC method; calcined at 650°C, 4h

* used as-prepared, after crushing to < 150 µm -size particles

Table 2. WGS Reaction Rates over various catalysts in 11%CO-23%H_2O-He gas mixture

Temperature (^{o}C)	Reaction rate (mol CO_2/g*s)		
	10%Cu-Ce(10%La)O_x	5%Cu-Ce(10%La)O_x	HNO$_3$-leached 10%Cu-Ce(10%La)O_x
450	1.26 x 10$^{-3#}$	8.39 x 10$^{-5#}$	4.71 x 10$^{-4#}$
400	8.47 x 10$^{-4#}$	4.95 x 10$^{-5#}$	2.53 x 10^{-4}
350	4.76 x 10$^{-4#}$	2.54 x 10^{-4}	1.14 x 10^{-5}
300	2.09 x 10^{-4}	9.67 x 10^{-5}	4.69 x 10^{-5}
250	7.21 x 10^{-5}	3.41 x 10^{-5}	

data may not be in the kinetics regime, i. e. CO conversion > 20%
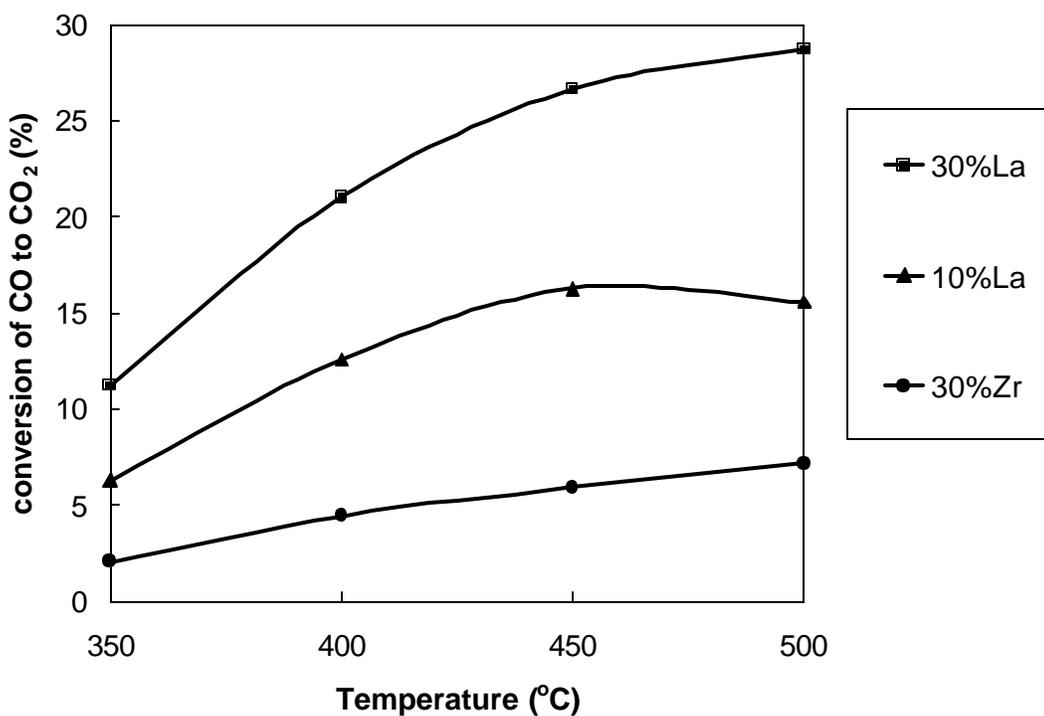
Figure 1. Effect of dopant in ceria on the WGS activity of 10%Cu-Ce(dopant)O_x catalyst 11%CO-23%H_2O-11%CO_2-17%H_2-He, contact time = 0.0101g*s/cc (STP)
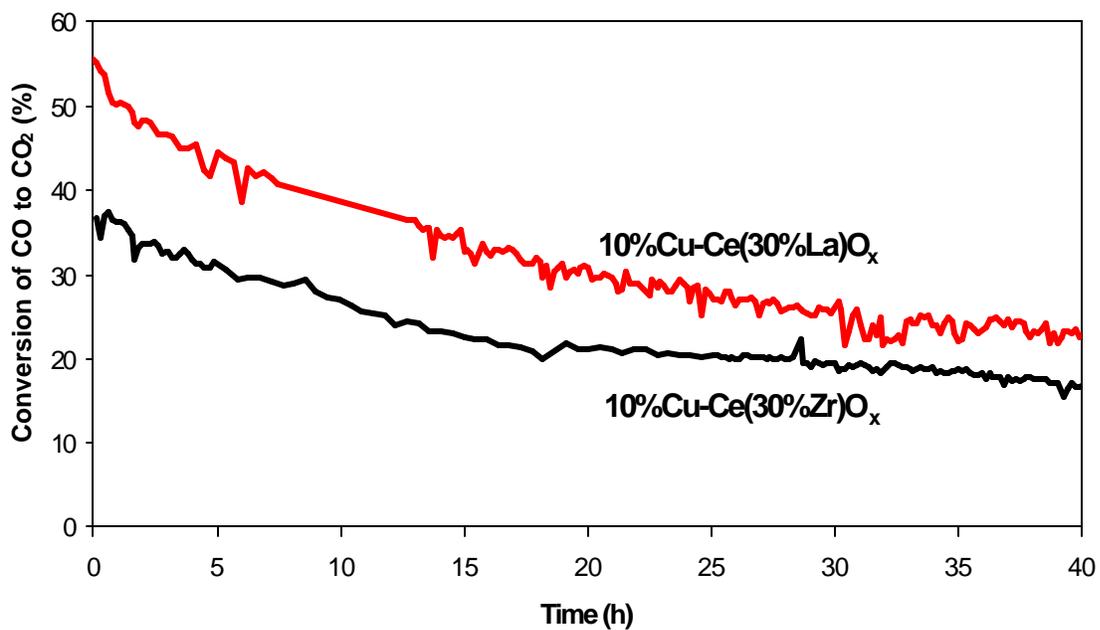
Figure 2. Long-term stability of 10%Cu-Ce(30%La)O_x and 10%Cu-Ce(30%Zr)O_x, 11%CO-23%H_2O-11%CO_2-17%H_2-He, contact time = 0.0101g*s/cc (STP), 450°C
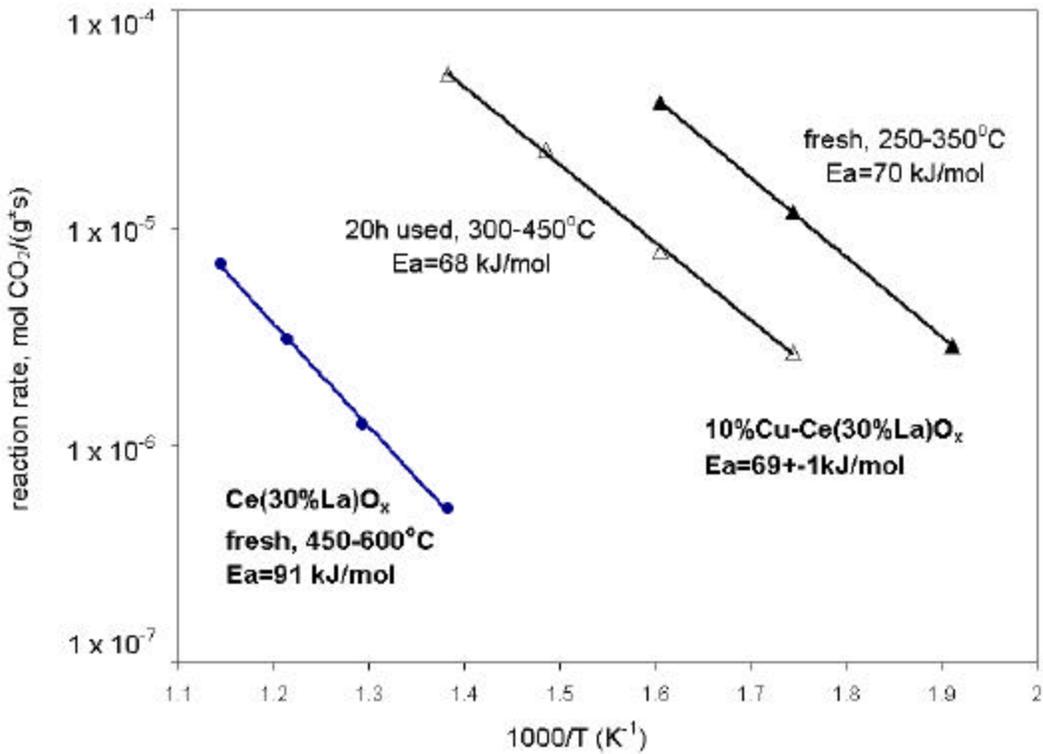
Figure 3. WGS reaction rates over 10%Cu-Ce(30%La)O_x and Ce(30%La)O_x
in 11%CO-23%H_2O-11%CO_2-17%H_2-He gas mixture
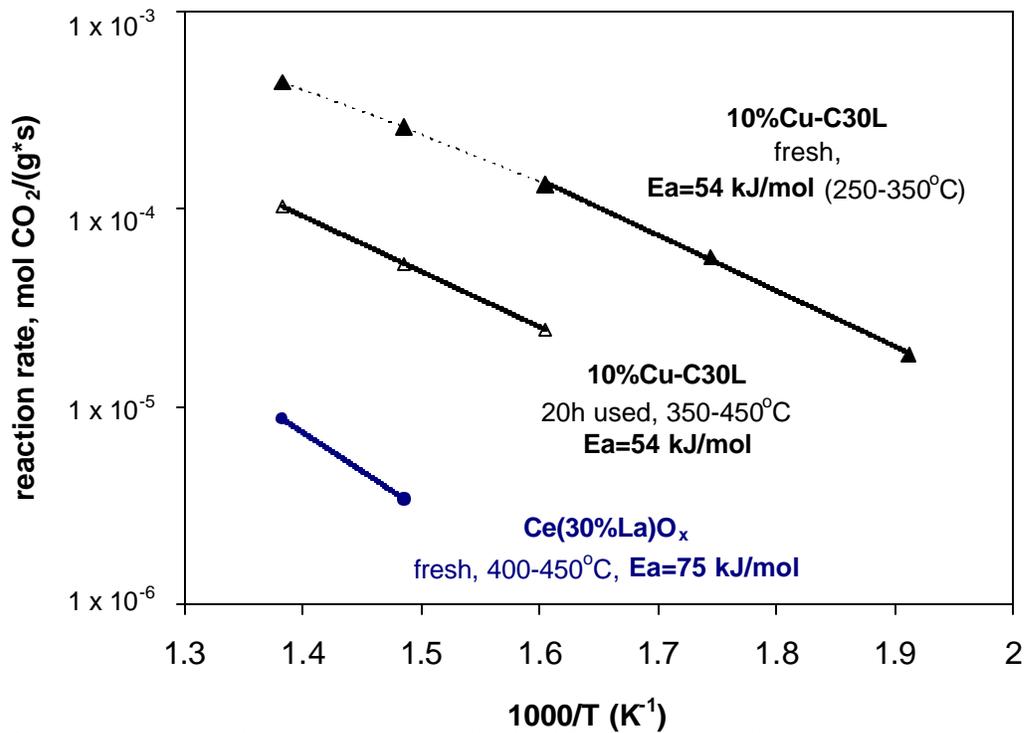
Figure 4. WGS reaction rates over 10%Cu-Ce(30%La)O_x and Ce(30%La)O_x
in 11%CO-23%H_2O-He gas mixture
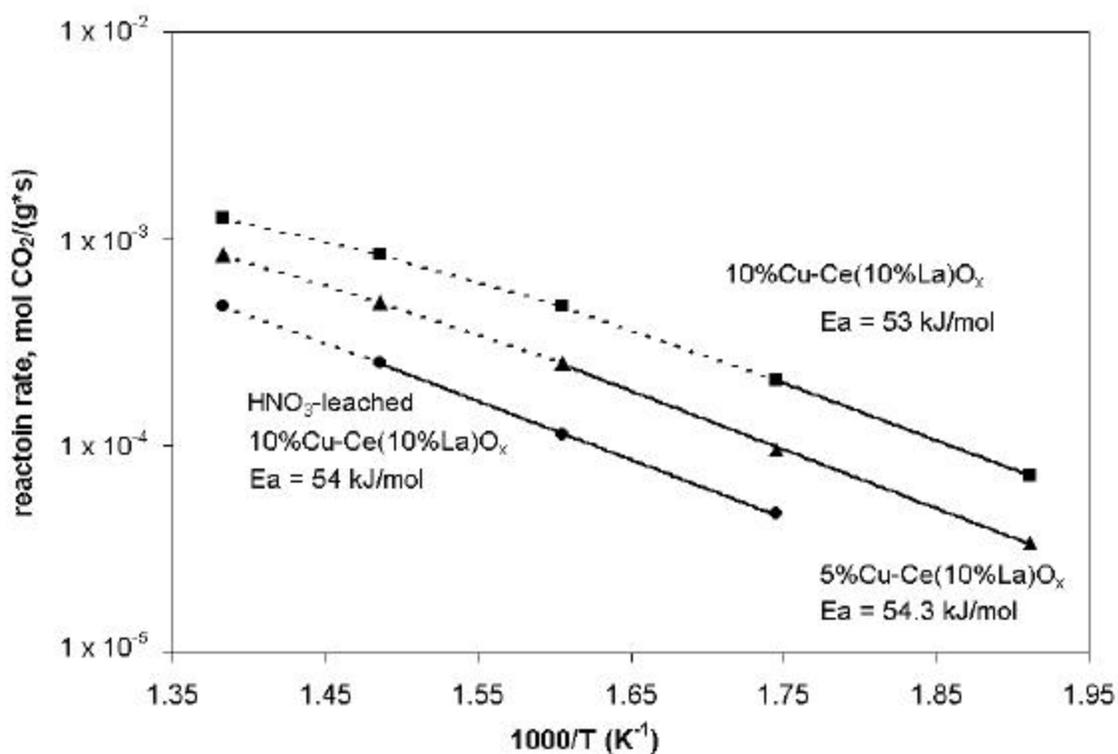
Figure 5. WGS reaction rates over 10%Cu-Ce(10%La)O_x, 5%Cu-Ce(10%La)O_x, and HNO$_3$-leached 10%Cu-Ce(10%La)O_x samples in 11%CO-23%H$_2$O-He gas mixture
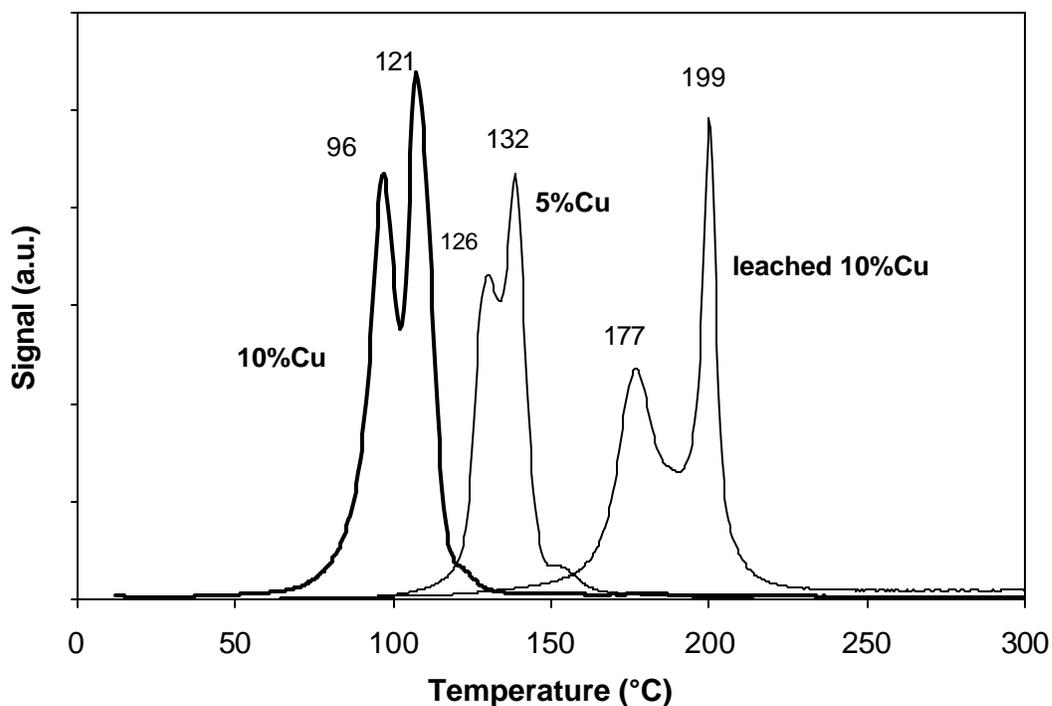
Figure 6. H$_2$-TPR: HNO$_3$-leached 10%Cu-Ce(10%La)O_x compared with the 10%Cu- and 5%Cu-Ce(10%La)O_x samples
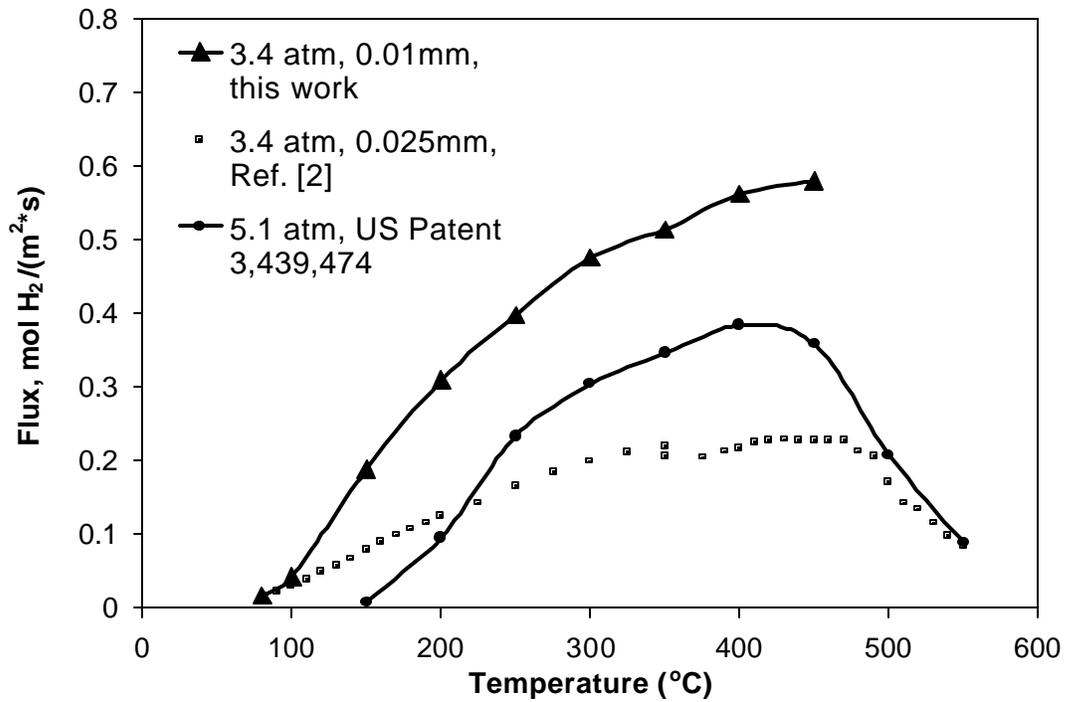
Figure 7. H_2 permeation through Pd_{60}-Cu_{40} membranes, 10μm –thick as compared with 25μm-thick membrane data in last year's report[2] and in the literature[4], P1=50psig (4.4 atm abs.), P2=0psig
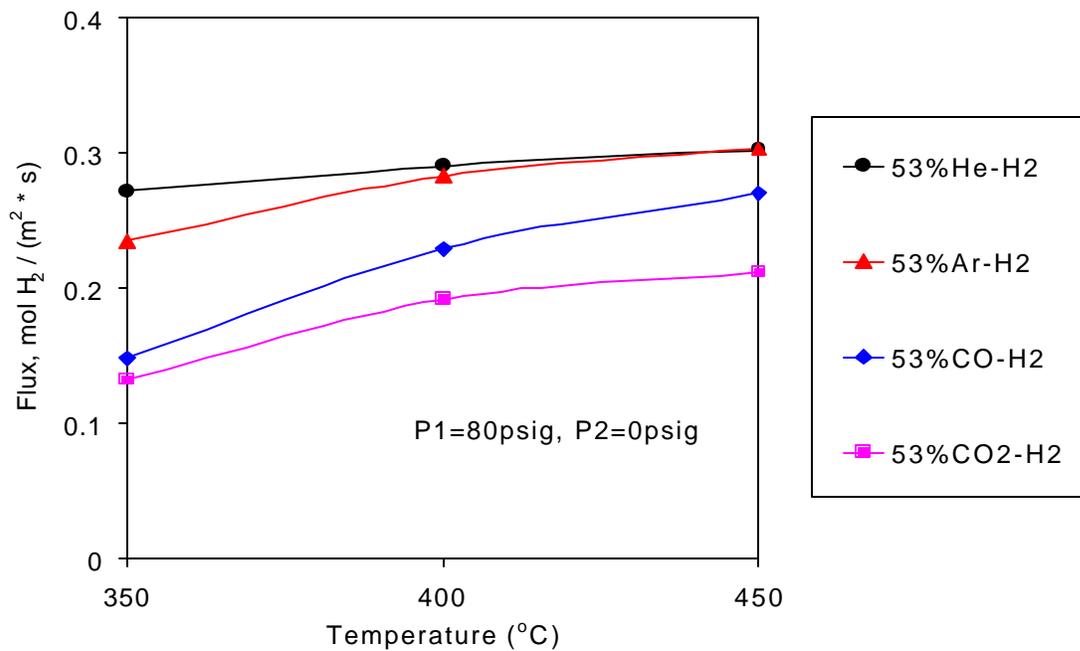
Figure 8. H_2 permeation through a 10μm-thick Pd-Cu membrane
53%CO-H_2, 53%CO_2-H_2, 53%He-H_2 and 53%Ar-H_2, P1=80psig, P2 =0psig
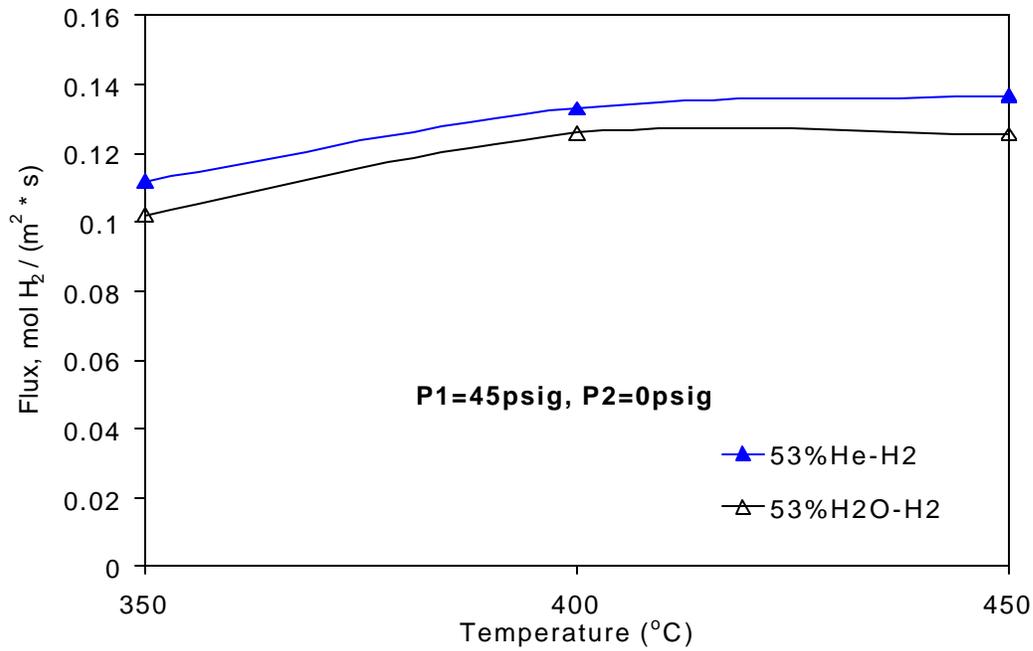
Figure 9. H_2 permeation through 10μm Pd-Cu membrane in 53%H_2O-H_2 and 53%He-H_2, P1=45psig, P2=0psig
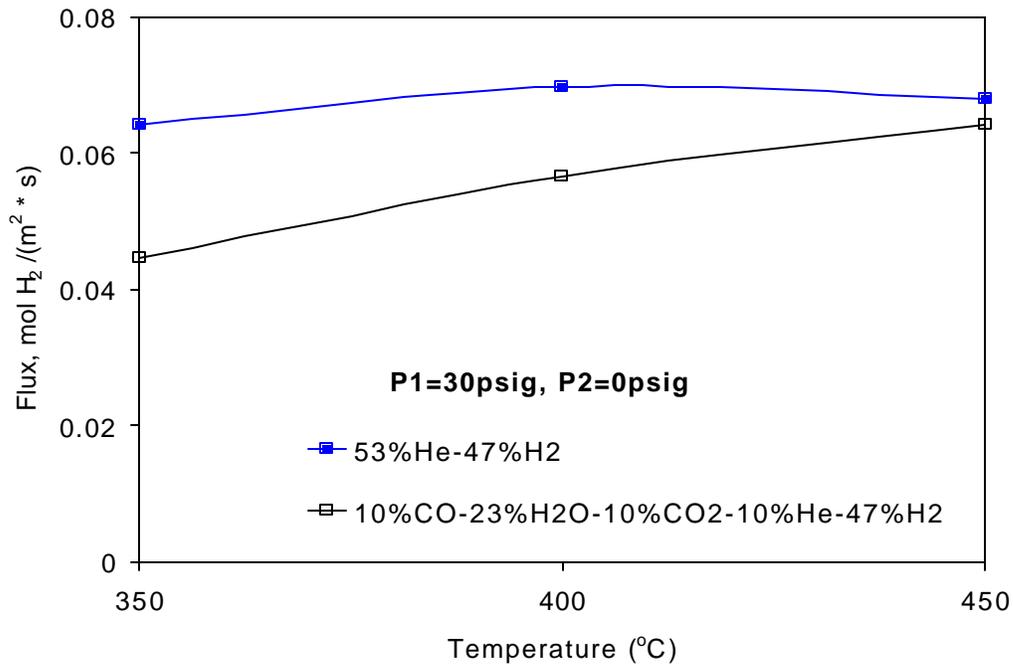
Figure 10. H_2 permeation through a 10μm-thick Pd-Cu membrane
10%CO-23%H_2O-10%CO_2-10%He-47%H_2 gas and 53%He-47%H_2 gas, P1=30psig, P2=0psig

Laboratory Membrane Reactor
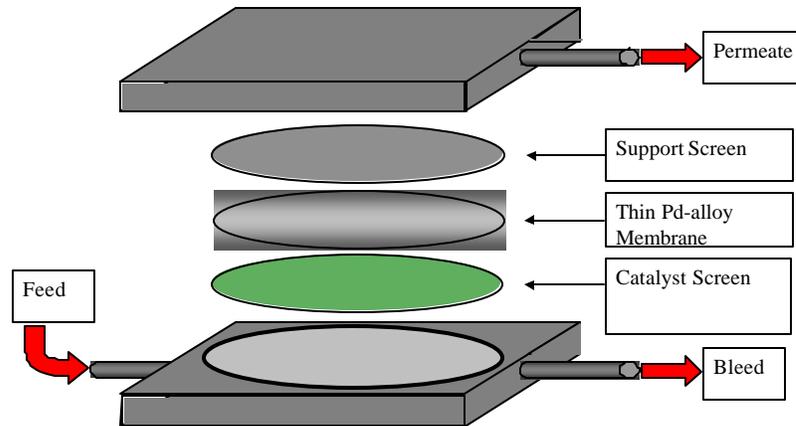
Figure 11. Schematic of WGS membrane reactor

Permeate

Support Screen

Thin Pd-alloy Membrane

Catalyst Screen

Feed

Bleed