CHAPTER 5

Experimental Procedure

5.1 Experimental Procedure

Five palladium test beds are employed in this experiment shown in Figure 5.1. Each of the test beds is loaded to a different pressure, for example, in the first data set, shown in Appendix A, the test beds were loaded at 20°C, to 667, 2502, 4022, 6340, and 10303 psia. The temperature of the environmental chamber was set to 20°C, and the test beds were allowed to equilibrate to this temperature.

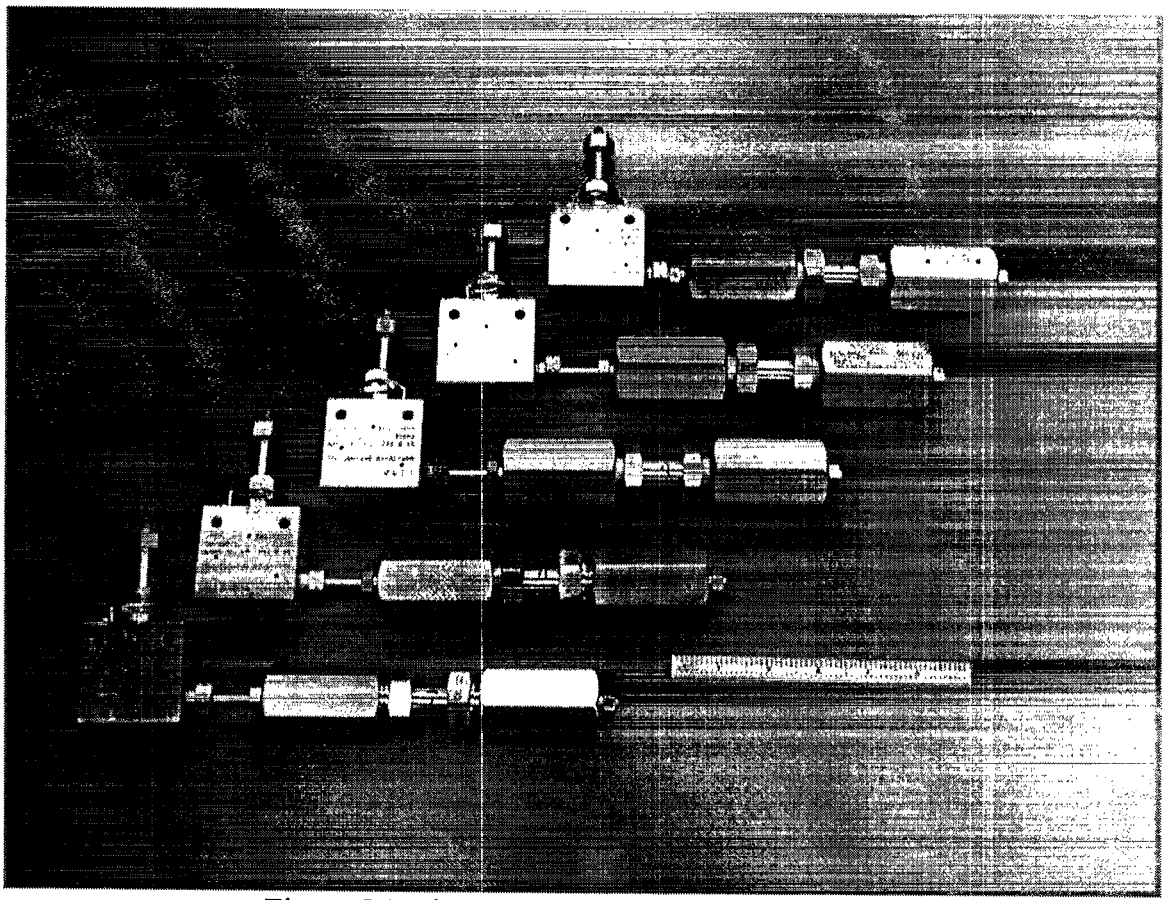

Figure 5.1 Five Beds Utilized in the Experiment.

Five different test beds are being utilized in this experiment, shown in Figure 5.1. The five beds will be achieve equilibrium at 20°C. Approximately 50 psia of hydrogen is

in the high-pressure manifold. The environmental chamber will be opened and the five beds will be opened with a half turn of the AE 30,000 psia valves. This is the beginning of the loading of the beds.

A calculation of the predicted pressures is done using the Fortran program Fill2, shown in the Appendix B, (Meyer, 1996). The inputs for the program are the moles of hydrogen, the amount of palladium in grams, the free volume of the un-hydrided unit, and the ambient temperature. The program can predict the pressure and the fugacity of the bed at different temperatures. When the maximum pressure at the highest temperature is needed a calculation of the initial loading pressure at 20°C is calculated.

5.2 Pressure Loading of the Beds

The initial loading of the each of the beds is at a different pressure. All of the beds are then controlled to an equilibrium baseline temperature of 20°C. The temperature and pressure are monitored every 15 minutes until equilibrium is reached. With this experimental setup the beds reach equilibrium in approximately 1 hour, the actual time for equilibration is shown in Appendix A. The baseline temperature, outside temperature of the outside valve and pressure are taken for each of the beds after equilibrium is reached. The temperature of the environmental chamber is then changed to another temperature and the process is repeated. In the first three P-C-T experiments the temperature of the baseline was 20°C. Then the beds were cycled through the temperatures 0°C, -20°C, -40°C, -60°C, 20°C, 40°C, 60°C, 80°C, 100°C, 120°C, 20°C and 20°C. Between the last two 20°C temperatures the chamber door is opened and the valve directly next to the palladium test beds is closed. When this valve is closed the

pressures within this now closed system change because of the movement of the valve seat slightly changes the internal volume. This volume change is on the order of ≈0.05 to 0.1 cm^3. At the 10,000 psia this change in volume increases the pressure by 35 psia. It is important to note that closing the valve will change the pressure in the palladium test bed.

5.3 Bed Weighing Procedure

When the test beds equilibrate at the last temperature the valves directly next to the beds are closed. When closed the last pressure and temperature data points are taken. Each of the beds is then individually disconnected from the high-pressure manifold, shown in Figure 5.2.

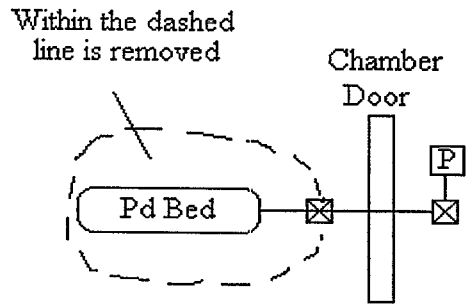

Figure 5.2. Diagram Showing Disconnection of Test Bed.

The beds are weighed to determine the amount of hydrogen within the bed. When handling a test beds, latex gloves are worn to minimize the possibility of adding contaminants to the beds. When measuring the weights one person is weighing the beds and another person is writing the weight down and verifying the correct weights are taken. The mass balance is first calibrated using a 2000.000gm weight. After the calibration the 2000gm standard weight is measured 3 times to insure that the balance is reading correctly. The standard weight must be repeatable to ±0.002 gm. The bed is then

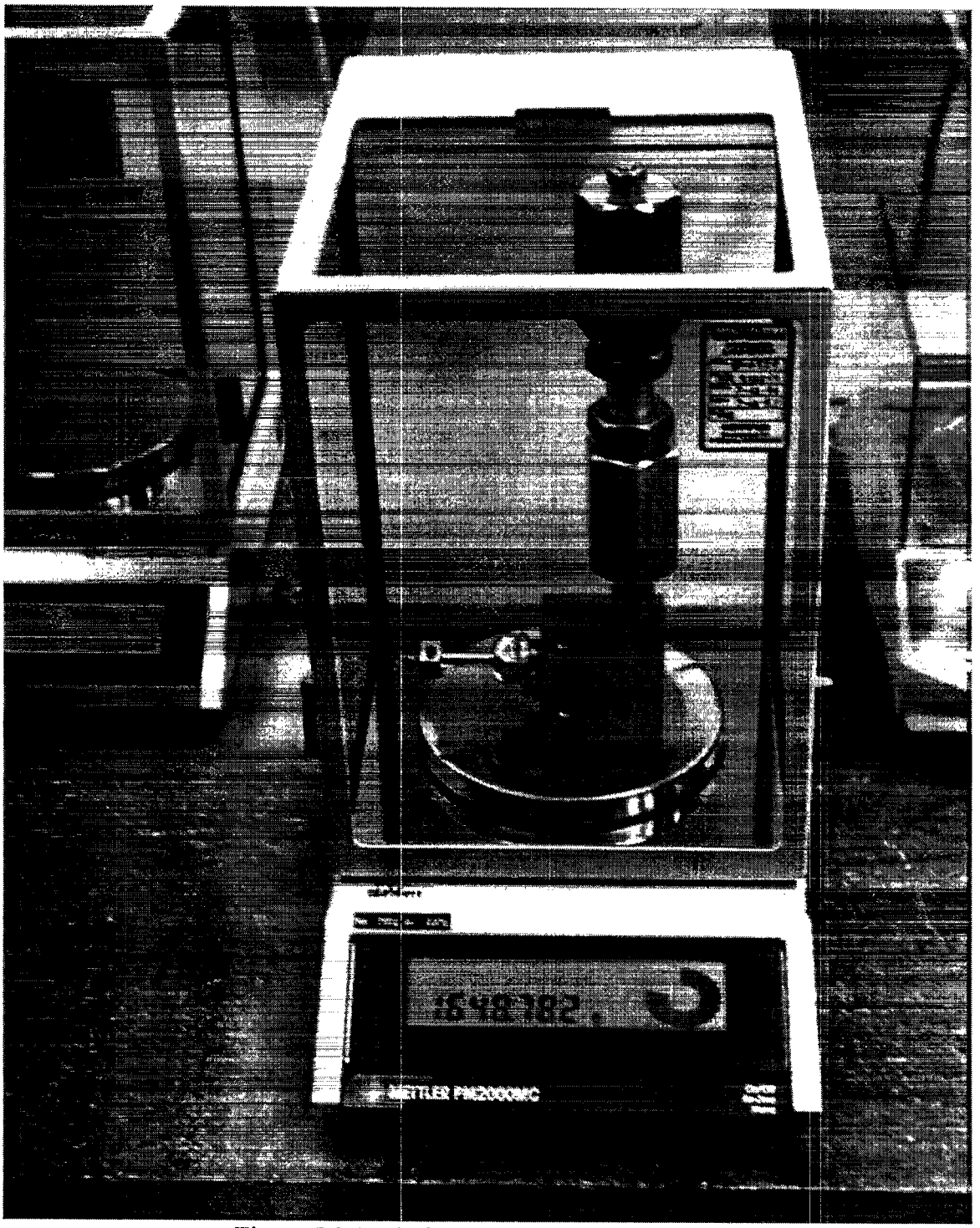

Figure 5.3 Bed Placement on the Balance Pan.

placed on the balance with special consideration to place the center of gravity of the bed in the center of the balance pan. The placement of a test bed is shown in a photograph in Figure 5.3.

The bed weight is then taken three times to insure that the test bed weight is within the 0.001 gm tolerance. After the three weights are taken the standard weight is then placed on the balance and weighed. The 2000gm standard weight traceable to NIST is weighed and must be within the ±0.002gm of the initial weight. Each bed is weighed using this procedure.

CHAPTER 6

Results and Discussion

6.1 Calculation of the Fugacity and Compressibility Factor

The fugacity-composition-temperature curves describe the equation of state for the palladium hydrogen system. The fugacity is calculated from the following integral equations:

$$\ln\left(\frac{f}{P'}\right) = \int_0^{P'} \frac{Z-1}{P}\,dP \tag{6-1}$$

where the compressibility factor, Z is defined by

$$Z \equiv \frac{Pv}{RT} \tag{6-2}$$

It is more convienient to write fugacity in terms of fugacity coefficient, Φ as:

$$\ln \Phi = \int_0^{Ptot} (Z-1)d\ln P \tag{6-3}$$

where

$$f \equiv \Phi P \tag{6-4}$$

The fugacity coefficient with a conversion to atmospheres becomes:

$$\Phi = e^{\left((C0+C1T+C2T^2)(\frac{P}{T})+(C3+C4T+C5T^2)\frac{1}{2}(\frac{P}{T})^2+(C6+C7T+C8T^2)\frac{1}{3}(\frac{P}{T})^3+(C9+C1T+C11T^2)\frac{1}{4}(\frac{P}{T})^4\right)} \tag{6-5}$$

Where C0, C1, C2, C3, C4, C5, C6, C7, C8, C9, C10, C11 are compressibility constants for the compressibility factor Z, pressure P is in atmospheres. The compressibility factor Z is in the form of the equation:

$$Z = 1 + (C0 + C1 * T + C3 * T^2)(\frac{P}{T}) + (C4 + C5 * T + C6 * T^2)(\frac{P}{T})^2 + (C7 + C8 * T + C9 * T^2)(\frac{P}{T})^3 + (C10 + C11 * T + C12 * T^2)(\frac{P}{T})^4 \quad (6\text{-}6)$$

The compressibility constants used in equations (6-5) and (6-6) are in Table 6.1, from (Meyer, 1996).

Table 6.1 Compressibility Constants Used in Compressibility Factor Calculation

C1=0.022456	C2=8.3057e-4	C3=-1.0193e-6
C4=0.056181	C5=-1.9111e-4	C6=1.5657e-7
C7=-0.0036149	C8=-8.1655e-6	C9=3.0139e-8
C10=-1.5121e-4	C11=2.7545e-6	C12=-4.6721e-9

6.2 Calculation of the Atomic Ratio

The moles of hydrogen within the palladium lattice (palladium solid) are calculated. The weight of the hydrogen measured on the mass balance is the physical property used in the calculation to determine the atomic ratio, H/Pd. Figure 6-1 describes the control volumes for the conservation of mass in the calculation of the atomic ratio.

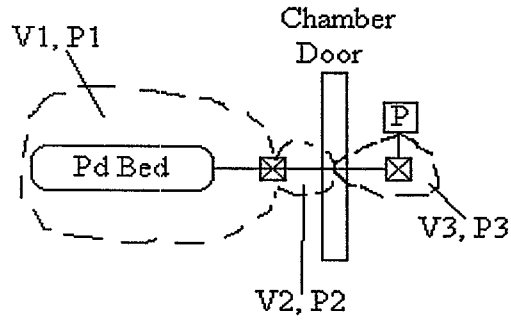

Figure 6.1 Diagram for the Calculation of the Atomic Ratio, H/Pd.

Equations (6-7) through (6-14) are used to determine how the actual weight measurements of the hydrogen are included in the calculation of the atomic ratio.

The moles of the hydrogen within the three dashed line volumes of Figure 6.1 are constant. Thus, equation (6-7) is true:

$$mol_1+mol_2+mol_3=constant \tag{6-7}$$

the moles of hydrogen in volume V2 are:

$$mol_2 = \frac{P_2V_2}{Z_2RT_2} \tag{6-8}$$

and the moles of hydrogen in volume V3 are:

$$mol_3 = \frac{P_3V_3}{Z_3RT_3} \tag{6-9}$$

In equation (6-7) "constant" is used for the total moles of hydrogen in the system, which remains constant throughout the experiment. Thus mol_1 is the moles of the hydrogen in the palladium solid plus the moles of the gaseous hydrogen in the volume 1 of the test bed. Equation (6-7), using (6-8) and (6-9) becomes equation (6-10), with mol_1 being split into two components. The two components of mol_1 are $mol_{H2\ solid}$ and the moles of the gaseous hydrogen, equation (6-7) becomes, at a specified temperature of 293.15K, equation (6-10):

$$constant = (mol_{H2\ solid} + \frac{P_1V_1}{Z_1R(293.15K)}) + \frac{P_2V_2}{Z_2R(293.15K)} + \frac{P_3V_3}{Z_3R(293.15K)} \tag{6-10}$$

$mol_{H2\ solid}$ is the moles of hydrogen in the palladium solid. P is the pressure for each respective volume V. The "constant" is determined at a specific temperature. R is the universal gas constant. The weight measured in grams at 293.15°K divided by the molecular weight of hydrogen is equation (6-11):

$$\frac{\text{weight measured at 293.15K}}{MW_{H2}} = (\text{mol}_{H2\ solid} + \frac{P_1V_1}{Z_1R(293.15K)}) \tag{6-11}$$

The constant in equation (6-10) using (6-11) becomes equation (6-12):

$$\text{constant} = \frac{\text{weight measured at 293.15K}}{MW_{H2}} + P(\frac{V_2}{Z_2R(293.15)} + \frac{V_3}{Z_3R(293.15)}) \tag{6-12}$$

The moles of the hydrogen in the solid the can be found with the measured weight of the hydrogen, and the pressure, volume, and temperature inputs using equations (6-11) and (6-12) combined to form (6-13):

$$\text{mol}_{H2\ solid} = \frac{\text{weight measured 293.13K}}{MW_{H2}} + P(\frac{V_2}{Z_2R(293.15K)} + \frac{V_3}{Z_3R(293.15K)}) - P(\frac{V_1}{Z_1RT_1} + \frac{V_2}{Z_2RT_2} + \frac{V_3}{Z_3RT_3}) \tag{6-13}$$

It must be noted that the V_1 is the volume of the dead space within the bed. As the palladium accepts hydrogen into the lattice, the palladium swells, thus reducing the volume of V_1. The reduction in the volume, V_1 is a direct function of the atomic ratio, H/Pd. Equation (6-14) is for the reduction in free volume due to the swelling of the palladium from (Wicke and Brodowsky, 1978).

$$\text{CorrectedVolume} = V_1 - \frac{(\text{Pd gm})(1.10777*(1+0.044*(H/Pd-0.607))^3 - 1}{12.02\frac{\text{gm Pd}}{\text{cm}^3}} \tag{6-14}$$

With equations (6-7) to (6-14) the atomic ratio can be calculated using the experimental weight measurement, the pressure, and temperature, and the known amount of palladium with equation (6-15):

$$\frac{H}{Pd} = \frac{2*\text{moles H}_2}{\text{moles Pd}} \tag{6-15}$$

The results of the experimentally measured atomic ratio are tabulated in the excel spreadsheet in Appendix A. The experimentally measured atomic ratio is in the plots of fugacity versus atomic ratio in Figures 6.2 and 6.3.

6.3 Plots of Fugacity Versus Atomic Ratio

The fugacity and the atomic ratio, H/Pd, can now be determined from the measured experimental data. The experimental data points for the temperatures -60°C, -40°C, -20°C, 0°C, 40°C, 60°C, 80°C, 100°C, and 120°C is plotted with the fugacity in psia versus the atomic ratio, H/Pd, in Figure 6.2. The curve fit to the data is also shown in Figure 6.2.

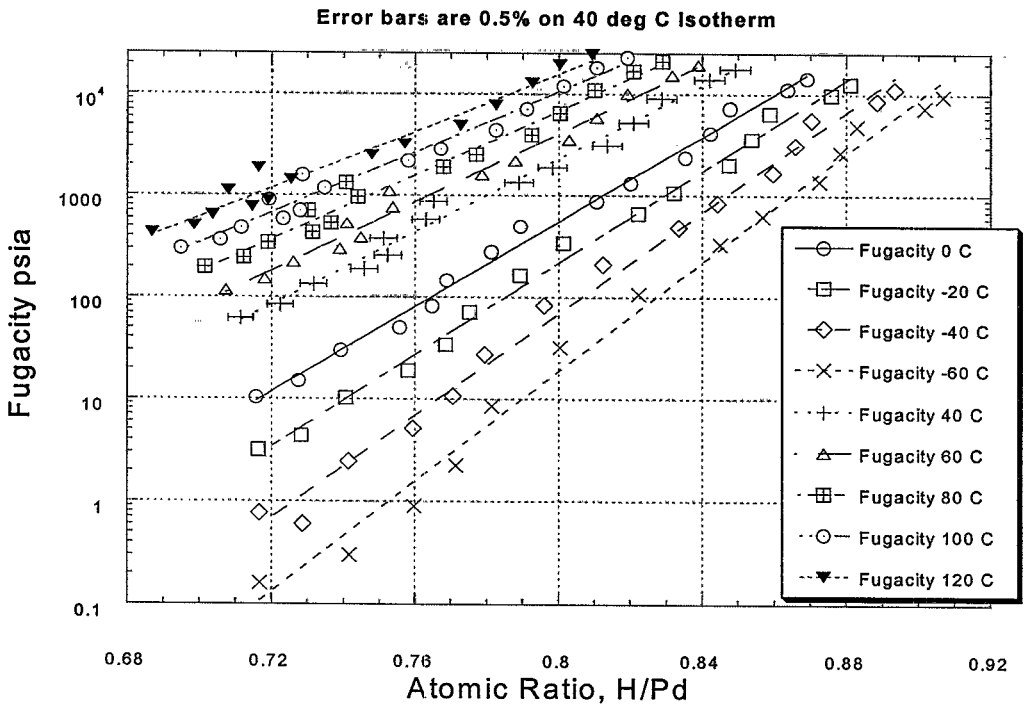

Figure 6.2 Experimental Data of Fugacity versus Atomic Ratio, H/Pd.

The experimental data points and the curve fit for the 20°C isotherm are shown in Figure 6.3. The error bars at 40°C and 20°C represent the 0.5% error on the atomic ratio axis.

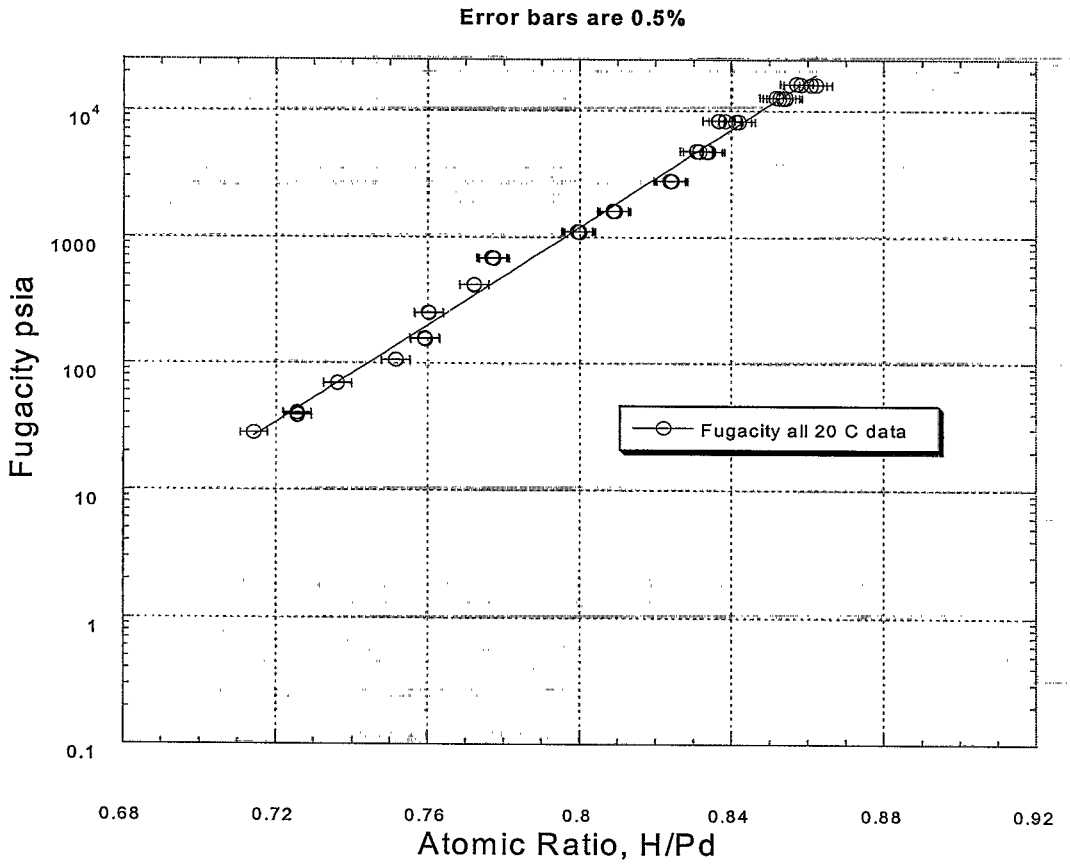

Figure 6.3 Experimental Data of Fugacity versus Atomic Ratio at 20°C.

Each of the isotherms in the experimental data shown in Figures 6.2 and 6.3 are curve fit to equation (6-16). The experimental constants, A and B, for the following curve fit are shown in Table 6.2.

$$\ln f\ (\text{psia}) = A(T) + B(T)(H/Pd) \qquad (6\text{-}16)$$

Table 6.2 Constants for the Palladium-Hydrogen Curve fit.

Temperature	A	B	Temperature	A	B
-60°C	-46.547	61.85	40°C	-25.461	41.516
-40°C	-41.743	57.472	60°C	-20.631	39.075
-20°C	-36.206	51.99	80°C	-20.494	36.669
0°C	-32.269	48.242	100°C	-18.297	34.465
20°C	-28.502	44.487	120°C	-16.276	32.422

6.4 Comparison of Curve Fit With the Literature Data

The data for each isotherm of the palladium-hydrogen equation of state has been curve fit to the equation (6-16).

Wicke and Nernst, and Perminov, Orlov, and Frumkin have all experimented in the β-phase region. Their low temperature data is shown in comparison with the experimental curve fits in Figure 6.4, the error bars represent 1% error in the atomic ratio. Perminov, Orlov, and Frumkins' experimental data was modified to express the pressure in terms of fugacity as shown in Appendix C.

Perminov, Orlov and Frumkins' modified data at 0°C is within the 1% error in atomic ratio to a fugacity of 1000 psia, as shown in Figure 6.4. At this point Perminov, Orlov and Frumkins' 0°C isotherm slopes up from this experimental curve fit. Perminov, Orlov and Frumkins' modified low-pressure data at -32°C begins between the -40°C and the -20°C curve fits. Perminov, Orlov and Frumkins' -32°C modified data then deviates from the curve fit of this study.

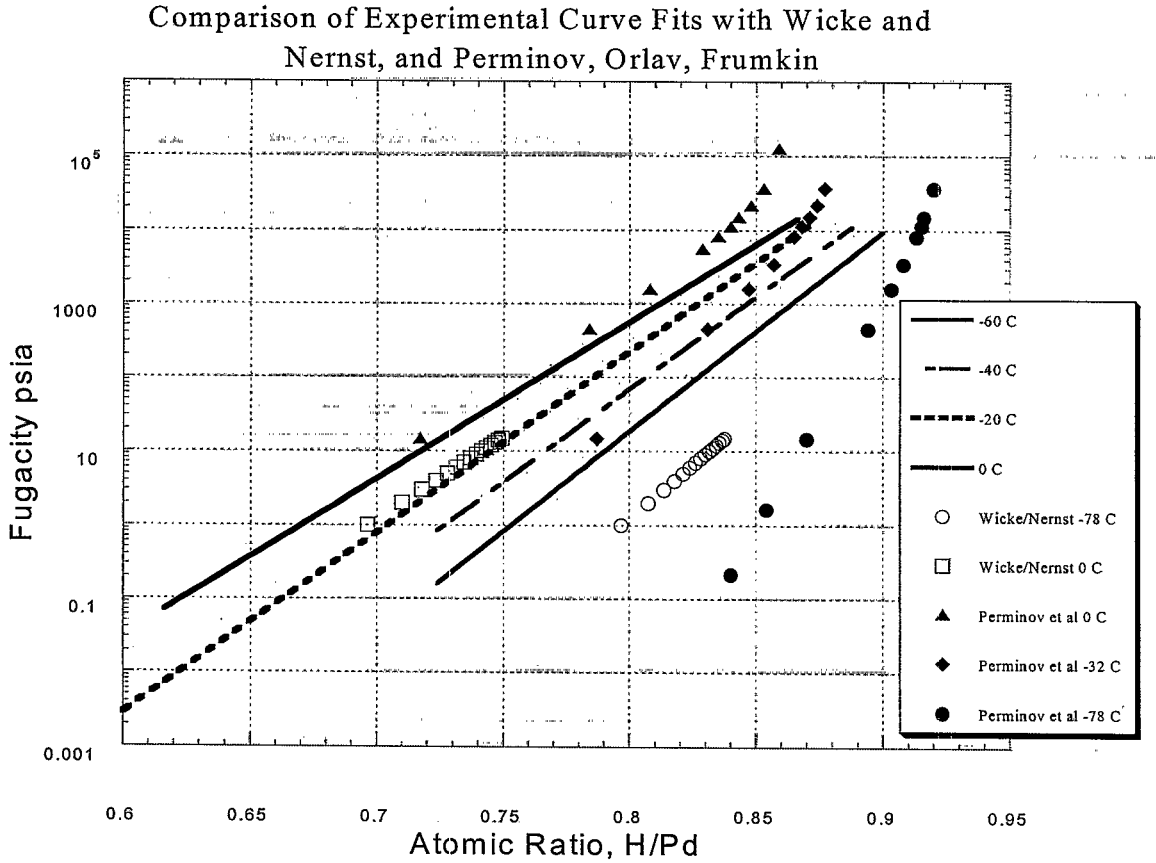

Figure 6.4 Comparison of Low Temperature Experimental Curve Fits (Perminov, et al, 1952) and (Wicke and Nernst, 1964) with literature data.

Wicke and Nernsts' -78°C and 0°C isotherms have the same general slope as the experimental isotherms in their vicinity. Wicke and Nernsts' 0°C isotherm is offset from the curve fit of this study by 2%. Wicke and Nernsts' 0°C isotherm is reads a higher atomic ratio than this study.

A comparison of Wicke and Nernst, and Perminov, Orlov and Frumkins' experimental data at -78°C is also shown in Figure 6.4. Perminov, Orlov and Frumkin noted that their data at this temperature was not very accurate.

A comparison of the higher temperature curve fits is shown in Figure 6.5. An error bar of 1% is shown on the experimental 40°C isotherm. All of Perminov, Orlov, and Frumkins' modified data at 50°C are within 1% of the experimental curve fit.

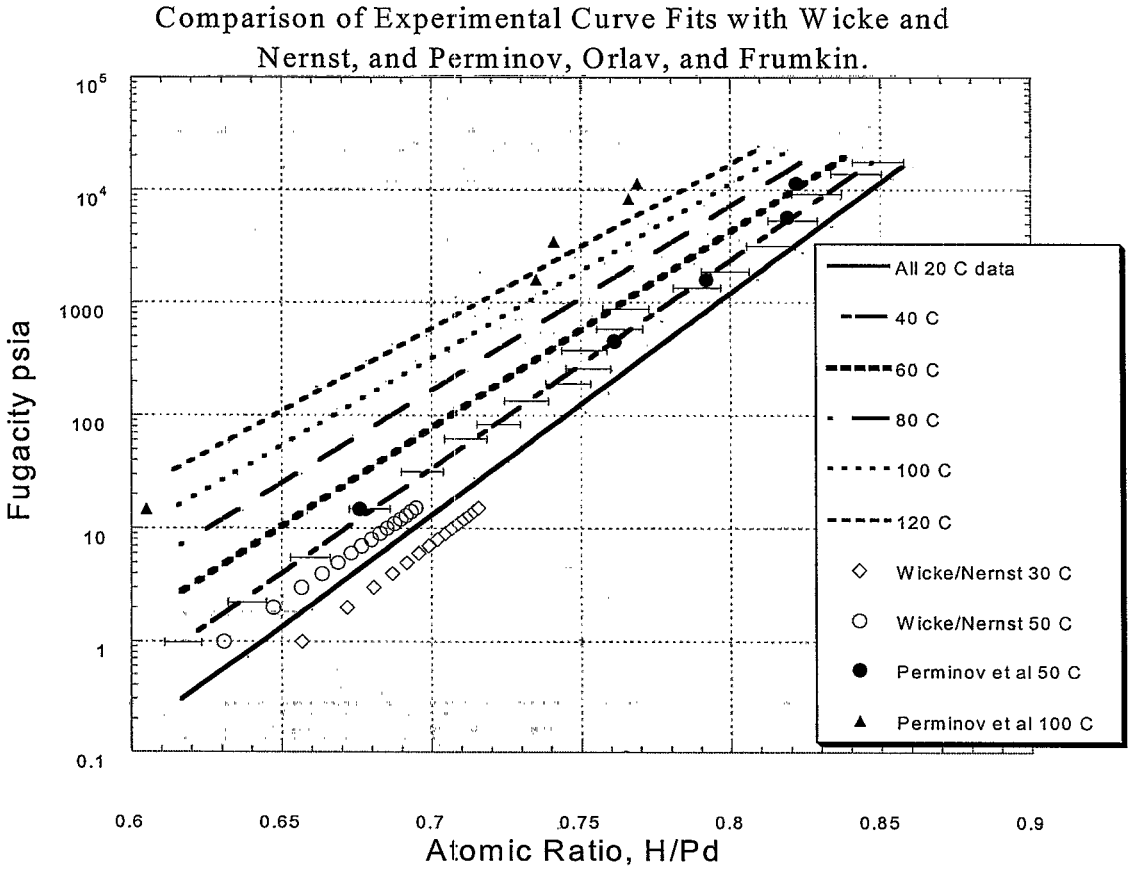

Figure 6.5 Comparison of High Temperature Experimental Curve Fits (Perminov et al, 1952) and (Wicke and Nernst, 1964) with literature data.

Perminov, Orlov, and Frumkins' 100°C modified data is within 1% of the experimental curve fit up to a fugacity of 1000 psia. At higher fugacities their 100°C isotherm has a lower atomic ratio than that shown by this study. Wicke and Nernsts' 30°C and 50°C data are offset and read a higher atomic ratio by 2% than the experimental curve fit. This 2% offset in Wicke and Nernsts' data was seen in the low temperature data as well.

6.5 Uncertainty Analysis

The general uncertainty analysis that is employed for the estimating the uncertainty in the fugacity and the atomic ratio is the Kline-McClintock method (Holman, 1989). This method uses the products of the sensitivities of the measured variable with respect to the independent variables and the uncertainties of the independent variables themselves. The form of the uncertainty for fugacity becomes equation (6-17),

$$U_f = \left[\left(\frac{\partial f}{\partial P} * U_P \right)^2 + \left(\frac{\partial f}{\partial T} * U_T \right)^2 \right]^{\frac{1}{2}} \tag{6-17}$$

Equation (6-18) is the partial derivative of fugacity with respect to pressure.

$$\frac{\partial f}{\partial P} = \frac{\partial}{\partial P} \left[P \exp \left(\begin{array}{l} \left[C_0 + C_1 T + C_2 T^2 \right] * \left(\frac{P}{T} \right) + \left[C_3 + C_4 T + C_5 T^2 \right] * \frac{1}{2} \left(\frac{P}{T} \right)^2 \\ + \left[C_6 + C_7 T + C_8 T^2 \right] * \frac{1}{3} \left(\frac{P}{T} \right)^3 + \left[C_9 + C_{10} T + C_{11} T^2 \right] * \frac{1}{4} \left(\frac{P}{T} \right)^4 \end{array} \right) \right] \tag{6-18}$$

Taking this partial derivative results in equation (6-19).

$$\begin{aligned} \frac{\partial f}{\partial P} = & \exp \left(\begin{array}{l} \left[C_0 + C_1 T + C_2 T^2 \right] * \left(\frac{P}{T} \right) + \left[C_3 + C_4 T + C_5 T^2 \right] * \frac{1}{2} \left(\frac{P}{T} \right)^2 \\ + \left[C_6 + C_7 T + C_8 T^2 \right] * \frac{1}{3} \left(\frac{P}{T} \right)^3 + \left[C_9 + C_{10} T + C_{11} T^2 \right] * \frac{1}{4} \left(\frac{P}{T} \right)^4 \end{array} \right) \\ & * \left(1 + P \left[\begin{array}{l} \frac{\left[C_0 + C_1 T + C_2 T^2 \right]}{T} + \frac{\left[C_3 + C_4 T + C_5 T^2 \right]}{T^2} P + \\ \frac{\left[C_6 + C_7 T + C_8 T^2 \right]}{T^3} P^2 + \frac{\left[C_9 + C_{10} T + C_{11} T^2 \right]}{T^4} P^3 \end{array} \right] \right) \end{aligned} \tag{6-19}$$

The equation for the partial derivative of fugacity with respect to temperature is:

$$\frac{\partial f}{\partial T} = P\left(\begin{array}{l} -\frac{C_0 P}{T^2} + C_2 P - \frac{C_3 P^2}{T^3} - \frac{C_4 P^2}{2(T^2)} - \frac{C_6 P^3}{T^4} - \left(\frac{2}{3}\right)\frac{C_7 P^3}{T^3} \\ -\frac{C_8 P^3}{3(T^2)} - \frac{C_9 P^4}{T^5} - \left(\frac{3}{4}\right)\frac{C_{10} P^4}{T^4} - \frac{C_{11} P^4}{2(T^3)} \end{array}\right) * \exp\left(\begin{array}{l} \left[C_0 + C_1 T + C_2 T^2\right] * \left(\frac{P}{T}\right) + \left[C_3 + C_4 T + C_5 T^2\right] * \frac{1}{2}\left(\frac{P}{T}\right)^2 \\ + \left[C_6 + C_7 T + C_8 T^2\right] * \frac{1}{3}\left(\frac{P}{T}\right)^3 + \left[C_9 + C_{10} T + C_{11} T^2\right] * \frac{1}{4}\left(\frac{P}{T}\right)^4 \end{array}\right) \quad (6\text{-}20)$$

With these two partial derivatives and the uncertainty in sensitivities of the pressure and temperature the uncertainty in the fugacity can be determined. This is shown in the equation (6-21).

$$U_f = \left[\begin{array}{l} \left(\exp\left(\begin{array}{l} \left[C_0 + C_1 T + C_2 T^2\right] * \left(\frac{P}{T}\right) + \left[C_3 + C_4 T + C_5 T^2\right] * \frac{1}{2}\left(\frac{P}{T}\right)^2 + \\ \left[C_6 + C_7 T + C_8 T^2\right] * \frac{1}{3}\left(\frac{P}{T}\right)^3 + \left[C_9 + C_{10} T + C_{11} T^2\right] * \frac{1}{4}\left(\frac{P}{T}\right)^4 \end{array}\right) * \left(1 + P\left[\begin{array}{l} \frac{\left[C_0 + C_1 T + C_2 T^2\right]}{T} + \frac{\left[C_3 + C_4 T + C_5 T^2\right]}{T^2} P + \\ \frac{\left[C_6 + C_7 T + C_8 T^2\right]}{T^3} P^2 + \frac{\left[C_9 + C_{10} T + C_{11} T^2\right]}{T^4} P^3 \end{array}\right]\right) * U_P\right)^2 + \\ \left(P\left(\begin{array}{l} -\frac{C_0 P}{T^2} + C_2 P - \frac{C_3 P^2}{T^3} - \frac{C_4 P^2}{2(T^2)} - \frac{C_6 P^3}{T^4} - \left(\frac{2}{3}\right)\frac{C_7 P^3}{T^3} \\ -\frac{C_8 P^3}{3(T^2)} - \frac{C_9 P^4}{T^5} - \left(\frac{3}{4}\right)\frac{C_{10} P^4}{T^4} - \frac{C_{11} P^4}{2(T^3)} \end{array}\right) * \exp\left(\begin{array}{l} \left[C_0 + C_1 T + C_2 T^2\right] * \left(\frac{P}{T}\right) + \left[C_3 + C_4 T + C_5 T^2\right] * \frac{1}{2}\left(\frac{P}{T}\right)^2 + \\ \left[C_6 + C_7 T + C_8 T^2\right] * \frac{1}{3}\left(\frac{P}{T}\right)^3 + \left[C_9 + C_{10} T + C_{11} T^2\right] * \frac{1}{4}\left(\frac{P}{T}\right)^4 \end{array}\right) * U_T\right)^2 \end{array}\right]^{\frac{1}{2}} \quad (6\text{-}21)$$

The general uncertainty analysis in the determination of the atomic ratio is calculated in terms of the uncertainty in the moles of hydrogen. Equation (6-22) is the general equation for the uncertainty in the moles of hydrogen.

$$U_{molH_2} = \left[\begin{array}{l} \left(\frac{\partial molH_2}{\partial \text{weight}} U_{\text{weight}} \right)^2 + \left(\frac{\partial molH_2}{\partial P} U_P \right)^2 + \left(\frac{\partial molH_2}{\partial V_1} U_{V1} \right)^2 + \left(\frac{\partial molH_2}{\partial V_2} U_{V2} \right)^2 \\ \left(\frac{\partial molH_2}{\partial V_3} U_{V3} \right)^2 + \left(\frac{\partial molH_2}{\partial T_{1,2}} U_{T1,2} \right)^2 + \left(\frac{\partial molH_2}{\partial T_3} U_{T3} \right)^2 \end{array} \right] \quad (6\text{-}22)$$

Equations for the partial deriatives involed in the determination of the uncertainty in the moles of hydrogen are shown in equations (6-23) thru (6-28)

$$\frac{\partial mol\,H_2}{\partial weight} = \frac{1}{MW_{H2}} \quad (6\text{-}23)$$

$$\frac{\partial mol\,H_2}{\partial P} = \left(\frac{V_2}{Z_2 R T_2} + \frac{V_3}{Z_3 R T_3} \right)_{20degC} - \left(\frac{V_1}{Z_1 R T_1} + \frac{V_2}{Z_2 R T_2} + \frac{V_3}{Z_3 R T_3} \right) \quad (6\text{-}24)$$

$$\frac{\partial mol\,H_2}{\partial V_1} = -\frac{P}{Z R T_1} \quad (6\text{-}25)$$

$$\frac{\partial mol\,H_2}{\partial V_2} = \left(\frac{P}{Z_2 R T_2} \right)_{20degC} - \frac{P}{Z_2 R T_2} \quad (6\text{-}26)$$

$$\frac{\partial mol\,H_2}{\partial V_3} = \left(\frac{P}{Z_3 R T_3} \right)_{20degC} - \frac{P}{Z_3 R T_3} \quad (6\text{-}27)$$

$$\frac{\partial mol\,H_2}{\partial T_{1,2}} = \left(-\frac{P V_1}{Z_1 R T_1^2} U_{T1,2} \right) \quad (6\text{-}28)$$

$$\frac{\partial mol\,H_2}{\partial T_3} = \left(\left(-\frac{PV_3}{Z_3 RT_3^2} \right)_{20\deg C} + \frac{PV_3}{Z_3 RT_3^2} \right) \tag{6-29}$$

The partial derivatives are inserted into the general uncertainty equation (6-22). The uncertainty in the moles of hydrogen can now be calculated using measured experimental quantities. With the collective equations (6-23) to (6-29) the uncertainty in the moles of hydrogen is stated in equation (6-30).

$$U_{mol\,H2} = \left[\begin{array}{l} \left(\frac{1}{MW_{H2}} U_{weight} \right)^2 + \left(\left(\left(\left(\frac{V_2}{Z_2 RT_2} + \frac{V_3}{Z_3 RT_3} \right)_{20\deg C} - \left(\frac{V_1}{Z_1 RT_1} + \frac{V_2}{Z_2 RT_2} + \frac{V_3}{Z_3 RT_3} \right) \right) U_P \right)^2 \right. \\ \left(\frac{P}{Z_1 RT_1} U_{V1} \right)^2 + \left(\left(\left(\frac{P}{Z_2 RT_2} \right)_{20\deg C} - \frac{P}{Z_2 RT_2} \right) U_{V2} \right)^2 + \\ \left(\left(\left(\frac{P}{Z_3 RT_3} \right)_{20\deg C} - \frac{P}{Z_3 RT_3} \right) U_{V3} \right)^2 \\ \left. \left(-\frac{PV_1}{Z_1 RT_1^2} U_{T1,2} \right)^2 + \left(\left(\left(-\frac{PV_3}{Z_3 RT_3^2} \right)_{20\deg C} + \frac{PV_3}{Z_3 RT_3^2} \right) U_{T3} \right)^2 \right] \end{array}^{\frac{1}{2}} \tag{6-30}$$

The calculation for the uncertainty in the atomic ratio is derived from the definition of the atomic ratio, which is shown in equation (6-31).

$$\frac{\mathrm{H}}{\mathrm{Pd}} = \frac{2 \cdot \mathrm{mols\,H_2}}{\mathrm{mols\,Pd}} \tag{6-31}$$

The uncertainty in the atomic ratio of hydrogen to palladium can be calculated with respect to the independent variables, moles of hydrogen and moles of palladium. Utilizing the uncertainty in the moles of hydrogen found in equation (6-30) and knowing the uncertainty in the moles of palladium, the uncertainty of the atomic ratio can be

calculated. The calculation for the uncertainty in the atomic ratio is shown in equation (6-32).

$$U_{\frac{H}{Pd}} = \left(\left(\frac{2}{\text{mol Pd}} U_{\text{mol H2}} \right) + \left(-\frac{2 \cdot \text{mol H}_2}{(\text{mol Pd})} U_{\text{molPd}} \right)^2 \right)^{\frac{1}{2}} \qquad (6\text{-}32)$$

The uncertainty in the atomic ratio is calculated for each of the individual atomic ratio data points, shown in Appendix A. A determination of the largest effect on the uncertainty in the atomic ratio was done by selectively zeroing the sensitivities of the each of the independent variables in the general uncertainty equation. The largest effect on the uncertainty of the atomic ratio is the uncertainty in the moles of hydrogen. This uncertainty is greatly affected by the sensitivity in the weight measurement. The uncertainty is larger for the palladium beds that have a low pressure and thus a lower mass of hydrogen. Conversely the uncertainty is smaller with high pressures and higher loads of hydrogen because of the higher weight of hydrogen. For instance the uncertainty at room temperature and 660 psia is 0.3% of the atomic ratio and at room temperature and 10,000 psia the uncertainty is 1.2% of the atomic ratio.

The sensitivity of the pressure transducers has the second largest effect on the uncertainty of our measurements. In this experiment many different ranges of the Heise pressure transducers are used. The sensitivity is quoted by the manufacturer to be better than ±0.005 of the span. The higher the range of the transducer the higher the uncertainty in the measurement. The pressure transducers were matched with each loading of the palladium bed to try to reduce unnecessary uncertainty in the measurements.

CHAPTER 7

Conclusions and Recommendations

7.1 Conclusions and Recommendations

This study demonstrates that taking a direct gravimetric weight measurement of a 50 gram hydrided palladium bed is a viable and accurate method of determining the equation of state for the palladium-hydrogen system. Curve fit data has been determined for the atomic ratio from -60°C to 120°C with a range in pressures of 0 to 20,000 psia.

The uncertainty of the measurement of the atomic ratio, H/Pd, is greatly influenced by the sensitivity, accuracy, and method of taking the weight measurement of the hydrided palladium bed. The largest contributor to the uncertainty is the sensitivity of the weight measurement. An increase in the accuracy and precision of the mass balance would further improve the results of a future experiment. The accuracy and precision of the pressure measurement is a secondary contributor to the uncertainty of the measurement. An improvement in the accuracy of the pressure transducers would improve the accuracy of the measurements. This improvement would not have as large an effect as an improvement in the weight measurement.

The accuracy in the determination of the equation of state of the palladium was improved upon in this investigation. The data was curve fit to a known equation used in the literature and 95% fugacity versus atomic ratio data points lie within 1% of the curve fit. The error in the data lie within the calculated uncertainty of the atomic ratio.

The experimental curve fit was compared to the literature. The experimental curve fit is within 1% of the atomic ratio of Perminov, Orlov, and Frumkins' data in the pressure region below 1,000 psia. Above this pressure Perminov, Orlov and Frumkins'

data slopes up and away from the experimental correlation. Wicke and Nernsts' data is also compared to this correlation and was found to have an atomic ratio that is 2% higher than this studies data. Factors that could cause this offset in the atomic ratio are impurities in the system, impurities in the palladium or hydrogen, or a systematic error in the volumetric method used by Wicke and Nernst.

An extension of this work could include the determination of the palladium-deuterium, palladium-tritium, metal-hydride equations of state. Other future work in this area could be to extend the pressure and temperature limits to determine if the experimental correlation could accurately predict the palladium-hydrogen equation of state in these ranges.

APPENDICIES

Compressibility constants			
-60 deg C to 80 deg C(Meyer 1996)			
a1	0.022456	8.306E-04	-1.019E-06
a2	0.056181	-1.911E-04	1.566E-07
a3	-0.0036149	-8.166E-06	3.014E-08
a4	-0.00015121	2.755E-06	-4.672E-09

PCT Constants, Meyer 1995	
for equation lnf=a-(b-cn)/T-dT	
a=	-0.37970
b=	10653
c=	12035
d=	0.02737

Units were opened with one full turn

	V1 cc	Pd	V2 cc	V2 cc	X-ducer	weight of unit (gm)	weight after	weight H/Pd
	unhydrided	gm	in	out	no.	before load	load	in grams
1	6.302	50.117	0.173	1.564	H 25743	1648.465	1648.855	0.3900
2	6.394	50.539	0.173	0.794	H 27163	1645.120	1645.758	0.6380
3	6.427	50.535	0.173	0.612	P 40454	1635.168	1635.636	0.4677
4	6.381	50.520	0.173	0.612	P 40456	1653.087	1653.596	0.5087
7	6.367	50.502	0.173	0.612	P 40457	1658.495	1659.056	0.5607

						Moles in the gas phase						hydrided	% Uncer
Date	time	unit	Pressure	Temp	Temp	moles bed	moles in	moles out	Fugacity	moles	H/Pd	free bed	tainty
			psia	bed °C	out °C	V1	V2	V3	psia	H2 solid		V1 cc	H/PD
2/28/98	7:45	1	666.9	21.4	22.3	0.0105	0.0003	0.0028	685.62	0.1829	0.777	5.74	0.29
		2	10303	21.4	22.3	0.1128	0.0034	0.0157	16160.86	0.2035	0.857	5.69	1.23
		3	2502	21.4	22.3	0.0364	0.0011	0.0039	2779.85	0.1956	0.824	5.72	0.45
		4	4022	21.4	22.3	0.0550	0.0017	0.0059	4771.87	0.1972	0.831	5.71	0.63
		7	6340	21.4	22.3	0.0794	0.0024	0.0085	8328.96	0.1985	0.837	5.70	0.88
2/28/98	8:16	1	664	21.2	22.2	0.0104	0.0003	0.0028	682.56	0.1830	0.777	5.74	0.29
		2	10292	21.2	22.4	0.1127	0.0034	0.0157	16139.69	0.2036	0.857	5.69	1.23
		3	2500	21.2	22.6	0.0364	0.0011	0.0039	2777.53	0.1956	0.824	5.72	0.45
		4	4020	21.2	22.8	0.0550	0.0017	0.0059	4769.49	0.1972	0.831	5.71	0.63
		7	6334	21.2	22.8	0.0794	0.0024	0.0085	8320.05	0.1986	0.837	5.70	0.88

Date	time	unit	Pressure	Temp	Temp	moles bed	moles in	moles out	Fugacity	moles	H/Pd	free bed	% Uncertainty
			psia	bed °C	out °C	V1	V2	V3	psia	H2 solid		V1 cc	H/PD
2/28/98	9:37	1	488.5	0.4	20.5	0.0083	0.0003	0.0021	498.96	0.1859	0.789	5.73	0.28
		2	9335	0.4	21.5	0.1110	0.0034	0.0147	14372.75	0.2064	0.869	5.68	1.19
		3	2168	0.6	21.6	0.0342	0.0010	0.0034	2385.37	0.1983	0.835	5.71	0.43
		4	3571	0.9	21.8	0.0529	0.0016	0.0053	4187.17	0.1999	0.842	5.70	0.60
		7	5707	1	21.6	0.0775	0.0024	0.0079	7388.96	0.2012	0.848	5.69	0.85
2/28/98	10:35	1	336.4	-19.6	20.8	0.0062	0.0002	0.0015	341.55	0.1887	0.801	5.73	0.27
		2	8417	-19.8	21.1	0.1092	0.0033	0.0137	12698.79	0.2092	0.881	5.68	1.15
		3	1854	-19.6	21.3	0.0318	0.0010	0.0030	2019.29	0.2012	0.847	5.70	0.40
		4	3144	-19.4	21.8	0.0508	0.0015	0.0048	3641.45	0.2027	0.854	5.69	0.57
		7	5104	-19.2	21.7	0.0756	0.0023	0.0072	6507.24	0.2038	0.859	5.69	0.81
2/28/98	11:45	1	209.7	-39.6	20.7	0.0042	0.0001	0.0009	211.78	0.1913	0.812	5.72	0.26
		2	7540	-39.6	20.8	0.1074	0.0033	0.0126	11130.61	0.2122	0.893	5.67	1.11
		3	1568	-39.4	21.1	0.0294	0.0009	0.0025	1690.83	0.2041	0.86	5.70	0.37
		4	2744	-39.1	21.5	0.0485	0.0015	0.0042	3138.08	0.2055	0.866	5.69	0.54
		7	4532	-38.9	21.6	0.0736	0.0022	0.0065	5684.79	0.2065	0.87	5.68	0.78
2/28/98	12:22	1	106.8	-60.3	20.3	0.0024	0.0001	0.0005	107.36	0.1937	0.822	5.72	0.26
		2	6656	-60.1	20.8	0.1054	0.0032	0.0115	9587.46	0.2153	0.907	5.67	1.07
		3	1296	-59.7	21.2	0.0269	0.0008	0.0021	1383.27	0.2072	0.872	5.69	0.35
		4	2361	-59.2	21.6	0.0462	0.0014	0.0037	2664.82	0.2085	0.878	5.68	0.51
		7	3965	-59.1	21.5	0.0714	0.0022	0.0058	4885.88	0.2095	0.883	5.68	0.74

Date	time	unit	Pressure psia	Temp bed °C	Temp out °C	moles bed V1	moles in V2	moles out V3	Fugacity psia	moles H2 solid	H/Pd	free bed V1 cc	% Uncertainty H/PD
2/28/98	13:35	1	661.4	21.1	21.5	0.0104	0.0003	0.0028	679.82	0.1830	0.777	5.74	0.29
		2	10227	20.1	21.6	0.1126	0.0034	0.0156	16013.01	0.2038	0.858	5.69	1.23
		3	2486	20.9	21.8	0.0362	0.0011	0.0039	2760.52	0.1958	0.824	5.72	0.45
		4	4001	20.9	22.1	0.0549	0.0017	0.0059	4743.61	0.1974	0.832	5.71	0.63
		7	6295	21	22.2	0.0791	0.0024	0.0085	8255.70	0.1989	0.838	5.70	0.87
2/28/98	14:27	1	846.9	41	22.5	0.0124	0.0004	0.0036	875.94	0.1802	0.765	5.75	0.31
		2	11126	40.9	22.3	0.1138	0.0035	0.0165	17650.39	0.2017	0.849	5.70	1.26
		3	2826	41	22.7	0.0383	0.0012	0.0043	3166.68	0.1932	0.813	5.73	0.48
		4	4455	40.9	22.7	0.0568	0.0017	0.0064	5338.50	0.1949	0.821	5.72	0.66
		7	6905	40.9	22.8	0.0807	0.0024	0.0091	9165.17	0.1967	0.829	5.71	0.90
2/28/98	15:08	1	1051.9	61.3	22.6	0.0144	0.0004	0.0044	1094.90	0.1773	0.753	5.76	0.33
		2	12059	61.1	22.8	0.1153	0.0035	0.0174	19375.11	0.1992	0.839	5.71	1.29
		3	3181	61	23.1	0.0403	0.0012	0.0048	3595.19	0.1907	0.803	5.74	0.51
		4	4922	60.9	23.1	0.0586	0.0018	0.0069	5955.76	0.1924	0.811	5.73	0.69
		7	7531	60.8	23.2	0.0823	0.0025	0.0097	10104.64	0.1944	0.819	5.72	0.93
2/28/98	16:22	1	1270	81.5	23.2	0.0164	0.0005	0.0053	1330.10	0.1744	0.741	5.77	0.35
		2	12992	81.2	23.3	0.1168	0.0035	0.0182	21089.90	0.1969	0.829	5.72	1.33
		3	3553	81.2	23.7	0.0422	0.0013	0.0053	4047.94	0.1882	0.792	5.75	0.53
		4	5410	81.1	24	0.0605	0.0018	0.0075	6605.17	0.1900	0.8	5.74	0.72
		7	8170	80.9	23.8	0.0839	0.0025	0.0102	11064.34	0.1922	0.81	5.73	0.96

Date	time	unit	Pressure	Temp	Temp	moles bed	moles in	moles out	Fugacity	moles	H/Pd	free bed	% Uncer tainty
			psia	bed °C	out °C	V1	V2	V3	psia	H2 solid		V1 cc	H/PD
2/28/98	17:30	1	1504.2	101.7	23.5	0.0183	0.0005	0.0062	1584.91	0.1716	0.728	5.78	0.37
		2	13926	101.3	23.8	0.1183	0.0036	0.0190	22784.89	0.1946	0.819	5.73	1.36
		3	3934	101.2	24.1	0.0441	0.0013	0.0057	4514.55	0.1858	0.782	5.76	0.56
		4	5891	101.1	24.2	0.0621	0.0019	0.0080	7245.29	0.1878	0.791	5.75	0.75
		7	8808	100.8	25.4	0.0854	0.0026	0.0108	12019.06	0.1902	0.801	5.74	0.99
2/28/98	18:35	1	1754	122.1	24.4	0.0202	0.0006	0.0071	1858.82	0.1687	0.716	5.79	0.40
		2	14877	121.6	23.9	0.1198	0.0036	0.0198	24483.13	0.1922	0.809	5.74	1.40
		3	4330	121.5	24.1	0.0459	0.0014	0.0062	5001.21	0.1835	0.773	5.77	0.59
		4	6375	121.2	24.8	0.0636	0.0019	0.0085	7887.36	0.1857	0.782	5.77	0.77
		7	9450	120.8	24.8	0.0868	0.0026	0.0113	12970.86	0.1881	0.793	5.75	1.02
2/28/98	20:15	1	658	21	22.5	0.0104	0.0003	0.0028	676.23	0.1831	0.777	5.74	0.29
		2	10145	20.8	22.6	0.1118	0.0034	0.0155	15812.05	0.2048	0.862	5.69	1.21
		3	2492	21.1	23	0.0363	0.0011	0.0039	2767.76	0.1957	0.824	5.72	0.45
		4	3962	21.2	23.3	0.0544	0.0016	0.0058	4688.82	0.1980	0.834	5.71	0.62
		7	6220	21.2	23.4	0.0783	0.0024	0.0084	8129.11	0.1998	0.842	5.70	0.86
2/28/98	20:20	1	658.8	21	22.5	0.0104	0.0003	0.0028	677.08	0.1831	0.777	5.74	0.29
		2	10180	20.8	22.6	0.1120	0.0034	0.0156	15891.66	0.2044	0.861	5.69	1.22
		3	2499	21.1	23	0.0364	0.0011	0.0039	2776.37	0.1956	0.824	5.72	0.45
		4	3973	21.2	23.3	0.0545	0.0017	0.0058	4704.09	0.1978	0.833	5.71	0.62
		7	6242	21.2	23.4	0.0785	0.0024	0.0084	8165.83	0.1996	0.841	5.70	0.87

Compressibility constants			
-60 to 80 °C			
a1	0.022456	8.31E-04	-1.02E-06
a2	0.056181	-1.91E-04	1.57E-07
a3	-0.003615	-8.17E-06	3.01E-08
a4	-0.000151	2.75E-06	-4.67E-09

PCT Constants, Meyer 1995		
for the equation lnf=a-(b-cn)/T-dT		
a=	-0.3797	
b=	10653	
c=	12035	
d=	0.027373	

Units were opened with a half turn

	V1 cc	Pd	V2 cc	V2 cc	X-ducer	weight of unit (gm)	weight after	weight measured
	unhydri	gm	in	out	no.	before load	load	in grams
1	6.302	50.1167	0.173	1.56	H 25743	1648.465	1648.834	0.3690
2	6.394	50.5392	0.173	0.79	H 27163	1645.12	1645.7297	0.6097
3	6.427	50.5348	0.173	0.61	P 40454	1635.168	1635.6013	0.4333
4	6.381	50.5204	0.173	0.61	P 40456	1653.087	1653.5027	0.4157
7	6.367	50.5015	0.173	0.61	P 40457	1658.495	1658.8777	0.3827

						Moles in the gas phase							% Uncer
Date	time	unit	Pressure	Temp	Temp	moles bed	moles in	moles out	Fugacity	moles	H/Pd	free bed	tainty
			psia	bed °C	out °C	V1	V2	V3	psia	H2 solid		V1 cc	H/PD
3/1/98	7:28	1	250.8	21.4	21.4	4.021E-03	1.209E-04	1.093E-03	253.42	0.17901	0.7601	5.754533	0.280635
		2	8673	21.3	21.4	1.002E-01	3.042E-03	1.395E-02	12644.66	0.20228	0.8517	5.695073	0.978963
		3	1509	21.4	21.4	2.287E-02	6.909E-04	2.445E-03	1607.21	0.19206	0.8088	5.726525	0.856459
		4	1061	21.5	21.4	1.639E-02	4.946E-04	1.751E-03	1108.92	0.18981	0.7995	5.73222	0.64719
		7	414	21.5	21.4	6.583E-03	1.982E-04	7.015E-04	421.16	0.18325	0.7722	5.746906	0.366162
3/1/98	8:30	1	144.5	0.6	21	2.501E-03	7.526E-05	6.331E-04	145.40	0.18104	0.7687	5.749169	0.275172
		2	7848	0.6	21.3	9.836E-02	2.992E-03	1.298E-02	11249.28	0.20509	0.8635	5.687702	0.942369
		3	1258	0.9	21.4	2.062E-02	6.238E-04	2.059E-03	1329.03	0.19476	0.8201	5.718115	0.7547
		4	849	1.1	21.6	1.417E-02	4.282E-04	1.412E-03	880.91	0.19244	0.8106	5.724041	0.554363
		7	276	1.2	21.8	4.732E-03	1.426E-04	4.697E-04	279.31	0.18539	0.7812	5.740336	0.31438

Date	time	unit	Pressure	Temp	Temp	moles bed	moles in	moles out	Fugacity	moles	H/Pd	free bed	% Uncertainty
			psia	bed °C	out °C	V1	V2	V3	psia	H2 solid		V1 cc	H/PD
3/1/98	10:47	1	70.8	-19.6	21	1.326E-03	3.993E-05	3.112E-04	71.03	0.18257	0.7752	5.746293	0.272147
		2	7053	-19.7	21.2	9.650E-02	2.938E-03	1.200E-02	9922.73	0.20799	0.8758	5.681547	0.90561
		3	1021	-19.6	21.3	1.821E-02	5.517E-04	1.688E-03	1069.64	0.19762	0.8322	5.710837	0.651849
		4	651	-19.3	21.7	1.181E-02	3.573E-04	1.091E-03	670.50	0.19519	0.8222	5.717106	0.463485
		7	164.5	-19.1	22.1	3.057E-03	9.221E-05	2.810E-04	165.72	0.1873	0.7892	5.735521	0.281068
3/1/98	11:46	1	27.3	-39.6	21	5.563E-04	1.675E-05	1.202E-04	27.33	0.18356	0.7794	5.746059	0.270704
		2	6290	-39.7	21.2	9.457E-02	2.882E-03	1.100E-02	8673.38	0.21098	0.8883	5.676498	0.868865
		3	809	-39.5	21.2	1.577E-02	4.781E-04	1.350E-03	840.71	0.20047	0.8442	5.704826	0.554061
		4	479	-39.2	21.8	9.481E-03	2.872E-04	8.084E-04	489.96	0.19787	0.8335	5.711488	0.383452
		7	82.5	-39	22	1.669E-03	5.035E-05	1.414E-04	82.82	0.18887	0.7959	5.733061	0.265569
3/1/98	12:36	1	8.4	-59.9	20.7	1.877E-04	5.649E-06	3.705E-05	8.40	0.18402	0.7814	5.747271	0.270159
		2	5545	-59.8	21.1	9.250E-02	2.821E-03	9.963E-03	7481.12	0.21414	0.9017	5.672425	0.831086
		3	614	-59.6	21.2	1.318E-02	4.001E-04	1.033E-03	632.97	0.20345	0.8567	5.699833	0.458957
		4	326	-59.2	21.5	7.097E-03	2.151E-04	5.542E-04	331.27	0.20058	0.8449	5.707158	0.314788
		7	32	-59	21.7	7.093E-04	2.140E-05	5.503E-05	32.05	0.18995	0.8004	5.733064	0.261892
3/1/98	13:40	1	248.8	21.2	21.8	3.992E-03	1.200E-04	1.083E-03	251.37	0.17905	0.7603	5.754523	0.280511
		2	8647	21.1	21.7	9.999E-02	3.037E-03	1.391E-02	12594.65	0.20249	0.8526	5.69504	0.976125
		3	1499	20.9	21.9	2.276E-02	6.877E-04	2.426E-03	1596.00	0.19219	0.8093	5.726347	0.851534
		4	1052	20.8	22.1	1.629E-02	4.917E-04	1.733E-03	1099.17	0.18993	0.8	5.731942	0.642931
		7	413	21	22.2	6.578E-03	1.980E-04	6.980E-04	420.13	0.18326	0.7722	5.746624	0.36593

Date	time	unit	Pressure	Temp	Temp	moles bed	moles in	moles out	Fugacity	moles	H/Pd	free bed	% Uncer tainty
			psia	bed °C	out °C	V1	V2	V3	psia	H2 solid		V1 cc	H/PD
3/1/98	14:56	1	372	40.9	22.5	5.577E-03	1.675E-04	1.607E-03	377.53	0.1769	0.7511	5.761554	0.28812
		2	9439	40.8	22.6	1.016E-01	3.082E-03	1.476E-02	13942.94	0.19998	0.842	5.703092	1.009596
		3	1763	40.9	22.7	2.492E-02	7.517E-04	2.816E-03	1891.97	0.18958	0.7983	5.735529	0.95348
		4	1283	40.8	22.8	1.850E-02	5.575E-04	2.088E-03	1350.46	0.1873	0.7889	5.740918	0.740761
		7	569	40.8	23	8.457E-03	2.542E-04	9.530E-04	582.02	0.18107	0.763	5.754507	0.430381
3/1/98	15:55	1	519.1	60.9	23	7.291E-03	2.186E-04	2.225E-03	529.45	0.17451	0.741	5.770239	0.298699
		2	10254	60.8	23.1	1.032E-01	3.126E-03	1.561E-02	15335.27	0.19749	0.8315	5.712308	1.043646
		3	2037	60.7	23.3	2.695E-02	8.114E-04	3.211E-03	2202.37	0.1871	0.7878	5.745656	1.053637
		4	1522	60.6	23.5	2.054E-02	6.180E-04	2.448E-03	1613.21	0.18484	0.7786	5.750932	0.836435
		7	740.5	60.5	23.6	1.032E-02	3.096E-04	1.229E-03	761.69	0.17888	0.7537	5.763722	0.503119
3/1/98	17:12	1	688.5	81.4	23.3	9.081E-03	2.718E-04	2.927E-03	705.94	0.17197	0.7302	5.780644	0.312436
		2	11106	81.2	23.5	1.049E-01	3.171E-03	1.645E-02	16793.43	0.19491	0.8207	5.722773	1.079827
		3	2341	81.2	23.7	2.904E-02	8.725E-04	3.642E-03	2550.23	0.18452	0.777	5.757155	1.161056
		4	1789	81.1	24	2.264E-02	6.798E-04	2.842E-03	1909.74	0.18228	0.7678	5.762343	0.939499
		7	937	80.9	24.1	1.225E-02	3.671E-04	1.540E-03	969.52	0.17657	0.744	5.774505	0.586369
3/1/98	18:23	1	872.7	101.5	23.3	1.086E-02	3.244E-04	3.683E-03	899.53	0.16938	0.7192	5.792124	0.328871
		2	11934	101.1	23.7	1.065E-01	3.212E-03	1.724E-02	18196.83	0.19252	0.8106	5.734061	1.114305
		3	2643	101.1	24	3.092E-02	9.272E-04	4.059E-03	2898.39	0.18216	0.7671	5.769379	1.262293
		4	2055	100.8	24.2	2.454E-02	7.353E-04	3.228E-03	2207.65	0.17994	0.7579	5.774429	1.037242
		7	1139	100.5	24.4	1.407E-02	4.205E-04	1.855E-03	1185.06	0.17439	0.7348	5.78609	0.670086

Date	time	unit	Pressure	Temp	Temp	moles bed	moles in	moles out	Fugacity	moles	H/Pd	free bed	% Uncer tainty
			psia	bed °C	out °C	V1	V2	V3	psia	H2 solid		V1 cc	H/PD
3/1/98	19:49	1	1075.4	122.1	23.5	1.265E-02	3.770E-04	4.498E-03	1114.31	0.16673	0.7079	5.805223	0.348227
		2	12792	121.5	23.7	1.081E-01	3.253E-03	1.803E-02	19633.60	0.19007	0.8003	5.746755	1.150502
		3	2967	121.5	24.1	3.280E-02	9.814E-04	4.497E-03	3274.26	0.17978	0.7571	5.783017	1.367369
		4	2343	121.2	24.3	2.645E-02	7.906E-04	3.638E-03	2532.53	0.17757	0.7479	5.787955	1.139406
		7	1362	120.7	24.4	1.591E-02	4.746E-04	2.199E-03	1424.96	0.17215	0.7254	5.799117	0.760423
3/1/98	22:06	1	248.7	21	22	3.993E-03	1.201E-04	1.081E-03	251.27	0.17905	0.7603	5.754396	0.280511
		2	8603	20.9	22.2	9.968E-02	3.028E-03	1.384E-02	12508.42	0.20288	0.8542	5.695052	0.970841
		3	1507	21.05	22.4	2.287E-02	6.908E-04	2.434E-03	1605.03	0.19208	0.8088	5.726347	0.855919
		4	1064	21.15	22.7	1.645E-02	4.965E-04	1.748E-03	1112.23	0.18975	0.7993	5.731969	0.649443
		7	413.8	21.15	22.8	6.587E-03	1.983E-04	6.979E-04	420.96	0.18325	0.7722	5.74669	0.366188
3/1/98	22:22	1	248.6	20.9	22	3.993E-03	1.200E-04	1.081E-03	251.17	0.17906	0.7603	5.754336	0.280509
		2	8618	20.8	22.2	9.983E-02	3.032E-03	1.386E-02	12539.95	0.20271	0.8535	5.694967	0.97316
		3	1508	20.9	22.4	2.289E-02	6.916E-04	2.436E-03	1606.20	0.19205	0.8087	5.72625	0.85697
		4	1063	21	22.7	1.644E-02	4.963E-04	1.746E-03	1111.15	0.18976	0.7993	5.731895	0.649129
		7	413	21	22.8	6.578E-03	1.980E-04	6.966E-04	420.13	0.18326	0.7722	5.746624	0.365893

Compressibility constants -60 to 80 °C			
Const			
a1	0.022456	0.000831	-1.02E-06
a2	0.056181	-0.000191	1.57E-07
a3	-0.00361	-0.000008	3.01E-08
a4	-0.00015	0.000003	-4.67E-09

PCT Constants, Meyer 1995	
for the equation lnf=a-(b-cn)/T-dT	
a=	-0.3797
b=	10653
c=	12035
d=	0.027373

Units were opened with a half turn

	V1 cc	Pd	V2 cc	V2 cc	X-ducer	weight of unit (gm)	weight after	weight measured
	unhydr	gm	in	out	no.	before load	load	in grams
1	6.302	50.1167	0.173	1.56355	H 25855	1648.465	1648.805	0.3400
2	6.394	50.5392	0.173	0.78414	P 60798	1645.12	1645.4687	0.3487
3	6.427	50.5348	0.173	0.78414	P 31381	1635.168	1635.5227	0.3547
4	6.381	50.5204	0.173	0.78414	P 31379	1653.087	1653.4501	0.3631
7	6.367	50.5015	0.173	0.78414	P 31380	1658.495	1658.8633	0.3683

						Moles in the gas phase							%Uncer
Date	time	unit	Pressure	Temp	Temp	moles bed	moles in	moles out	Fugacity	moles	H/Pd	free bed	tainty
			psia	bed °C	out °C	V1	V2	V3	psia	H2 solid		V1 cc	H/PD
3/2/98	10:45	1	27.81	20.87	22.3	4.534E-04	1.355E-05	0.000122	27.84	0.16820	0.714182	5.79	0.885000
		2	38.5	20.5	22.5	6.277E-04	1.878E-05	0.000085	38.56	0.17234	0.725665	5.78	0.863723
		3	68.5	20.67	23.3	1.113E-03	3.335E-05	0.000150	68.69	0.17483	0.736212	5.77	0.851444
		4	105.2	20.7	23.6	1.705E-03	5.114E-05	0.000230	105.66	0.17841	0.751486	5.77	0.834417
		7	154.9	20.72	24	2.502E-03	7.514E-05	0.000337	155.90	0.18019	0.759269	5.76	0.826250
3/2/98	11:58	1	10.3	0.41	22.5	1.806E-04	5.399E-06	0.000045	10.30	0.16856	0.715701	5.79	0.883119
		2	14.95	0.4	22.7	2.618E-04	7.835E-06	0.000033	14.96	0.17277	0.727469	5.78	0.861573
		3	29.9	0.65	23	5.220E-04	1.565E-05	0.000066	29.94	0.17553	0.739131	5.77	0.848060
		4	50.4	0.9	23.2	8.772E-04	2.633E-05	0.000110	50.51	0.17938	0.755578	5.76	0.829851
		7	81.3	1	23.4	1.411E-03	4.239E-05	0.000178	81.59	0.18147	0.764676	5.76	0.820313

Date	time	unit	Pressure psia	Temp bed °C	Temp out °C	moles bed V1	moles in V2	moles out V3	Fugacity psia	moles H2 solid	H/Pd	free bed V1 cc	tainty H/PD
3/2/98	13:02	1	3.18	-19.77	22.5	6.021E-05	1.800E-06	0.000014	3.18	0.16871	0.71636	5.79	0.882307
		2	4.34	-19.9	22.8	8.216E-05	2.458E-06	0.000010	4.34	0.17298	0.728346	5.78	0.860535
		3	10.12	-19.6	23.1	1.909E-04	5.723E-06	0.000022	10.12	0.17591	0.740749	5.77	0.846205
		4	18.8	-19.3	23.3	3.537E-04	1.062E-05	0.000041	18.82	0.17999	0.758141	5.76	0.827038
		7	33.9	-19.1	23.5	6.360E-04	1.911E-05	0.000074	33.95	0.18237	0.768476	5.76	0.816234
3/2/98	16:01	1	0.77	-39.8	22.1	1.584E-05	4.734E-07	0.000003	0.77	0.16877	0.716599	5.79	0.882013
		2	0.6	-40	22.2	1.236E-05	3.692E-07	0.000001	0.60	0.17306	0.728684	5.79	0.860137
		3	2.47	-39.8	22.5	5.068E-05	1.518E-06	0.000005	2.47	0.17607	0.741428	5.77	0.845431
		4	5.14	-39.55	22.7	1.052E-04	3.156E-06	0.000011	5.14	0.18027	0.759345	5.77	0.825728
		7	10.63	-39.3	22.8	2.169E-04	6.518E-06	0.000023	10.64	0.18286	0.770509	5.76	0.814083
3/2/98	16:35	1	0.16	-59.36	21.3	3.594E-06	1.074E-07	0.000001	0.16	0.16878	0.716664	5.79	0.881933
		2											
		3	0.3	-59.4	22.2	6.732E-06	2.013E-07	0.000001	0.30	0.17612	0.741639	5.78	0.845192
		4	0.9	-58.9	22.5	2.011E-05	6.026E-07	0.000002	0.90	0.18037	0.759753	5.77	0.825286
		7	2.25	-58.9	22.7	5.018E-05	1.506E-06	0.000005	2.25	0.18305	0.77131	5.76	0.813241
3/2/98	18:37	1	27.82	20.92	22.2	4.535E-04	1.356E-05	0.000122	27.85	0.16820	0.714182	5.79	0.885000
		2	39.05	20.8	22.4	6.360E-04	1.903E-05	0.000086	39.11	0.17233	0.725624	5.78	0.863772
		3	69.2	20.88	22.8	1.124E-03	3.367E-05	0.000152	69.40	0.17482	0.736159	5.77	0.851506
		4	105.87	20.9	22.8	1.714E-03	5.143E-05	0.000232	106.33	0.17840	0.751435	5.77	0.834474
		7	155.65	20.9	22.7	2.513E-03	7.545E-05	0.000340	156.66	0.18018	0.75921	5.76	0.826315

Date	time	unit	Pressure	Temp	Temp	moles bed	moles in	moles out	Fugacity	moles	H/Pd	free bed	tainty
			psia	bed °C	out °C	V1	V2	V3	psia	H2 solid		V1 cc	H/PD
3/3/98	7:35	1	27.76	20.87	21.5	4.526E-04	1.353E-05	0.000122	27.79	0.16820	0.714186	5.79	0.884996
		2	39.04	20.8	21.6	6.358E-04	1.902E-05	0.000086	39.10	0.17233	0.725623	5.78	0.863772
		3	69.5	20.93	21.6	1.128E-03	3.381E-05	0.000153	69.70	0.17481	0.736134	5.77	0.851536
		4	106.33	20.92	21.8	1.722E-03	5.165E-05	0.000233	106.80	0.17839	0.751396	5.77	0.834518
		7	158.8	20.95	22	2.563E-03	7.696E-05	0.000348	159.85	0.18012	0.758961	5.76	0.826592
3/3/98	8:42	1	61.59	40.95	22.3	9.393E-04	2.806E-05	0.000270	61.74	0.16755	0.711431	5.79	0.888434
		2	83.3	40.9	22.3	1.268E-03	3.793E-05	0.000183	83.58	0.17158	0.722473	5.79	0.867572
		3	133.4	40.9	22.5	2.025E-03	6.062E-05	0.000292	134.11	0.17375	0.731661	5.78	0.856816
		4	188.7	40.87	22.6	2.855E-03	8.557E-05	0.000412	190.12	0.17704	0.745728	5.77	0.840997
		7	256.8	40.8	22.7	3.873E-03	1.162E-04	0.000558	259.43	0.17856	0.752388	5.77	0.834032
3/3/98	9:40	1	115.41	60.6	23.2	1.655E-03	4.939E-05	0.000503	115.92	0.16658	0.707313	5.80	0.893641
		2	148.5	60.4	23.3	2.126E-03	6.351E-05	0.000324	149.34	0.17056	0.71816	5.79	0.872868
		3	220.5	60.54	23.6	3.143E-03	9.401E-05	0.000479	222.36	0.17241	0.726022	5.78	0.863640
		4	293.85	60.48	23.7	4.175E-03	1.250E-04	0.000636	297.15	0.17546	0.739059	5.78	0.848848
		7	378.04	60.25	24	5.353E-03	1.604E-04	0.000815	383.52	0.17678	0.744884	5.78	0.842813
3/3/98	10:55	1	195.54	81.2	23.6	2.637E-03	7.861E-05	0.000848	196.93	0.16522	0.701553	5.80	0.901062
		2	243.1	80.9	23.8	3.273E-03	9.764E-05	0.000527	245.26	0.16918	0.71233	5.80	0.880203
		3	335.9	80.9	24	4.503E-03	1.345E-04	0.000725	340.02	0.17077	0.71909	5.79	0.872281
		4	426	80.74	24.1	5.690E-03	1.701E-04	0.000916	432.65	0.17362	0.731306	5.79	0.858290
		7	525.25	80.5	24.5	6.991E-03	2.091E-04	0.001123	535.39	0.17478	0.736477	5.78	0.853021

Date	time	unit	Pressure	Temp	Temp	moles bed	moles in	moles out	Fugacity	moles	H/Pd	free bed	tainty
			psia	bed °C	out °C	V1	V2	V3	psia	H2 solid		V1 cc	H/PD
3/3/98	12:34	1	296.5	101.3	24.3	3.778E-03	1.124E-04	0.001277	299.56	0.16362	0.694742	5.81	0.910046
		2	357.8	100.85	24.6	4.552E-03	1.356E-04	0.000770	362.27	0.16762	0.705764	5.81	0.888713
		3	470.5	100.84	24.8	5.957E-03	1.776E-04	0.001007	478.25	0.16899	0.711596	5.80	0.881944
		4	575.9	100.66	25	7.264E-03	2.167E-04	0.001227	587.54	0.17169	0.723172	5.80	0.868566
		7	687.5	101.27	25.1	8.620E-03	2.573E-04	0.001457	704.10	0.17277	0.728001	5.80	0.863718
3/3/98	14:15	1	420.27	121.8	25	5.069E-03	1.506E-04	0.001797	426.14	0.16177	0.686892	5.82	0.920685
		2	495.5	121.25	25.3	5.966E-03	1.773E-04	0.001058	503.69	0.16587	0.698421	5.82	0.898541
		3	626.4	121.2	25.7	7.505E-03	2.232E-04	0.001329	639.52	0.16707	0.703534	5.82	0.892720
		4	745.6	120.95	26	8.898E-03	2.649E-04	0.001572	764.24	0.16966	0.714631	5.81	0.879776
		7	868.25	120.4	26.1	1.033E-02	3.076E-04	0.001821	893.62	0.17065	0.719059	5.81	0.875456
3/3/98	16:30	1	27.95	20.95	23.2	4.556E-04	1.362E-05	0.000122	27.98	0.16820	0.714172	5.79	0.885013
		2	40.1	20.8	23.2	6.530E-04	1.954E-05	0.000088	40.17	0.17231	0.725541	5.78	0.863871
		3	69.7	20.96	23.5	1.131E-03	3.390E-05	0.000152	69.90	0.17481	0.736122	5.77	0.851549
		4	106.85	21	23.7	1.730E-03	5.188E-05	0.000233	107.32	0.17838	0.751363	5.77	0.834555
		7	155.95	21	23.9	2.517E-03	7.557E-05	0.000339	156.96	0.18017	0.759196	5.76	0.826331
3/3/98	16:48	1	27.95	20.98	23.2	4.555E-04	1.362E-05	0.000122	27.98	0.16820	0.714172	5.79	0.885012
		2	40.17	20.8	23.2	6.541E-04	1.957E-05	0.000088	40.24	0.17231	0.725536	5.78	0.863877
		3	69.7	20.94	23.5	1.131E-03	3.390E-05	0.000152	69.90	0.17481	0.736122	5.77	0.851549
		4	106.8	20.95	23.7	1.729E-03	5.187E-05	0.000233	107.27	0.17838	0.751366	5.77	0.834552
		7	156	20.97	23.9	2.518E-03	7.560E-05	0.000339	157.01	0.18017	0.759191	5.76	0.826337

Compressibility constants -60 to 80 °C			
Const			
a1	2.246E-02	8.306E-04	-1.019E-06
a2	5.618E-02	-1.911E-04	1.566E-07
a3	-3.615E-03	-8.166E-06	3.014E-08
a4	-1.512E-04	2.755E-06	-4.672E-09

PCT Constants, Meyer 1995		
for the equation lnf=a-(b-cn)/T-dT		
a=	-0.3797	
b=	10653	
c=	12035	
d=	0.027373	

Units were opened with a half turn

	V1 cc unhydride	Pd gm	V2 cc in	V2 cc out	X-ducer no.	weight of unit (gm) before load	weight after load	weight H/Pd in grams
1	6.302	50.1167	0.173	1.56	H 25855	1648.465	1648.797	0.3320
2	6.394	50.5392	0.173	4.96	H 27164	1645.12	1645.446	0.3260
3	6.427	50.5348	0.173	4.96	H 25854	1635.168	1635.484	0.3160
4	6.381	50.5204	0.173	4.96	H 25851	1653.087	1653.393	0.3057
7	6.367	50.5015	0.173	0.78	P 60828	1658.495	1658.79	0.2953

						Moles in the gas phase							% Uncer
Date	time	unit	Pressure	Temp	Temp	moles bed	moles in	moles out	Fugacity	moles	H/Pd	free bed	tainty
			psia	bed °C	out °C	V1	V2	V3	psia	H2 solid		V1 cc	H/PD
3/10/98	9:32	1	299.56	121.42	23.6	0.00363	0.00011	0.00129	302.54	0.15971	0.67814	5.83	0.93230
		2	178.73	120.9	24.3	0.00218	0.00006	0.00246	179.79	0.157084	0.66142	5.84	0.94771
		3	100.54	120.9	24.3	0.00123	0.00004	0.00139	100.87	0.154118	0.64898	5.85	0.96587
		4	56.4	120.62	24.1	0.00070	0.00002	0.00078	56.51	0.150152	0.63247	5.86	0.99135
		7	32.37	120.2	24	0.00040	0.00001	0.00007	32.40	0.145996	0.61519	5.87	1.01957
3/10/98	11:26	1	198.92	100.96	23.6	0.00255	0.00008	0.00086	200.30	0.161259	0.68472	5.82	0.92321
		2	113.86	100.6	24.2	0.00147	0.00004	0.00157	114.31	0.158709	0.66826	5.83	0.93795
		3	57.76	100.6	24.3	0.00075	0.00002	0.00080	57.88	0.155207	0.65357	5.84	0.95907
		4	29.55	100.43	24.2	0.00038	0.00001	0.00041	29.58	0.150844	0.63538	5.86	0.98680
		7	15.813	100.1	24.2	0.00021	0.00001	0.00003	15.82	0.146232	0.61618	5.87	1.01791

Date	time	unit	Pressure psia	Temp bed °C	Temp out °C	moles bed V1	moles in V2	moles out V3	Fugacity psia	moles H2 solid	H/Pd	free bed V1 cc	% Uncertainty H/PD
3/10/98	12:31	1	121.45	80.79	23.2	0.00165	0.00005	0.00053	121.99	0.162521	0.69008	5.81	0.91596
		2	65.92	80.52	23.8	0.00090	0.00003	0.00091	66.08	0.159952	0.67349	5.82	0.93063
		3	30.18	80.75	24	0.00041	0.00001	0.00042	30.21	0.155932	0.65662	5.84	0.95461
		4	14.21	80.67	23.8	0.00019	0.00001	0.00020	14.22	0.15125	0.63709	5.85	0.98415
		7	7.05	80.38	23.8	0.00010	0.00000	0.00002	7.05	0.146364	0.61674	5.86	1.01700
3/10/98	13:18	1	65.92	60.45	23.3	0.00095	0.00003	0.00029	66.09	0.163481	0.69415	5.81	0.91055
		2	33.35	60.25	23.6	0.00048	0.00001	0.00046	33.39	0.16083	0.67719	5.82	0.92554
		3	13.57	60.35	23.7	0.00020	0.00001	0.00019	13.58	0.156383	0.65852	5.83	0.95185
		4	5.87	60.25	23.6	0.00009	0.00000	0.00008	5.87	0.151478	0.63805	5.85	0.98267
		7	2.75	60.1	23.6	0.00004	0.00000	0.00001	2.75	0.146432	0.61702	5.86	1.01653
3/10/98	15:32	1	31.51	40.42	23.2	0.00048	0.00001	0.00014	31.55	0.164111	0.69683	5.80	0.90704
		2	14.9	40.3	23.6	0.00023	0.00001	0.00021	14.91	0.161346	0.67936	5.81	0.92257
		3	5.54	40.5	23.7	0.00009	0.00000	0.00008	5.54	0.156609	0.65947	5.83	0.95048
		4	2.23	40.5	23.6	0.00003	0.00000	0.00003	2.23	0.151581	0.63848	5.84	0.98200
		7	0.982	40.4	23.7	0.00002	0.00000	0.00000	0.98	0.146461	0.61715	5.86	1.01632
3/10/98	18:42	1	12.92	20.36	22.3	0.00021	0.00001	0.00006	12.93	0.164471	0.69836	5.80	0.90505
		2	5.68	20.2	22.5	0.00009	0.00000	0.00008	5.68	0.161613	0.68049	5.81	0.92105
		3	1.92	20.33	22.5	0.00003	0.00000	0.00003	1.92	0.156714	0.65992	5.83	0.94984
		4	0.72	20.35	22.6	0.00001	0.00000	0.00001	0.72	0.151625	0.63867	5.84	0.98171
		7	0.297	20.37	22.6	0.00000	0.00000	0.00000	0.30	0.146473	0.6172	5.85	1.01624

Compressibility constants -60 to 80 °C			
Const			
a1	2.246E-02	8.306E-04	-1.019E-06
a2	5.618E-02	-1.911E-04	1.566E-07
a3	-3.615E-03	-8.166E-06	3.014E-08
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3	6.427	50.5348	0.173	4.96	H 25854	1635.168	1635.484	0.3160
4	6.381	50.5204	0.173	4.96	H 25851	1653.087	1653.393	0.3057
7	6.367	50.5015	0.173	0.78	P 60828	1658.495	1658.79	0.2953

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Date	time	unit	Pressure	Temp	Temp	moles bed	moles in	moles out	Fugacity	moles	H/Pd	free bed	tainty
			psia	bed °C	out °C	V1	V2	V3	psia	H2 solid		V1 cc	H/PD
3/10/98	9:32	1	299.56	121.42	23.6	0.00363	0.00011	0.00129	302.54	0.15971	0.67814	5.83	0.93230
		2	178.73	120.9	24.3	0.00218	0.00006	0.00246	179.79	0.157084	0.66142	5.84	0.94771
		3	100.54	120.9	24.3	0.00123	0.00004	0.00139	100.87	0.154118	0.64898	5.85	0.96587
		4	56.4	120.62	24.1	0.00070	0.00002	0.00078	56.51	0.150152	0.63247	5.86	0.99135
		7	32.37	120.2	24	0.00040	0.00001	0.00007	32.40	0.145996	0.61519	5.87	1.01957
3/10/98	11:26	1	198.92	100.96	23.6	0.00255	0.00008	0.00086	200.30	0.161259	0.68472	5.82	0.92321
		2	113.86	100.6	24.2	0.00147	0.00004	0.00157	114.31	0.158709	0.66826	5.83	0.93795
		3	57.76	100.6	24.3	0.00075	0.00002	0.00080	57.88	0.155207	0.65357	5.84	0.95907
		4	29.55	100.43	24.2	0.00038	0.00001	0.00041	29.58	0.150844	0.63538	5.86	0.98680
		7	15.813	100.1	24.2	0.00021	0.00001	0.00003	15.82	0.146232	0.61618	5.87	1.01791

Appendix B

```
      program fill2
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccc
c     !Date of modification. 11-28-96
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccc
c  This program is for gaseous replensihment & uses the following equation
s:
c     The separation factors need to be confirmed if a mixed gas is used!!
!
c     Relationships of two authors give significantly different results.
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccc
c     The accuracy of the PCT relationship in the beta region is ±0.5%
c         to 5,000 psia at 25¡C and 0 yrs. for pure gases.
c         lnf=a-(b-cn)/T+dT-2mew/RT (Meyer, 1995) beta region
c         lnf=c1+c2/T+c3(n-.3)-2mew/RT Plateau region (Ivar, Meyer Modifie
d)
c     z=1+ccP/T+dd(P/T)^2 (constants good to 1500 atm(22ksi))
c     Bed heating T=Tambient + DTexp(-k*age)
c     Separation factors:  The accuracy is not known and needs to be confi
rmed exp.
c     A flow program is used to determine the mass deficit from equilibriu
m.
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccc
      implicit none

      real*8  a(3),b(3),c(3),d(3),e(75),rr,r,k,sf(3,3),temp,tempa,
     1       phi,ptot,pold,mb(5),mbop(5),mbs(5),x(5),mpd,mt20b,mhe30b,
     2       moptot,vb,vrep,delp,zeff,vb0,mfn,nall,pall,yold,YY(3),
     3       tmp1,tmp2,tmp3,tmp4,tmp5,tmp6,tmp7,tmp8,tmp9,tmp10,tmp11,
     4       deld,delt,yoldd,yoldt,dtemp,aal1,ball,call1,dal1,aa,y(5),tot,
     5         cc(5),dd(5),ee(5),ff(5),gg(5),a1,a2,a3,a4,a5,de1,zold
      integer igas,i,nep
      character*30 tmp
      character*1 good
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccc
c     data infomation
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccc
      data a / 5.83, 32.39, 28.49 /                    ! PCT constants
      data b /12640., 15313., 15349. /                 ! PCT constants
      data c /12832., 12832., 13832. /                 ! PCT constants
      data d /0.01853, -.03127, -.02363 /              ! PCT constants
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccc
c     data for compressibility factors for:
c         Form of the equation: z=1 + a1(P/T) + a2(P/T)^2 + a3(P/T)^3 +a4(
P/T)^4 [1]
c     where a, b, c & d are function of temperature f(T):
c     a1 = b(1) + b(2)*T + b(3)*T^2    [2]
c     a2 = b(4) + b(5)*T + b(6)*T^2    [2] etc.
c     H2, D2, T2, He-3, He-4, & N2 (note N2 has 5 pressure terms)
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccc
      data e/
     1        .022456,8.3057e-4,-1.0193e-6,           !Z constants a1 H2
     2        .056181,-1.9111e-4,1.5657e-7,               ! Z constants a2
 H2
     3        -.0036149,-8.1655e-6,3.0139e-8,             ! Z constants a3
 H2
     4        -1.5121e-4,2.7545e-6,-4.6721e-9,            ! Z constants a4
 H2
```

```
     5         0,0,0,                                          ! Z constants a4
 H2
     1         .0062142,8.9821e-4,-1.1201e-6,                  ! Z constants a1 D2
     2          .058847,-2.1402e-4,2.0254e-7,                  ! Z constants a2
 D2
     3          -.0047847,1.3188e-6,1.2312e-8,                 ! Z constants a3
 D2
     4          -1.388e-5,1.6842e-6,-2.7094e-9,                ! Z constants a4
 D2
     5         0,0,0,                                          ! Z constants a4
 D2
     1         -2.109e-5,9.2529e-4,-1.1593e-6,                 ! Z constants a1 T2
     2          .06076,-2.2813e-4,2.273e-7,                    ! Z constants a2
 T2
     3          -.0055349,6.8348e-6,2.7755e-9,                 ! Z constants a3
 T2
     4          7.1281e-5,1.0614e-6,-1.6328e-9,                ! Z constants a4
 T2
     5         0,0,0,                                          ! Z constants a4
 T2
     1         .15849,-2.6866e-5,-5.2201e-8,                   ! Z constants a1 He-
3
     2          .0018328,-4.0134e-5,5.8146e-8,                 ! Z constants a2
 He-3
     3          -5.2376e-4,5.0454e-6,-6.9967e-9,               ! Z constants a3
 He-3
     4          6.6813e-6,-2.3159e-8,-2.0797e-10,              ! Z constants a4
 He-3
     5         0,0,0,                                          ! Z constants a4
 He-3
     1         -1.7618,.0085229,-9.695e-6,                     ! Z constants a1 N2
     2          1.3616,-.0053109,6.0426e-6,                    ! Z constants a2
 N2
     3          -.35382,.0012511,-1.3966e-6,                   ! Z constants a3
 N2
     4          .037694,-1.0501e-4,1.1648e-7,                  ! Z constants a4
 N2
     5          -.00096532,-2.4486e-7,0./                      ! Z constants a5
 N2
      rr = 1.9872                                             ! gas constant c
al/gmole ¡K
      r = 82.057                                              ! cm^3 atm/gmole
 ¡K
      k = 0.056216
      zold=1.0                                               ! dk constant 1/year
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccc
c     read & confirm input file
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccc
      call read input(mb,vb,vrep,tempa,mpd,mfn,dtemp,y,tot)
      good(1:1) = 'N'
      write(6,*) 'Are these parameters correct [y or n]?'
          read(5,*) good
      if (good(1:1) .eq. 'n' .or. good(1:1) .eq. 'N') then
          write(6,*) 'Please edit the file delivery.inp and restart'
          goto 999
      endif
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccc
c     initialize
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccc
      call initialize(mb,mbop,mt20b,mhe30b,ptot,pall,tempa,vb0,vb,YY,y,
     1  aall,ball,calll,dall,a,b,c,d,cc,dd,ee,ff,gg,x,zeff,a1,a2,a3,a4,a5,
```

```
e)
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccc
c     open output file fill2.out1
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccc
      open(unit=2,file='fill2.out1',status='unknown')
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccc
c     write headers for output
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccc
      write(6,910)' stoic(all)   ptot(psia)  pTDH(psia)  mbs(H2)     mbop(
H2)    vb(cc)       '
      write(2,910)' stoic(all)   ptot(psia)  pTDH(psia)   mbs(t2)      mbo
p(t2)     vb(cc)       '
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccc
c     determine stoic given P & T
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccc
1     temp=tempa+dtemp                                 ! bed temp. with self he
ating
      sf(2,1) = exp(277.5/temp - 0.025)                ! deuterium/protium sepa
ration factor
      sf(3,1) = exp(430.4/temp - 0.092)                ! tritium/protium separa
tion factor
      sf(3,2) = exp(133.5/temp - 0.021)                ! tritium/deuterium sepa
ration factor
      sf(1,2) = 1/sf(2,1)                           ! protium/deuterium separati
on factor
      sf(1,3) = 1/ sf(3,1)                          ! protium/tritium separation
 factor
      sf(2,3) = 1/ sf(3,2)                          ! deuterium/tritium separati
on factor
      phi = a1*(ptot/temp)+a2*(ptot/temp)**2/2+a3*(ptot/temp)**3/3+a4*(pto
t/temp)**4/4
      phi = phi + a5*(ptot/temp)**5/5
      phi = exp(phi)        ! fugacity coefficient
      write(6,*) 'phi=',phi
      tmp11=aall-ball/temp+dall*temp
      nall=mfn*(log(pall*phi)-tmp11)*temp/calll        ! total stoic
              ! total moles tritium
       tmp2=exp(11.017-1000.*4.006/temp+0.12*(nall-.3)) !plateau pressure
atm
      if ((pall-tmp2) .lt. .001) then
       vb=vb0-(mpd*106.4/12.02)*nall*(.179*x(3)+.182*x(2)+.185*x(1))
       nall = 2*(mb(1) + mb(2) + mb(3) - tmp2*(vb+vrep)/(zeff*r*temp))/mpd
      endif
2     continue
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccc
cc    determine the pressure in the system
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccc
      mb(4) = mhe30b       ! total moles helium
       vb=vb0-(mpd*106.4/12.02)*nall*(.179*x(3)+.182*x(2)+.185*x(1))
      do 16 i = 1,3
          mbs(i) = x(i)*nall*mpd/2.0                        ! hydrogen solid
16    continue
      mbs(4)=0.
      do 18 i = 1,4
          mbop(i) = mb(i) - mbs(i)                          ! moles overpres
sure
18    continue
```

```
c      sum moles of gas
       moptot = 0.0
       do 20 igas = 1,5
           moptot = moptot + mbop(igas)                          ! moles total ga
s
20     continue
           a1 = 0
           a2 = 0.
           a3 = 0.
           a4 = 0.
           a5 = 0.
       if(moptot .le. 0) then
           ptot=tmp2
           pall=tmp2
           goto1
           endif
           nep = 1
       do 40 i = 1,5
           cc(i) = e(nep) + e(nep+1)*temp + e(nep+2)*temp**2        !
           dd(i) = e(nep+3) + e(nep+4)*temp + e(nep+5)*temp**2      !
           ee(i) = e(nep+6) + e(nep+7)*temp + e(nep+8)*temp**2      !
           ff(i) = e(nep+9) + e(nep+10)*temp + e(nep+11)*temp**2        !
           gg(i) = e(nep+12) + e(nep+13)*temp + e(nep+14)*temp**2       !
40         nep = 5*3*i +1
       do 30 i = 1,5
           y(i) = mbop(i)/moptot                              ! mole fraction
           a1 = a1 + cc(i)*y(i)
           a2 = a2 + dd(i)*y(i)
           a3 = a3 + ee(i)*y(i)
           a4 = a4 + ff(i)*y(i)
           a5 = a5 + gg(i)*y(i)
30     continue
101    zeff = 1 + a1*(ptot/temp)+a2*(ptot/temp)**2+a3*(ptot/temp)**3+a4*(pt
ot/temp)**4
       zeff = zeff + a5*(ptot/temp)**5
       ptot = zeff*(moptot*r*temp)/(vb+vrep)        ! atm
       del = abs((zeff-zold)/zold)
       zold = zeff
c      write(6,*) 'nall,zeff,ptot,pall=',nall,zeff,ptot,pall
       if(del .gt. .0001) goto 101
       aa = calll/temp
       pall = (mb(1) + mb(2) + mb(3) + (1/aa - nall)*mpd/2.0)/
     1  ((vb+vrep)/(zeff*r*temp) + mpd/(pall*2.0*aa))      !partial hydroge
n pressure
       if ((pall - tmp2) .lt. .002) pall = tmp2         !plateau pressure
       delp = abs(pall-pold)
       pold = pall
       phi = a1*(ptot/temp)+a2*(ptot/temp)**2/2+a3*(ptot/temp)**3/3+a4*(pto
t/temp)**4/4
       phi = phi + a5*(ptot/temp)**5/5
       phi = exp(phi)       ! fugacity coefficient
       nall=mfn*(log(pall*phi)-tmp11)*temp/calll
       if ((pall - tmp2) .lt. 0.002) then
           pall=tmp2
        nall = 2*(mb(1) + mb(2) + mb(3) - pall*(vb+vrep)/(zeff*r*temp))/mpd
    !two phase
           endif
       if (delp .gt. 0.0001) goto 2
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
cccc
c      mixture convergence on x(i)'s and YY(i)'s given p and stoic
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
cccc
c      Yi*pall*v/ZRT=Mb(i)-x(i)*n*mpd/2
c    iteratating the combined three eq. for protium, deuterium & tritium
```

```
cccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
cccc
c    solving for fraction of protium in gas phase
cccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
cccc
      yold = YY(1)
220   tmp1 = pall*(vb+vrep)*(1. - sf(2,3))/(zeff*r*temp)
      tmp2 = pall*(vb+vrep)*(sf(2,1)*yold + sf(2,3)*(1. - yold))/(zeff*r*t
emp) +
     1      nall*mpd/2. + mb(2)*(sf(2,3) - 1.)
      tmp3 = -mb(2)*(sf(2,3)*(1. - yold) + sf(2,1)*yold)
      tmp4 = pall*(vb+vrep)*(sf(2,1) - sf(2,3))*tmp2/(zeff*r*temp) +
     1      2.*tmp1*mb(2)*(sf(2,1) - sf(2,3))
      tmp4=tmp4/(tmp2**2. - 4.*tmp1*tmp3)
      tmp5 = ((pall*(vb+vrep)*sf(2,3)/(zeff*r*temp) + nall*mpd/2 + mb(2)*(
sf(2,3) - 1))
     1      *tmp2 + 2.*tmp1*mb(2)*sf(2,3) - 2.*tmp1*tmp3)/(tmp2**2. - 4.*tm
p1*tmp3)
      tmp6 = tmp4*sqrt(tmp2**2. - 4.*tmp1*tmp3)/(2.*tmp1) - (sf(2,1) - sf(
2,3))/
     1      (2.*(1 - sf(2,3)))
      tmp7 = tmp5*sqrt(tmp2**2. - 4.*tmp1*tmp3)/(2.*tmp1) - sf(2,3)/
     1      (2.*(1. - sf(2,3))) - (nall*mpd/2. + mb(2)*(sf(2,3) -1))/(2.*tm
p1)
      tmp8 = pall*(vb+vrep)*((1. - sf(1,3)) + (sf(1,2) - sf(1,3))*tmp6)/(z
eff*r*temp)
      tmp9 = pall*(vb+vrep)*(sf(1,3) + (sf(1,2) - sf(1,3))*tmp7)/(zeff*r*t
emp)
      tmp9 = tmp9 + nall*mpd/2 + mb(1)*(sf(1,3) - 1. - (sf(1,2) - sf(1,3))
*tmp6)
      tmp10 = -mb(1)*(sf(1,3) + (sf(1,2) - sf(1,3))*tmp7)
      YY(1) = (-tmp9 + sqrt(tmp9**2. - 4.*tmp8*tmp10))/(2.*tmp8)
      if(YY(1) .lt. 0.) YY(1)=0.
      if(YY(1) .gt. 1.) YY(1)=1.
      yold=(yold+yy(1))/2.
      if (abs(YY(1) - yold) .gt. 0.0002) goto 220
      yold = YY(1)
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccccccc
c  tritium in gas phase
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccccccc
      tmp1 = pall*(vb+vrep)*(1.0 - sf(3,2))/(zeff*r*temp)
      tmp2 = pall*(vb+vrep)*(sf(3,1)*YY(1)+sf(3,2)*(1 - YY(1)))/(zeff*r*te
mp) +
     1      nall*mpd/2 + mb(3)*(sf(3,2) - 1)
      tmp3 = -mb(3)*(sf(3,2)*(1 - YY(1)) + sf(3,1)*YY(1))
      YY(3) = (-tmp2 + sqrt(tmp2**2 - 4*tmp1*tmp3))/(2*tmp1)
      if(YY(3) .gt. 1.) YY(3)=1.
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
cccccccc
c  deuterium in gas phase
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
cccccccc
      tmp1 = pall*(vb+vrep)*(1.0 - sf(2,3))/(zeff*r*temp)
      tmp2 = pall*(vb+vrep)*(sf(2,1)*YY(1)+sf(2,3)*(1 - YY(1)))/(zeff*r*te
mp) +
     1      nall*mpd/2 + mb(2)*(sf(2,3) - 1)
      tmp3 = -mb(2)*(sf(2,3)*(1 - YY(1)) + sf(2,1)*YY(1))
      YY(2) = (-tmp2 + sqrt(tmp2**2 - 4*tmp1*tmp3))/(2*tmp1)
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
cccccccc
c  protium, deuterium & tritium in solid phase
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
cccccccc
```

```
      x(1) = YY(1)/(YY(1) + sf(1,2)*YY(2) + sf(1,3)*YY(3))
      x(2) = YY(2)/(YY(2) + sf(2,3)*YY(3) + sf(2,1)*YY(1))
      x(3) = YY(3)/(YY(3) + sf(3,1)*YY(1) + sf(3,2)*YY(2))
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
cccccccc
c  calculate new constants for PCT
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
cccccccc
      aall = 0.0
      ball = 0.0
      calll = 0.0
      dall = 0.0
      do 310 i = 1,3
          aall = x(i)*a(i) + aall
          ball = x(i)*b(i) + ball
          calll = x(i)*c(i) + calll
          dall = x(i)*d(i) + dall
310   continue
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
cccccccc
c  convergence check
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
cccccccc
      deld=abs(yoldd-YY(2))
      yoldd=YY(2)
      delt=abs(yoldt-YY(3))
      yoldt=YY(3)
      if(deld .gt. 0.00001) goto 1
      if(delt .gt. 0.00001) goto 1
      write(6,*) 'converged  on bed'

      write(6,900) nall,ptot*14.696,pall*14.696,mbs(1),mbop(1),vb,phi*ptot
*14.696
      write(2,900) nall,ptot*14.696,pall*14.696,mbs(3),mbop(3),vb,phi*ptot
*14.696
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
cccccccc
      close(unit=2)
900   format(10e12.5)
910   format(a80)
999   stop
      end
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
cccccccc
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
cccccccc
      subroutine read input(mb,vb,vrep,tempa,mpd,mfn,dtemp,y,tot)
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
cccccccc
      implicit none
      real*8 mb(1),vb,vrep,tempa,mpd,mfn,dtemp,y(1),tot
      character*30 tmp
      integer igas
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
cccccccc
ccc   hydrogen, igas = 1
ccc deuterium, igas = 2
ccc tritium, igas = 3
ccc helium 3, igas = 4
ccc others, igas = 5
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
cccccccc
c     read inputs from file fill2.inp
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
cccccccc
```

```
      open(unit=1,file='fill2.inp',status='old')
c     bed gas
      call skip
      call skip
      call skip
      tot = 0.
      do 110 igas = 1,5
          read(1,*) mb(igas),tmp
          write(6,*) mb(igas),tmp
          tot = mb(igas) + tot
110   continue
          do 100 igas = 1,5
          y(igas) = mb(igas)/tot
          write(6,*) y(igas)
100   continue

c     Volumes
      call skip
      call skip
      call skip
          read(1,*) vb,tmp                                    ! unhydrided
 bed
          write(6,*) vb,tmp
          read(1,*) vrep,tmp                                  ! replen
          write(6,*) vrep,tmp
c     System temperature
      call skip
      call skip
      call skip
          read(1,*) tempa,tmp                                 ! ambient te
mp.
          write(6,*) tempa,tmp
          read(1,*) dtemp,tmp                                 ! bed heatin
g rise
          write(6,*) dtemp,tmp
      call skip
          read(1,*) mpd,tmp                                   ! grams pall
adium
          write(6,*) mpd,tmp
          mpd=mpd/106.4                                       ! moles pall
adium
      call skip
          read(1,*) mfn,tmp                                   ! stoic muli
tiplier
          write(6,*) mfn,tmp                                  ! for hydrog
ens
      call skip
      close (unit = 1)
      return
      end
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccccc
      subroutine skip
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccccc
      character*30 tmp
      read(1,*) tmp
      write (6,*) tmp
      return
      end
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccccc
      subroutine initialize(mb,mbop,mt20b,mhe30b,ptot,pall,tempa,vb0,vb,YY
,y,
     1  aall,ball,calll,dall,a,b,c,d,cc,dd,ee,ff,gg,x,zeff,a1,a2,a3,a4,a5,
```

```
e)
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccccc
      implicit none
      real*8 mb(1),mbop(1),mt20b,mhe30b,ptot,pall,tempa,vb0,vb,YY(1),aall,
ball,
     1     calll,dall,a(1),b(1),c(1),d(1),cc(1),dd(1),x(1),zeff,a1,a2,a3,a
4,a5,e(1),
     2    ee(1),ff(1),gg(1),y(1)
      integer i,nep
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccccc
c     initial conditions
ccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
ccccc
      zeff=1.
      mt20b = mb(3)
      mhe30b = mb(4)
      ptot = 50.0                    ! atm total pressure
      pall=ptot
      mbop(5) = mb(5)
      tempa = tempa + 273.15
      vb0=vb
      aall=0.
      ball=0.
      calll=0.
      dall=0.
      do 10 i=1,3
          YY(i) = mb(i)/(mb(1)+mb(2)+mb(3))
          x(i) = YY(i)
          aall=x(i)*a(i) + aall
          ball=x(i)*b(i) + ball
          calll=x(i)*c(i) + calll
          dall=x(i)*d(i) + dall
10    continue
      nep = 1
      do 40 i = 1,5
          cc(i) = e(nep) + e(nep+1)*tempa + e(nep+2)*tempa**2      !
          dd(i) = e(nep+3) + e(nep+4)*tempa + e(nep+5)*tempa**2        !
          ee(i) = e(nep+6) + e(nep+7)*tempa + e(nep+8)*tempa**2        !
          ff(i) = e(nep+9) + e(nep+10)*tempa + e(nep+11)*tempa**2      !
          gg(i) = e(nep+12) + e(nep+13)*tempa + e(nep+14)*tempa**2
!
40        nep = 5*3*i +1
          a1 = 0.
          a2 = 0.
          a3 = 0.
          a4 = 0.
          a5 = 0.
      do 30 i = 1,5
          a1 = a1 + cc(i)*y(i)
          a2 = a2 + dd(i)*y(i)
          a3 = a3 + ee(i)*y(i)
          a4 = a4 + ff(i)*y(i)
          a5 = a5 + gg(i)*y(i)
30    continue
      return
      end
```

Data From Wicke and Nernst, and Perminov, Orlov and Frumkin													
Wicke :	Constants A and B												
deg C	lnP=-A+B*(H/Pd)		H/Pd										
	A	B											
50	26.6124247	42.2	0.695										
30	30.2124247	46	0.715										
0	35.5124247	51	0.749			Perminov, Orlov and Frumkin							
-78.5	52.9124247	66.4	0.838			Pressure	Fugacity	H/Pd	H/Pd	H/Pd	H/Pd	H/Pd	
Press	H/Pd (calc)	Wicke and Nernst				(atm)	(Meyer)	meas	meas	meas	meas	meas	
(psia)	-78.5 deg C	0 C	30 C	50 C				-78.5 C	-32 C	0 C	50 C	100 C	
1	0.79687387	0.69632	0.657	0.63		0.0146	0.21456	0.84					
2	0.80731283	0.70991	0.672	0.65		0.112	1.6461	0.854	0.787	0.717	0.676	0.605	
3	0.81341923	0.71786	0.681	0.66		1	14.707	0.87	0.831	0.784	0.761		
4	0.81775179	0.7235	0.687	0.66		30	451	0.894	0.847	0.808	0.792	0.735	
5	0.82111239	0.72788	0.692	0.67		100	1588.4	0.903	0.857			0.741	
6	0.82385819	0.73145	0.696	0.67		200	3452.6	0.908		0.829	0.819		
7	0.82617974	0.73448	0.699	0.68		300	5654		0.865	0.835		0.766	
8	0.82819076	0.7371	0.702	0.68		400	8260.1	0.913	0.868	0.84	0.822	0.769	
9	0.8299646	0.7394	0.705	0.68		500	11346	0.915	0.871	0.843			
10	0.83155135	0.74147	0.707	0.69		600	14992	0.916	0.874	0.848			
11	0.83298675	0.74334	0.709	0.69		750	21714		0.877	0.853			
12	0.83429716	0.74505	0.711	0.69		1000	37127	0.92		0.859			
13	0.83550262	0.74662	0.713	0.69		1700	1.26E+05						
14	0.8366187	0.74807	0.714	0.69									
15	0.83765775	0.74942	0.716	0.69									

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