Title:Water-gas Shift with Integrated Hydrogen SeparationProcess

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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 temperature to improve reaction kinetics. 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.

In the first year of the project, we prepared a series of nanostructured Cu- and Fe-containing ceria catalysts by a special gelation/precipitation technique followed by air calcination at 650° C. Each sample was characterized by ICP for elemental composition analysis, BET-N2 desorption for surface area measurement, and by temperature-programmed reduction in H₂ to evaluate catalyst reducibility. Screening WGS tests with catalyst powders were conducted in a flow microreactor at temperatures in the range of 200-550°C. On the basis of both activity and stability of catalysts in simulated coal gas, and in CO₂-rich gases, a Cu-CeO₂ catalyst formulation was selected for further study in this project. Details from the catalyst development and testing work are given in this report. Also in this report, we present H₂ permeation data collected with unsupported flat membranes of pure Pd and Pd- alloys over a wide temperature window.

Table of Contents

Page

Abstract	3
Introduction	5
Experimental	. 6
Results and Discussion	. 7
Conclusions	10
Literature cited	. 10
Table 1	11
Table 2	12
Fig.1-1313-	-19

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/H₂ 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 -Cr₂O₃ 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 H₂ 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 the 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 reaction¹, 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. Their results showed that to make membrane reactors possible as alternatives to traditional apparatus, the Pd thickness should be less than or equal to 20μ m, 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.

An active and robust catalyst based on Cu-CeO₂, previously identified by our group as a most promising WGS catalyst², was selected for study in this project. This was compared to iron oxide-ceria catalysts and to a commercial iron oxide-based HTS catalyst. Several catalyst formulations were prepared, characterized and tested in the first year of study. Details from the catalyst development and testing work are given in this report.

Also in this report, we present H_2 permeation data collected with unsupported flat membranes of pure Pd and Pd- alloys over a wide temperature window.

Experimental

1. Catalyst preparation

The Cu- and Fe-containing 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^{2,3}.

The following steps are involved in the UGC method:

1) Dissolving 72 gram urea and $(NH_4)_4Ce(NO_3)_2$, La $(NO_3)_3$, Cu $(NO_3)_2$ · 3H₂O or Fe $(NO_3)_3$ · 9H₂O in the amount needed for 75 mmol sample in 600 ml de-ionized water;

- 2) Heating the solution to boiling;
- 3) Adding water to 900 ml when the precipitation begins (about 30 min after boiling);
- 4) Aging the precipitate in boiling water with constant stirring for 8 hours;

5) Filtering and washing the precipitate twice in 50-70°C water with constant stirring for 30 min each time;

- 6) Drying the precipitate at about 100°C for 12 h;
- 7) Crushing the dried lump into particles smaller than 150 µm in diameter;
- 8) Calcining the powder by heating slowly (2°C/min) to 650°C and keeping it at this temperature for 4 h;

Many formulations of the catalyst were prepared. In most cases, La was used as a dopant in ceria in small amounts (10 at %). This was added to improve the surface area and thermal stability of ceria. Copper or iron oxide was a minor component in ceria. All formulations are written with the atomic amounts of the additive indicated. As an example, the formulation $5at\%Cu-Ce(10at\%La)O_x$ means:

Cu/(Cu+Ce+La) = 0.05, $Ce/(Cu+Ce+La) = 0.95 \times 0.90$, and $La/(Cu+Ce+La) = 0.95 \times 0.10$.

C12-4-02, a commercial catalyst containing 80-95 wt% Fe₂O₃, 5-10 wt% Cr₂O₃, <5 wt% CrO₃, 1-5 wt% CuO and 1-5 wt% graphite, was provided by United Catalysts, Inc. in pellet form, and used after crushing it into <150 μ m size powder.

 $5wt\%CeO_2/C12-4-02$ was prepared by deposition/precipitation of cerium hydroxide onto the C12-4-02 particles through the following steps: adding Ce(NO₃)₃ solution dropwise into a slurry of the particles held at a constant pH=7 in a (NH₄)₂CO₃ solution and under constant stirring; aging the precipitate for 1 hour at 70°C; filtering it and washing with 70°C water for 30 min; drying the precipitate at about 100°C overnight; calcining the resultant powder at 400°C for 10 hours using a heating rate 2°C/min. to reach this temperature.

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.

Temperature programmed reduction (TPR) tests with H_2 were run in the same instrument according to the following procedure: heat about 1 gram sample to 350°C (10°C/min) in 50ml/min (STP) 20%O₂/He and keep it at this temperature for 30 min to fully oxidize the sample. Stop heating and let the temperature drop; at 200°C, switch in 50ml/min N₂ (99.999%). Start the temperature-programmed reduction after the

sample cools down to room temperature. The reduction gas was $20\%H_2/N_2$ (50 ml/min) and the heating rate was 5°C/min.

3. Activity tests in a packed-bed microreactor

Activity tests were performed at atmospheric pressure with 150 mg catalyst load on a quartz frit at the center of a flow quartz-tube microreactor (1.0 cm ID, 50cm long), which was heated inside an electric furnace. The contact time was $0.09g \cdot \text{s/cc}$, with the total gas flow rate of 100ml/min (STP). This corresponds to a gas hourly space velocity of 80,000 h⁻¹. All samples were used without activation. The initial screening tests were run in mixtures of CO and H₂O in He gas. Water was injected into the flowing gas stream by a calibrated water pump and 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 Carle gas chromatograph, equipped with a thermal conductivity detector and a ¹/₄-inch dia. x 6 ft long Carbosphere column for CO and CO₂ separation. The CO conversion was calculated from the concentration of CO and CO₂ detected. There was no methane formation even when H₂ and CO₂ were included in the feed gas mixture.

4. Membrane Apparatus and Testing Procedure

A stainless steel membrane holder with copper gaskets was constructed and used for hydrogen permeability tests. Hydrogen (99.97% purity) pressure was measured with an Omega PX880 pressure transducer with \pm 0.25% accuracy. The bleed flow rate was varied using an outlet valve. Bleed and permeate flow rates were measured by a bubble flow-meter and had a deviation of \pm 3-5%. The temperature of the membrane holder was regulated by heating tape and an Omega CN76000 temperature controller with \pm 1°C accuracy. The temperature was measured by a thermocouple at the outer edge of the membrane holder.

Membrane samples (disk area 0.358 cm²) were punched out, washed with acetone using ultrasound and mechanically cleaned with fine blotting paper. Fresh membranes were heated to 350°C at a hydrogen upstream pressure of 3.4 atm (50 psig) and hydrogen downstream pressure 1 atm (0 psig=14.7 psia). Hydrogen permeation was measured as the system was heated for 2-3 hours. When flux was stable, the system was cooled by steps $\Delta T=10^{\circ}$ C at the rate of 1°C/min. After about 20 min of stabilization, the H₂ flux was measured over a period of 30-40 min at each temperature to ensure steady-state operation.

Results and Discussion

1. Catalyst Characterization

Table 1 shows the BET surface areas of the catalyst samples prepared and tested during this reporting period. The surface area, after the 650° C calcination, was around 90 m²/g for samples containing about 10 at% Cu or Fe. At higher than 15at% content, addition of Cu or Fe decreases the surface area of the La - doped ceria.

Figure 1 shows H₂-TPR profiles of Ce(10at%La)O_x (C10L) and Cu-Ce(10at%La)O_x (Cu-C10L) with different atomic % of Cu. The C10L sample had two peaks. The first one (centered at 408°C) is attributed to reduction of the surface oxygen of ceria. The second one (766°C, not shown here but shown in Figure 2), lower and broader, is attributed to bulk oxygen reduction of ceria. These are well established in the literature.^{3,4} Each Cu-C10L sample has two or three peaks below 500°C. The H₂ consumption (mol

 H_2 per mol CeO₂) below 500°C, after subtraction of the H_2 needed to reduce all the CuO present, is close to the H_2 consumption of the first peak of C10L. It is not possible to separate the peaks due to reduction of CuO from those due to reduction of the surface oxygen of ceria. However, all consumption of H_2 over the Cu-containing ceria samples occurs at much lower temperatures than the first peak of C10L. This means that Cu addition dramatically increases the reducibility of the surface oxygen of CeO₂. The samples containing 10-15 at% Cu and are the most reducible. Addition of Cu does not affect the reducibility of the bulk oxygen of ceria. The peak temperatures and areas of the below-500°C peaks for each sample are shown in Table 2.

Figure 2 shows TPR profiles of C10L and Fe-C10L samples. Addition of Fe shifts the first peak of ceria slightly to the left, showing that addition of Fe slightly increases the reducibility of ceria. The second peak of ceria (around 770°C) remains at the same position, but a peak centered around 630°C appears and grows when adding Fe. The 630°C - peaks are probably due to the last step of Fe_2O_3 reduction, i.e. reduction of FeO into Fe. We assign the low temperature peaks of Fe-C10L samples to the reduction of surface oxygen of ceria, reduction of Fe_2O_3 into Fe_3O_4 , and Fe_3O_4 into FeO. The peak temperatures for the H₂-TPR of Fe-ceria samples are also shown in Table 2.

2. Catalyst screening tests

Catalysts were screened in a gas mixture containing 2 mol%CO-10 mol%H₂O-He. CO conversion plots are shown in Figure 3. Fe-containing doped ceria (Fe-C10L) samples have higher activities than C10L, showing that addition of Fe increases the activity of ceria. But the commercial catalyst (C12-4-02) is more active than the Fe-C10L samples and all Cu-C10L samples are more active than both the Fe-C10L samples and C12-4-02. Among the Cu-ceria samples, 10at%Cu-C10L and 15at%Cu-C10L are the most active. The activity test results correlate well with the H₂- TPR results, i.e. a higher WGS activity is seen for the more reducible samples. The CuO contained in C12-4-02 may be the reason that C12-4-02 is more active than the Fe-C10L samples.

The 10at%Cu-C10L sample, which was the most active of all the catalyst formulations screened, and the 13at%Fe-C10L, the most active of the Fe-C10L samples, were chosen for further study.

Effect of adding ceria into the commercial Fe-Cr catalyst C12-4-02

We did not find any evidence that ceria increases the reducibility of iron oxide by comparing the TPR results of 13at%Fe-C10L and the commercial Fe-Cr catalyst C12-4-02 (Figure 4). However, a small peak at 119°C in Fig.4 is indicative of reduction of some of the surface oxygen of ceria due to the interaction with the iron oxide. To find out whether ceria can increase the reducibility and activity of iron oxide, as it has been reported in the literature⁵⁻⁸, ceria was deposited onto the commercial C12-4-02 catalyst as described above. Figure 5 shows the H₂-TPR profile of C12-4-02 and 5wt%CeO₂/C12-4-02. For each sample, the first two overlapped peaks are attributed to reduction of CuO because C12-4-02 contains 1-5wt%CuO. The peak at 350°C for C12-4-02 and the one at 312°C for CeO₂/C12-4-02 are assigned to reduction of Fe₂O₃. The ceria -containing sample is reduced at lower temperatures. Thus, we verified that ceria addition increases the reducibility of iron oxide in the commercial Fe-Cr catalyst. The activity of C12-4-02 is also increased after the addition of ceria, as shown in Figure 6, but it is still lower than that of the copper-containing sample, 10at%Cu-C10L.

The TPR peak temperatures of both $CeO_2/C12-4-02$ and C12-4-02 are shown in Table 2.

3. WGS activity and stability in a simulated coal-gas composition

In many practical cases, hydrogen will be obtained from coal gas mixtures. Therefore, the WGS activity and stability of the 10at%Cu-C10L and 13at%Fe-C10L catalysts were tested and compared with the commercial catalyst C12-4-02 in a simulated coal gas composition, 10%CO-34%H₂O-10%CO₂-15%H₂-He (here helium is used instead of N₂). 10at%Cu-C10L is still more active than C12-4-02, which is more active than 13at%Fe-C10L in this gas mixture (Figure 7). All three catalysts showed stable performance in the simulated coal gas mixture at 450°C, but each achieved a different CO conversion (Figure 8). Only the Cu-ceria catalyst was active enough for the equilibrium CO conversion to be approached at these conditions.

4. Catalyst stability in CO₂- and H₂- rich gases

The effect of the product gases on the kinetics of the WGS reaction on ceria-based catalysts will be studied systematically in the second year of the project. However, it was important to evaluate here the influence of H_2 or CO_2 on the catalyst activity and stability, because reformate and coal gas streams contain both of these gases. Of particular interest for a H₂-selective membrane reactor is the effect of CO_2 , since the continuous H₂ removal through the membrane will create a CO_2 -rich condition over the shift catalyst.

Figure 9 shows the performance of 10at%Cu-C10L, C12-4-02 and 13at%Fe-C10L in the H₂-rich gas mixture of 2%CO-10%H₂O-35%H₂-He at 450°C, 0.09g•s/cc. For 10 hours the CO conversion kept close to the equilibrium conversion on all three catalysts, and no methane was detected. However, as shown in Figure 10, in a CO₂- rich gas (2%CO-10%H₂O-35%CO₂-He) at the same temperature and contact time, only the CO conversion on 10at%Cu-C10L stayed close to the equilibrium value, while the CO conversions on the other two catalysts dropped to much lower levels.

Figure 11 shows the WGS performance of 10at%Cu-C10L in three gas mixtures with different contents of CO_2 (10%CO-16%H₂O-He, 10%CO-16%H₂-3.4%CO₂-He and 10%CO-16%H₂O-64%CO₂-He). The presence of CO_2 inhibits the reaction on 10at%Cu-C10L, but unlike iron oxide, severe CO_2 inhibition does not occur on this catalyst. On iron oxide-based catalysts, Carl Lund's group⁹ has reported a dramatic poisoning effect by CO_2 , whereby the reaction rate decreased by several orders of magnitude in CO_2 -rich gas.

5. Hydrogen permeation tests

Hydrogen permeation through pure palladium foil, palladium-copper and palladium-silver alloy foils has been investigated at different operating conditions in the membrane reactor system described above.

The effect of temperature on the hydrogen flux (J) (hydrogen permeation) through pure Pd, $Pd_{60}Cu_{40}$ and $Pd_{75}Ag_{25}$ alloy membranes has been studied experimentally in the temperature range from 250°C to 500°C at upstream pressure 50 psig (3.4 atm) and permeate hydrogen pressure of 1 atm. Hydrogen permeability as a function of temperature for the studied membranes is presented in Fig.12. The points are calculated from the average permeability values for all membrane samples at each temperature. Hydrogen flux through the palladium-silver membranes is higher than for pure palladium and palladium-copper samples. Flux decay is observed for the palladium-copper membrane with increasing temperature. These

results are in good agreement with data in the literature (US Patent, 3,439,474). A direct comparison of our results of hydrogen permeability for a 25μ m palladium-copper membrane with those in the literature is shown in Fig. 13.

Conclusions/Future Work Plans

The first-year screening studies of WGS catalysts have identified Cu-ceria as the most promising shift catalyst for integration with H₂-selective membranes. The specific formulation 10at%Cu-C10L is both highly active and stable in operation at 450° C and in CO₂-rich or H₂-rich gas streams. Further detailed evaluation of this type catalyst will be performed in the second year of the project, including reaction kinetics, effect of H₂S adsorption, and structural analysis after exposure to various reaction gases. A larger flat- membrane (10cm x 5cm oval) reactor will be used for H₂ permeation tests from synthetic coal gas mixtures. Preparation of catalysts on screens to be used in the membrane reactor will also take place during the second year of the work. Global reaction rate measurements will be made using the flat-membrane reactor and the modeling effort will be underway.

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Table 1. Specific surface area of catalysts[#]

Sample	BET surface area (m ² /g)		
C10L	98		
5at%Cu-C10L	107		
10at%Cu-C10L	124		
15at%Cu-C10L	85		
40at%Cu-C10L	66		
6.2at%Fe-C10L	109		
13at%Fe-C10L	98		
20at%Fe-C10L	86		
29at%Fe-C10L	80		
50at%Fe-C10L	76		
C12-4-02*	96		

all catalysts were prepared by the UGC method; calcined at 650° C, 4h * used in the as-received form, after crushing to < 150 μ m -size particles

Sample	Pea	Peaks above		
	Temperature (°C)	Total H_2 consumption (mmol H_2/g)	H_2 consumption for reduction of ceria only [#]	500°C Temperature (°C)
	((mol H ₂ /molCe)		
C10L	408	0.560	0.106	766
1at%Cu-C10L	135, 302	0.539	0.092	N/A
5at%Cu-C10L	126, 132, 145(small)	0.737	0.085	N/A
10at%Cu-C10L	96, 103, 129(small), 164(small)	1.156	0.108	N/A
15at%Cu-C10L	90, 121(small), 134	1.340	0.080	N/A
40at%Cu-C10L	94, 154(shoulder), 195	3.414	0.078	766
6.2at%Fe-C10L	123, 367			N/A
13at%Fe-C10L	119, 359			641, 766
29at%Fe-C10L	362			616,
				~790 (shoulder)
C12-4-02	197, 223, 350			
5wt%CeO ₂ -(C12- 4-02)	160, 199, 312			

Table 2. H₂-TPR: Peak temperatures and areas (H₂ consumption)

N/A: tests stopped at lower temperatures.

Some peak areas are not calculated because of overlap; deconvolution would not be accurate.

 $^{\scriptscriptstyle\#}$ after subtraction of the H_2 consumed by CuO



Figure 1. H₂-TPR of Cu-Ce(10at%La)O_x samples, all calcined at 650°C



Figure 2. H₂-TPR of Fe-Ce(10at%La)O_x samples, all calcined at 650°C



Figure 3. WGS activity of Cu- and Fe-Ce(10at%La)O_x in 2%CO-10%H₂O-He $0.09g\bullet$ s/cc, S.V.=80,000 h⁻¹ (except C12-4-02)



Figure 4. H₂-TPR of 13at%Fe-Ce(10at%La)O_x and C12-4-02







Figure 6. Effect of ceria addition on the WGS activity of C12-4-02, 10%CO-16%H₂O-He, 0.09g•s/cc



 $\label{eq:Figure 7. WGS activity in a simulated coal-gas composition, $$10\%CO-34\%H_2O-10\%CO_2-15\%H_2-He, 0.09g•s/cc, $$S.V.= 80,000 h^{-1} (except C12-4-02)$}$



Figure 8. WGS activity / stability in a simulated coal-gas composition, 10% CO-34%H₂O-10%CO₂-15%H₂-He, 0.09g•s/cc, 450°C, S.V.= 80,000 h⁻¹ (except C12-4-02)



 $\begin{array}{l} \mbox{Figure 9. WGS activity / stability of Cu- and Fe-Ce(10at\%La)O_x in H_2-rich gas $2\%CO-10\%H_2O-35\%H_2$-He, 0.09gs/cc, $S.V.= 80,000 h^{-1} (except C12-4-02, 48,000 h^{-1}) $ \end{array}$



Figure 10. WGS activity / stability of Cu- and Fe-Ce(10at%La)O_x in CO₂-rich gas, 2%CO-10%H₂O-35%CO₂-He, 0.09gs/cc, 450 °C



Figure 11. CO₂ effect on the WGS Activity of 10at%Cu-C10L, 10%CO-16%H₂O-x%CO₂-balance He, 0.09g*s/ml, S.V.= 80,000h⁻¹



Figure 12. Hydrogen Permeation through Pd and Pd-alloy membranes (p₁= 3.4 atm, p₂ =1 atm, L_{membrane}= 25 μ m)

Figure 13. Hydrogen permeation through Pd-Cu membranes, 25 μm

