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APPENDICES

EXPERIMENTAL DATA

APPENDIX A: Gas Chromatograph Calibration

Due to the detector could not detected hydrogen signal in range of our research (less than 4% for our scope in LTWGS). The calibrations had been done for all composition in WGS reaction before the experiments performed. This calibration was not reprogrammed with calibration table in gas chromatograph (GC) software, it is an external calibration determined from actual inlet flow and the calculated area in GC. The Calibration curve was plotted. The way to get the area from the result was changed from auto-integrated to hand drawing baseline integrated instead.

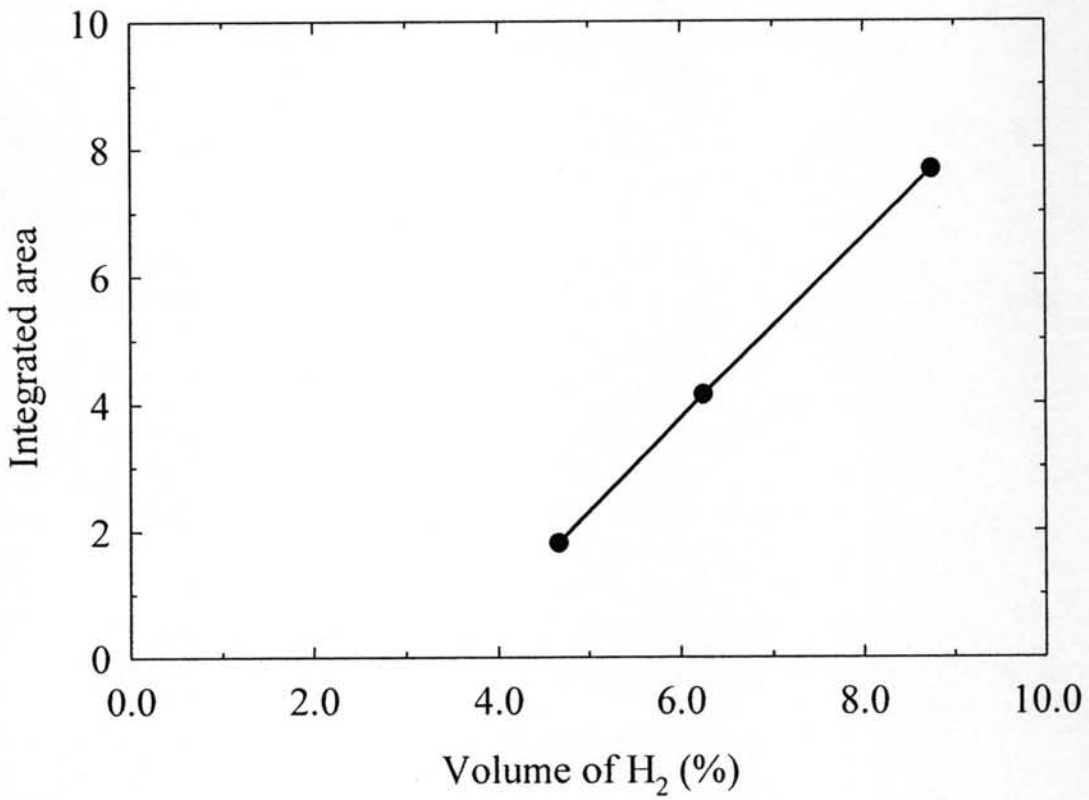

Figure A.1 Calibration curve of hydrogen gas with total flow 80 ml/min.

Fig. A.1 showed the calibration curve of H_2 diluted in helium before passing through the GC. The concentration of H_2 is in the range of 4-9%. For CO calibration, Fig. A.2 shows the calibration curve of CO in the range of 2-8%. The 10% CO/He was also diluted with helium to desired concentration.

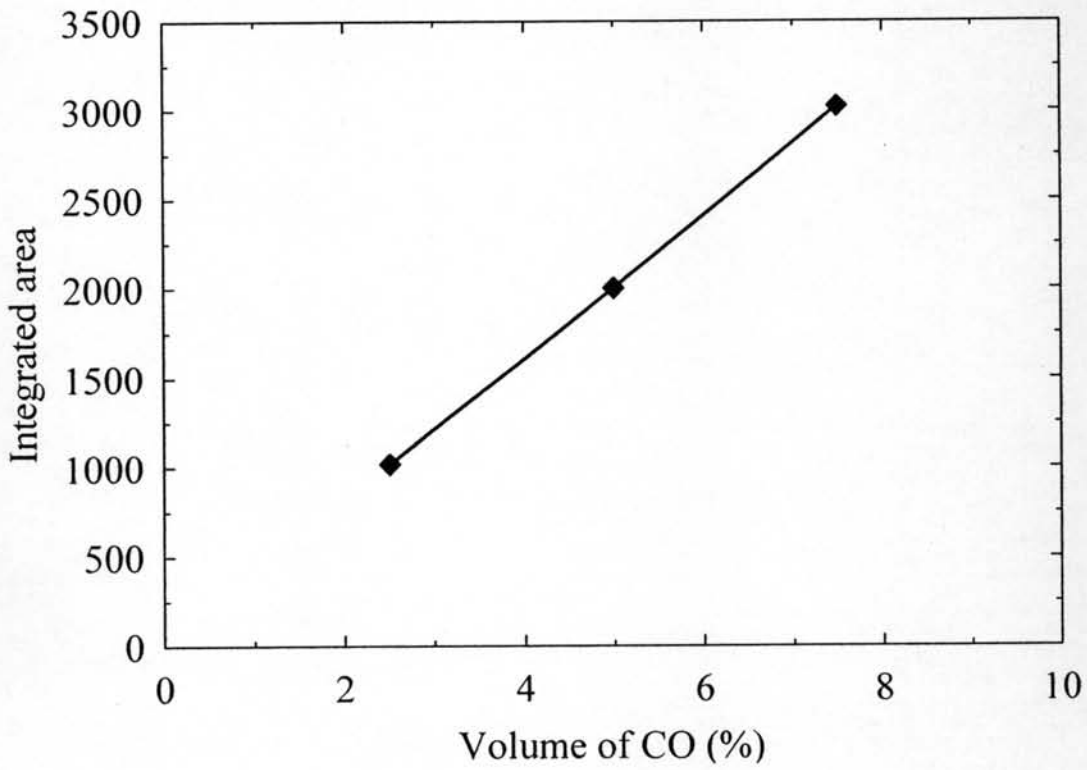

Figure A.2 Calibration curve of carbon monoxide gas with total flow 80 ml/min.

APPENDIX B: Moisture Calculation

Amount of steam used in this work was calculated from moisture conversion table from Universal Analyzer Inc., Nevada. Table A.1 shows moisture conversion as a function of temperature.

Table A.1 Moisture conversion table

DEW POINT Degree Celsius	% WATER BY VOLUME AT SATURATION
-100	0.0000014
-50	0.00388
-30	0.0347
-20	0.102
-10	0.256
-5	0.396
-4	0.432
-3	0.469
-2	0.510
-1	0.555
0	0.602
1	0.649
2	0.696
3	0.750
4	0.803
5	0.861
6	0.922
7	0.988
8	1.06
9	1.13
10	1.21
11	1.29
12	1.38
13	1.48
14	1.58
15	1.68
20	2.31
25	3.13
30	4.19
35	5.55
40	7.28

DEW POINT Degree Celsius	% WATER BY VOLUME AT SATURATION
45	9.46
50	12.2
55	15.5
60	19.7
65	24.4
70	30.7
75	38.0
80	46.7
85	57.2
90	69.2

In order to generate the desired amount of water in the feed steam, the temperature of water was set at the point that there is a desired amount of moisture in air. The flow rate of carrier gas was also determined too.

For example, 4% vol. of steam in total flow rate, 90 ml/min (4% CO balanced with Helium) can be determined from equation A.1.

There are 2 flows of reactant for 90 ml/in total flow

- 36 ml/min of 10%CO in Helium, 3.6 ml/min of CO (to get 4% of CO)
- 54 ml/min of Helium as a carrier gas

The desired percent of steam is 4% which is also 3.6 ml/min as same as amount of CO to get 1/1 ratio of water/CO.

Equation A.1

$$Volume\ of\ steam\ in\ carrier\ gas = Flow\ rate\ of\ carrier\ gas \times \frac{\%\ water\ by\ volume}{100}$$

$$3.6\ \frac{ml}{\min} = 54\ \frac{ml}{\min} \times \frac{\%water\ by\ volume}{100}$$

$$\%water\ by\ volume = 6.67\ \%$$

At 6.67% of water by volume, the temperature is ~ 38° C

APPENDIX C: Particle size Calculation

The Scherrer's formula (3.3) relates crystal size to line width;

$$t = K * \lambda / \beta \cos \theta$$

In which

t is a measure for the dimension of the particle in the direction perpendicular to reflecting plane;

λ is the X-ray wavelength,1.5406 nm;

β is the peak width (FWHM in radians);

θ is the angle between the beam and normal on the reflecting plane; and

K is a constant (often taken as 0.94)

Table A.2 Calculated crystallite size of Au from 2% Au/CeO_2 catalyst compared with commercial CeO_2 support from XRD technique table

Sample	FWHM (in radi-ans,β)	Plane,2θ	t (Particle size in Å) ($K \times \lambda / \beta\cos\theta$)	Particle Size in nm.
2% Au/CeO_2 (LS)	0.00164	[111], 2θ = 38.20	894.71	89.47
	-	[200], 2θ = no peak	-	-
2% Au/CeO_2 (HS)	0.00246	[111], 2θ = 38.20	596.47	59.64
	-	[200], 2θ = no peak	-	-
5% Au/CeO_2 (LS)	0.00206	[111], 2θ = 38.62	713.20	71.32
	0.00206	[200], 2θ = 44.26	726.60	72.66

Table A.3 Calculated crystallite size of CeO_2 from 2% Au/CeO_2 catalyst compared with commercial CeO_2 support from XRD technique table

Sample	FWHM (in radians,β)	Plane,2θ	t (Particle size in Å) $(K \times \lambda / \beta\cos\theta)$	Particle Size in nm.
CeO_2 low surface	0.00328	[111], 2θ = 28.64	436.28	43.62
	0.00246	[220], 2θ = 47.58	615.97	61.59
CeO_2 high surface	0.00410	[111], 2θ = 28.60	348.99	34.89
	0.00452	[220], 2θ = 47.48	335.11	33.51
1% Au/CeO_2 (LS)	0.00328	[111], 2θ = 28.62	436.26	43.62
	0.00328	[220], 2θ = 47.54	461.91	46.19
2% Au/CeO_2 (LS)	0.00328	[111], 2θ = 28.62	436.26	43.62
	0.00287	[220], 2θ = 47.56	527.94	52.79
2% Au/CeO_2 (HS)	0.00452	[111], 2θ = 28.56	316.54	31.65
	0.00492	[220], 2θ = 47.52	307.92	30.79
5% Au/CeO_2 (LS)	0.00328	[111], 2θ = 28.54	436.18	40.797
	0.00246	[220], 2θ = 47.48	615.74	41.234

APPENDIX D: Catalytic Activity of Commercial Catalyst

High-Temperature Water-Gas Shift Activity

0.9 g (0.148-0.224 mm) of Shiftmax 120 was used. The total flow rate was 75 ml/min, 60 ml/min of 10% CO/He and 15 ml/min of helium (to get 8% of CO) at the temperature range of 300-450° C.

Table A.4 HT-WGS activities of Shiftmax 120 as a function of reaction temperature

Temperature (°C)	CO conversion (%)
300	36.80
350	46.47
400	51.44
450	60.87

Low-Temperature Water-Gas Shift Activity

0.16 g (0.148-0.224 mm) of Shiftmax 230 was used. The total flow rate was 80 ml/min, 32 ml/min of 10% CO/He and 48 ml/min of helium (to get 4% of CO) at the temperature range of 100-325° C.

Table A.5 LT-WGS activities of Shiftmax 230 as a function of reaction temperature

Temperature (°C)	CO conversion (%)
100	3.32
125	4.14
150	10.43
175	67.63
200	73.58
225	79.92
250	82.16
275	80.62
300	82.40
325	74.92

APPENDIX E: CO Conversion of Prepared Au/CeO_2 Catalysts

Effect of Space Velocity

0.45 g (0.148-0.224 mm) of 2% Au/CeO_2-LS was used at varied space velocities on LT-WGS at the temperature range of 100-350° C. The total flow rates were varied from 90, 60, and 30 ml/min for space velocity (SV) 12,000, 8,000, and 4,000 $ml \cdot h^{-1} \cdot g^{-1}$ with same amount of catalyst. 10% CO/He was used and the flow rate of 10% CO/He were 36, 24, and 12 ml/min (to get 4% of CO). For SV = 30,000 $ml \cdot h^{-1} \cdot g^{-1}$, the flow rate condition was used as same as from the LT-WGS reaction of commercial catalyst.

Table A.6 LT-WGS activities of 2% Au/CeO_2 catalyst at different space velocities as a function of the reaction temperature

Temperature, °C	CO Conversion, %			
	SV = 4,000	SV = 8,000	SV = 12,000	SV = 30,000
100	1.51	1.14	1.45	2.30
150	4.69	3.35	2.47	2.36
200	12.31	6.99	3.83	2.46
250	19.87	11.69	6.19	2.91
275	-	-	7.27	-
300	29.99	19.95	8.35	3.82
350	35.00	29.09	12.53	5.93

Effect of Gold Loading

0.45 g (0.148-0.224 mm) of 1%, 2%, 5% Au/CeO_2-LS, and commercial CeO_2 support were used on LT-WGS at the temperature range of 100-350 °C. The total flow rate was 30 ml/min at SV = 4,000 $ml \cdot h^{-1} \cdot g^{-1}$ with same amount of catalyst. 10% CO/He was used and the flow rate of 10% CO/He were 12 ml/min (to get 4% of CO).

Table A.7 LTS activities of 1% and 2% Au/CeO_2 as a function of different temperature

Temperature, °C	CO conversion, %			
	CeO_2-LS	1% Au/CeO_2	2% Au/CeO_2	5% Au/CeO_2
100	0.58	0.95	1.51	2.42
150	0.77	2.13	4.69	5.64
200	1.10	4.30	12.31	10.87
250	1.67	8.57	19.87	16.86
275	-	11.13	-	-
300	1.98	13.80	29.99	24.27
325	-	19.21	-	-
350	2.10	25.06	35.00	35.56

Effect of H_2-Pretreatment

0.45 g (0.148-0.224 mm) of H_2 pretreated and unpretreated 2% Au/CeO_2-LS were used on LT-WGS at the temperature range of 100-350° C. The total flow rate was 30 ml/min at SV = 4,000 $ml \cdot h^{-1} \cdot g^{-1}$ with same amount of catalyst. 10% CO/He was used and the flow rate of 10% CO/He were 12 ml/min (to get 4% of CO).

Table A.8 LT-WGS activities of pretreated 2% Au/CeO_2 and 2% Au/CeO_2-LS as a function of temperature

Temperature, °C	CO conversion, %	
	Pretreated 2% Au/CeO_2-LS	2% Au/CeO_2-LS
100	2.13	1.51
150	7.13	4.69
200	17.45	12.31
250	25.29	19.87
300	33.50	29.99
350	37.38	35.00

Effect of Type of CeO_2

Two type of commercial ceria supports, micro-powder and nano-particle ceria, were examined with the same amount of gold loading by DP method. The prepared catalysts were performed in the same LT-WGS reaction condition without any pretreatment. 0.45 g (0.148-0.224 mm) of 2% Au/CeO_2-HS and 2% Au/CeO_2-LS were used on LT-WGS at the temperature range of 100-350° C at SV = 4,000 ml g^{-1} h^{-1}. The reaction composition was 4% CO and 4% H_2O balanced with helium.

Table A.9 LT-WGS activities of 2% Au/CeO_2-HS and 2% Au/CeO_2-LS compared with commercial supports as a function of temperature

Temperature, °C	CO conversion, %			
	2% Au/CeO_2-HS	2% Au/CeO_2-LS	CeO_2-LS	CeO_2-HS
100	5.72	1.51	0.58	3.56
150	14.48	4.69	0.77	5.52
200	21.20	12.31	1.10	7.91
250	32.71	19.87	1.67	8.23
300	43.97	29.99	1.98	8.44
350	55.69	35.00	2.10	8.65

Effect of Water/CO ratio

0.45 g (0.148-0.224 mm) of 2% gold-ceria-HS was used to study the effect of water/CO ratio without any pretreatment at SV = 4,000 ml g^{-1} h^{-1}. The reaction composition was 4% CO and with varied % of H_2O (12%, 4%, 0%) balanced with helium.

Table A.10 LT-WGS activities of 2% Au/CeO_2 at different water/CO ratio as a function of temperature

Temperature, °C	CO conversion, %		
	Water/CO = 0/1	Water/CO = 1/1	Water/CO = 3/1
100	3.83	5.72	3.55
150	7.81	14.48	5.93
200	12.42	21.20	16.65
247	-	32.71	-
250	12.71	-	28.96
299	-	43.97	-
300	11.75	-	44.02
349	-	55.69	-
350	5.79	-	59.32

Effect of H_2-Mixing

0.45 g (0.148-0.224 mm) of 2% gold-ceria-HS catalyst was used without any pretreatment at SV = 4,000 ml g^{-1} h^{-1}. The reaction composition was 4% CO, 4% H_2O and 60% H_2 balanced with helium.

Table A.11 LT-WGS activities of 2% Au/CeO_2 at different water/CO ratio as a function of temperature

Temperature, °C	CO conversion, %	
	Ideal reactant	60% H_2 mixing in reactant
100	5.72	1.33
150	14.48	1.62
200	21.20	4.04
250	32.71	6.74
300	43.97	8.21
350	55.69	15.74

Stability of prepared Au/CeO_2 catalyst

The stability of 2% Au/CeO_2-HS catalyst (0.45 g with size 0.148-0.224 mm) was performed in LT-WGS at 4,000 ml g^{-1} h^{-1}. The reaction was performed at 250 °C for 500 minutes. The composition of reactant was 4% CO and 4% H_2O diluted with helium.

Table A.12 LT-WGS activities of 2% Au/CeO_2 at different water/CO ratio as a function of temperature

Time on stream, min	CO conversion, %	Time on stream, min	CO conversion, %
0	42.00	250	29.09
10	40.61	260	29.00
20	38.92	270	29.18
30	37.70	280	29.76
40	37.13	290	29.37
50	36.65	300	29.11
60	36.61	310	29.47
70	35.92	320	29.14
80	35.64	330	29.85
90	35.01	340	29.19
100	34.71	350	29.28
110	34.45	360	29.51
120	34.10	370	29.80
130	32.57	380	30.35
140	32.20	390	29.90
150	31.99	400	30.11
160	31.80	410	30.51
170	32.32	420	30.74
180	32.04	430	31.04
190	31.25	440	30.78
200	30.81	450	30.44
210	30.86	460	30.53
220	29.40	470	30.99
230	28.71	480	30.39
240	29.64	490	30.28

APPENDIX F: Bench Scale Experiment

Both prepared Au/CeO_2-HS and commercial catalyst were performed in bench scale experiment. The experimental data was shown below.

CO Conversion of LT-WGS Activity

In bench scale, 4.5 g of catalyst was used with 20-60 mesh. The space velocity was set to ~ 4,000 ml g^{-1} h^{-1}. The LT-WGS reaction was performed at 250 °C for ~500 minutes.

Table A.13 Bench-scale LT-WGS activities of Shiftmax 230 catalyst as a function of temperature

Time on stream, hrs	Inlet CO concentration, ppm	Outlet CO concentration, ppm	CO conversion, %
2.00	42191.38	4574.81	89.16
2.35	34564.17	7315.04	78.84
3.15	35357.98	4323.64	87.77
4.00	34775.33	4344.99	87.51
4.45	47227.00	3736.03	92.09
5.30	41924.52	5440.06	87.02
6.15	40488.62	2745.57	93.22
7.25	46010.49	2804.29	93.91
8.15	37380.63	1948.29	94.79

Table A.14 Bench-scale LT-WGS activities of 2% Au/CeO_2-HS catalyst as a function of temperature

Time on stream, hrs	Inlet CO concentration, ppm	Outlet CO concentration, ppm	CO conversion, %
3.00	40216.69	19796.28	50.78
4.50	38476.49	21442.38	44.27
6.00	43632.03	21731.80	50.19
6.50	42722.73	19953.16	53.29
7.00	41839.67	21559.71	48.47
8.00	39903.48	22796.28	42.87

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