LA-UR-98-2111

Title: Ultrasonically Determined Fill Pressure and Density in Closed Spherical Shells

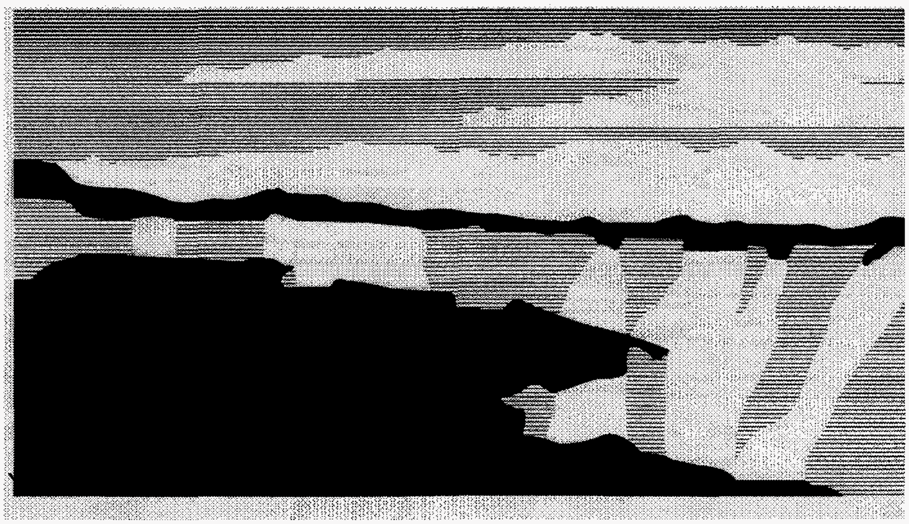
CONF-980496--

Author(s): T. Asaki

MASTER

Submitted to: Target Fabrication Specialists Meeting

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED



Los Alamos
NATIONAL LABORATORY

Form No. 836 R5
ST 2629 10/91

DISCLAIMER

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Ultrasonically determined fill pressure and density in closed spherical shells.

Thomas J. Asaki

Condensed Matter and Thermal Physics Group

MST-10, Mail Stop K764

Los Alamos National Laboratory

Los Alamos, New Mexico 87545

Abstract

Experiments have been conducted in which the D_2 fill pressure has been determined for several closed millimeter-size aluminum and beryllium shells. The vibrational resonance frequency spectrum of the shells was used to calculate the sound velocity of the interior gas. This velocity, along with the equation-of-state, determined the gas pressure and density. The accuracy in determining the fill conditions is within 0.5% in both pressure and density for near critical density ($\rho \gtrsim 9$ mol/L) gas over a wide range of temperatures (190 K to 300K). Reduced accuracy was apparent at low density. An attempt was made to determine the fill density of one shell by acoustic observation of the dew point temperature. While this temperature was recorded very accurately, the uncertainty in the saturated vapor density curve near the critical point yielded inaccurate results. These methods were shown to be unaffected by small deviations in the sphericity of the gas-filled cavity.

I. Introduction

Inertial confinement fusion (ICF) targets present a unique challenge for gas properties measurement. The success of these millimeter-size shells as a fusion target requires that they be highly spherically symmetric. The ideal shell encloses a cavity without defects (e.g. fill tubes or seams). The filling of these shells has progressed along one of several paths including hemispherical shell bonding under high pressure, the use of temperature-dependent semipermeable materials, and diffusion-bonded micron-sized plugs. In any case, it is generally not possible to accurately determine the resultant fill pressure by standard means.

This paper focuses on the determination of the fill pressure of closed spherical shells through the use of resonant ultrasound spectroscopy (RUS). Asaki *et al.*[1] suggested that the fill pressure (as well as other target and gas properties) can be accurately determined from the elastic/fluid resonance spectrum of a spherical target. A brief summary of the ideas relevant to pressure determination are presented here. The target is assumed to belong to the set of objects whose physical properties depend only upon the radial coordinate *r*. The resonant mode structures and frequencies of the target are then computed numerically in a one-dimensional calculation.[2] The first class of resonant modes (toroidal) is characterized by nonradial shear motion in the shell. These modes do not couple into the interior gas except through weak boundary layer effects and are not of direct interest in this study. The second class of resonant modes (spheroidal) is characterized by motion involving both compressional and shear effects. Motion can be purely radial or can exhibit more complicated shape oscillations. In the limit that the interior cavity is evacuated, the spheroidal modes reduce to the spheroidal modes of an elastic shell. In the limit of an infinitely rigid shell, the spheroidal modes reduce to the acoustic modes of a spherical cavity. Beryllium and beryllium/copper mixtures characterize a large class of shells of interest to the ICF community. Since beryllium is an exceptionally stiff material, the spheroidal modes can be effectively divided into elastic shell modes and acoustic cavity modes. This division of modes is only a classification of convenience. Pure elastic shell modes and pure acoustic cavity modes do not exist in these shells, and the distinction vanishes when two types of modes are nearly degenerate. With these cautions, a general spherical cavity resonance mode notation will be used here. In an ideal cavity the resonant frequencies are given by

$$f_{ns} = \frac{c z_{ns}}{2\pi a} \tag{1}$$

where a is the cavity radius, c is the speed of sound in the gas, and z_{ns} is the s^{th} root of

$$\frac{\partial j_n(z)}{\partial z} = 0 \tag{2}$$

and $j_n(z)$ is the n^{th} order spherical Bessel function.

It is also important to note that any deviations in the cavity from sphericity will break the degeneracy of nonradial modes. The number of observable resonance peaks per mode is $2n+1$. Consider the spherical harmonic decomposition of the shape of a nearly spherical cavity:

$$r = a\left[1 - \sum_{\ell,m} \varepsilon_{\ell m} Y_\ell^m(\theta,\phi)\right]. \tag{3}$$

For small perturbation amplitudes $\varepsilon_{\ell m}/a \ll 1$, the average of the single mode frequencies is unshifted to order ε/a. Thus, insofar as the cavity volume is known, small deviations from sphericity should not affect the measurements.[3]

II. Experimental

Resonant ultrasound spectroscopy (RUS) was the method for observing the elastic/fluid target response. In this scheme, two or more acoustic transducers served as target mounts. One transducer was used as a frequency-swept sine-wave excitation source. The target was driven into a resonant vibrational mode as the source frequency passed through the resonance. Any of the remaining transducers were used as a receiver. The response signal was passed through a heterodyne receiver yielding a real-time amplitude response frequency spectrum. An excellent and complete review of this technique is given in Ref. 4 and need not be given here.

The target mount consisted of a tetrahedral array of pinducers. Three were rigidly mounted within a stainless steel base ring. The fourth was vertically mounted in a sleeve of a top plate and a small spring provided a clamping tension. Each pinducer element was capped with a small piece of alumina in the shape of a rounded cone. This wear plate served as an efficient sound channel and allowed for the mounting of very small targets.

A liquid helium flow-through cryostat was designed to accommodate the target mount. The cryostat design included electrical feed-throughs for the transducers and a gas handling manifold for in situ filling of sample targets with fill tubes. A combination of diode and germanium resistance sensors at the target mount provided temperature measurement accuracy of 5 mK below 100 K and 10 mK above 100 K. Temperature control stability was better than 10 mK over the entire range 20 K to 300 K.

Two methods were used to determine the gas density within the closed spherical shell. The first method begins with the use of Eq. (1) to calculate the sound speed of the gas. Because of the modest accuracy requirements (0.5% in pressure) various corrections for soft boundary effects, as well as boundary layer dissipative effects can be ignored. The sound speed, along with the temperature T and a suitable equation-of-state,[5] then yields the gas density and pressure. This method requires knowledge of the cavity radius a. Experiments can be carried out at a wide range of temperatures and with several acoustic modes for statistical averaging. Calculations for a wide range of temperatures must include the thermal expansion and contraction of the shell. This first method is designated the EOS method.

A second method was also used to determine the initial fill conditions of one of the targets used in this study. Along an isochor, the sound speed passes through a minimum at the dew point temperature T_D. Thus, by slowly varying the temperature while observing a single resonance, T_D can be determined very accurately. The density is then determined through a saturated vapor density expression $\rho_V(T_D)$ or possibly an equation of state. The density determination is independent of the size and shape of the cavity. This method is designated the dew-point method.

The frequency used in a calculation of the sound speed must be accompanied by positive mode identification. This involves both the distinction of acoustic modes from the background of elastic shell modes and proper identification of the indices (n and s) associated with a given resonance. The elastic shell modes of targets made with hemispheres and containing other anisotropies (e.g. fill tubes or adhesives) have low values of the resonance Q, typically a few hundred. The acoustic cavity modes, relatively unaffected by shell material details, have larger Q's on the order of a few thousand. This alone serves to distinguish the modes of interest. Once the acoustic cavity modes are identified and the frequencies listed, it only remains to show that the ratios of frequencies match the ratios of the roots of Eq. (2).

Additional consideration must be given to the observation of degenerate modes. The degree of frequency splitting is on the order of the nonsphericity ε/a while the average frequency remains unshifted to this order.[3] Thus, it is appropriate to compute an approximate frequency average given by the unweighted average of the observed resonances for a given mode. The relative uncertainty introduced by this computation will be no greater than (and typically much less than) ε/a . Frequencies observed in highly nonspherical cavities may not yield reliable results.

Several deuterium-filled sample targets were examined by the EOS method. One target was also examined by the dew-point method. The results are described in the following three sections.

III. AL53

The AL53 is an aluminum shell of 5.00 mm outer radius and 3.00 mm inner radius constructed of two hemispheres that were diffusion bonded. It was filled with D_2 (0.2% He-3) at nominal room temperature through a fill tube to a pressure of 5240 psia and the fill tube was pinched closed. The closed fill tube remains an integral part of this sample target. The construction and filling took place approximately 7 years prior to the present study.

Acoustic cavity mode frequency f_{20} was observed at six temperatures ranging from 190.00 K to 294.80 K. Since the assembly temperature remains unknown, for the calculation it was assumed that the cavity radius was 3.000 mm at 298 K. Taking into account the thermal contraction of the shell, it was found from these measurements that the target pressure P = 5244(21) psia and density ρ = 11.95 mol/L at 298 K. This good agreement with the reported fill pressure may be somewhat fortuitous considering the uncertainties in the fill conditions and construction. The precision of measurements over this wide range of temperatures suggests that the accuracy is limited by knowledge of the cavity size. The presence of He-3 has been ignored, although a simple mass correction to the sound speed indicates a probable error of less than 0.15%.

The dew point temperature in the AL53 was measured to be 37.859(5) mK by observing the minimum in the frequency f_{24}. Various calculations of the dew point density ρ_D and pressure P_D are summarized in Table 1. Friedman *et al.*[6] made direct measurements of the saturated vapor pressure and the reported pressure is based upon their four-constant equation valid from 29 K to the critical temperature $T_C = 38.34$ K. Young and Souers[7] provide approximate relations for the saturated vapor properties based upon the 1967 NBS EOS. The close agreement with the pressure of Friedman *et al.* is due to the fact that their data was used in constructing the vapor pressure equation. Finally, the saturated vapor properties calculated by the NIST-12 EOS[5] are reported. Clearly, the saturated vapor density is not well known in the vicinity of the critical point and the fill density of the AL53 cannot be determined with any reasonable accuracy. The technique itself does not appear to be in question and it can be expected to yield significantly improved results for cavities not filled to near critical density.

IV. BE43

The BE43 is a beryllium shell of 4.00 mm outer radius and 3.00 mm inner radius. This target was also constructed of two hemispheres and retains an open stainless steel fill tube (4 mil ID, 9 mil OD). The target was mounted within the cryostat and the fill tube attached to a deuterium gas manifold exterior to the cryostat. The target was initially evacuated then cryogenically filled by cooling to 20 K while connected to a large D_2 reservoir (158 cm^3). Once the target was filled with liquid D_2 it was isolated from the reservoir and manifold by closing the manifold valve nearest the target. The cryostat was then allowed to warm to room temperature, the target and fill line (volume ≈ 3.50 cm^3) now containing deuterium gas at elevated pressures. The remainder of an experiment proceeded as follows. Cavity mode acoustic frequencies were measured and the gas pressure calculated by the EOS method. The gas was expanded into a small manifold volume (4.57 cm^3) which was connected to a pressure gauge (Precise Sensors 0-500 psi strain gauge, accuracy $\pm$ 0.1 psi). The acoustic calculations were repeated at this new pressure. The target was isolated from the manifold and the manifold was evacuated. Then the process was repeated beginning with expansion of the gas into the manifold. In this way a single target filling allows data to be taken at several different pressures. Since no direct reading of the pressure is available for the highest test pressure (before the initial fill was expanded into the manifold), it must be calculated based upon the manifold, target, and fill line volumes.

Two complete experiments were carried out at 297.45 $\pm$ 0.15 K and the combined results are shown in Fig. 1. The calculations were based upon observation of the lowest frequency mode f_{11}. Data are represented by open circles and equal pressures are indicated by the solid line. The agreement is quite good considering that the pressure dependence of the sound speed is quite weak. In fact, the largest pressure discrepancy represents only a 0.2% difference in sound speed. The systematic overestimation of the pressure is most likely due to the equation-of-state calculation for which even somewhat greater uncertainties in the sound speed may be common for certain regions of the phase diagram. Thermal and viscous boundary effects, bulk viscosity, and shell admittance are expected to have even less effect on the sound velocity calculation.[8]

V. BE31-n Series

The BE31-n series of shells were constructed of beryllium hemispheres of 3.000 mm outer radius and variable inner radii of approximately 1.05 mm. Each hemisphere was machined with specific axisymmetric perturbations for the purposes of another study. Spherical Harmonic decomposition of the shapes reveals that the maximum perturbation amplitude for any shell

$\varepsilon_{\ell 0}^{\max} / a = 0.0237$ and, thus, to a good approximation it is only necessary to consider a spherical cavity of volume equal to that of the machined cavity. The exception to the above discussion is the spherically machined hemishells for constructing shell #1.

The fill pressures were determined by the EOS method for each of the shells for temperatures ranging from 200 K to 300 K. The results are shown in Table II. Each set of hemishells was adhesively bonded in a deuterium atmosphere at temperature T_0 and pressure P_0. The EOS calculated pressure P_A and standard deviation of the measurements σP_A are given as corrected to temperature T_0. With the exception of shell #5, which is discussed in detail below, each calculated pressure significantly exceeds P_0. The large range of calculated pressures, from 4407 to 5225 psia, suggests that systematic metrologic or calculation errors are not responsible. Rather, evaluation of the assembly process reveals that the pressure within the cavity could rise to as much as 1.5 times the ambient pressure (~6000 psia) during the final stage of hemishell joining. It is believed that the calculated pressures P_A reflect an accurate measure of the cavity pressure and as such are not directly comparable to the ambient construction conditions P_0.

In addition, the standard errors associated with shells #3 and #4 are unreasonably large. The degeneracy splitting observed for the acoustic modes in these shells was quite large (2% - 4%) compared to that of the other shells (0.2% to 1%). This suggests that the degree of nonsphericity in these two shells is great enough induce mode frequency shifts unaccounted for by this calculation. Indeed, the shells with the greatest degree of machined nonsphericity were these two shells. However, it is not clear that the small differences in machining should make such a significant impact on the acoustically determined nonsphericity. It appears that very precise pressure measurements can be made for cavities whose acoustic modes show degeneracy splitting of 1% or less.

The pressure in shell #5 was initially measured at 1996 psia on March 16, 1998. Subsequent measurements revealed a decrease in the pressure over time. These results are shown in Fig. 2 in which the acoustically determined pressure is plotted for seven days. The exponential fit to the data is

$$P = 4961 e^{-D/18.8} \quad (4)$$

where the pressure P (psia) is given at any time D (days elapsed since midnight March 1, 1998). This equation gives a pressure "half-life" of 13.0 days. Unfortunately, the exact date and time of the hemishell bonding has been lost, but occurred during the period February 26 to March 6 ($D \approx$ -1.5 to 6.5 days). The wide range of pressures computed by Eq. (4) for these dates (3500-5400 psia) renders any figure for the initial fill pressure unreliable.

VI. Conclusion

Resonant Ultrasound Spectroscopy has been used to determine the fill pressure and density of closed spherical shells. It is believed that the acoustically determined pressures and densities are accurate to within 0.5% for D_2 fill densities greater than about 9 mol/L. Low-pressure measurements were made at reduced accuracy. These measurements rely on the accuracy of the deuterium equation-of-state.

A second method of determining the fill density requires the acoustic observation of the dew point temperature. This experiment was performed with the AL53, but was limited severely by the lack of knowledge of the saturated vapor density near the critical point. Conversely, this technique suggests a method for measuring the saturated vapor density curve.

This work is supported by the U.S. Department of Energy under contract number W7405-ENG36.

References

1. Asaki, T. J., Hoffer, J. K., and Sheliak, J. D., "Ultrasonic characterization of inertial confinement fusion targets," *Fusion Tech.* **33**, 171 (1998).
2. Alterman, Z., Jarosch, H., and Pekeris, C. L., "Oscillations of the earth," *Proc. Roy. Soc. London*, **252**, 80 (1959).
3. Mehl, J. B., "Acoustic resonance frequencies of deformed spherical resonators. II," J. Acoust. Soc. Am., 79, 278 (1986).
4. Migliori, A. and Sarrao, J. L., "Resonant ultrasound spectroscopy: Applications to physics, materials measurement and nondestructive evaluation," John Wiley & Sons, New York (1997).
5. The deuterium equation-of-state used in the calculations was that of the NIST Standard Reference Database 12 (1992) provided by the Fluid Mixtures Data Center, Thermophysical Division, NIST, Boulder, CO 80303.
6. Friedman, A. S., White, D., and Johnston, H. L., "The direct determination of the critical temperature and critical pressure of normal deuterium. Vapor pressures between the boiling and critical points," J. Am. Chem. Soc. 73, 1310 (1951).
7. Young, D. A. and Souers, P. C., "A three phase equation of state model for the hydrogen isotopes," Lawrence Livermore National Laboratory Report #UCRL-83829 (1980).
8. Moldover, M. R., Mehl, J. B. and Greenspan, M., "Gas-filled spherical resonators: Theory and experiment," *J. Acoust. Soc. Am.* **79**, 253 (1986).

Figure Captions

Fig. 1. Room temperature deuterium pressure measurements in the BE43. The measured pressures are those determined by a pressure gauge connected to the cavity space via a gas manifold and fill tube. The calculated pressures are those given by the acoustically determined sound velocity and the D_2 equation-of-state. The solid line indicates pressure equality. The deviation at the highest pressures likely reflects the uncertainties associated with an equation-of-state calculation. In addition, the two highest measured pressures have an additional uncertainty related to uncertain volumes in the gas handling system (see text).

Fig. 2. Acoustically determined pressure for the BE31-5 over the course of one week. This shell was shown to leak with a pressure "half-life" of 13.0 days.

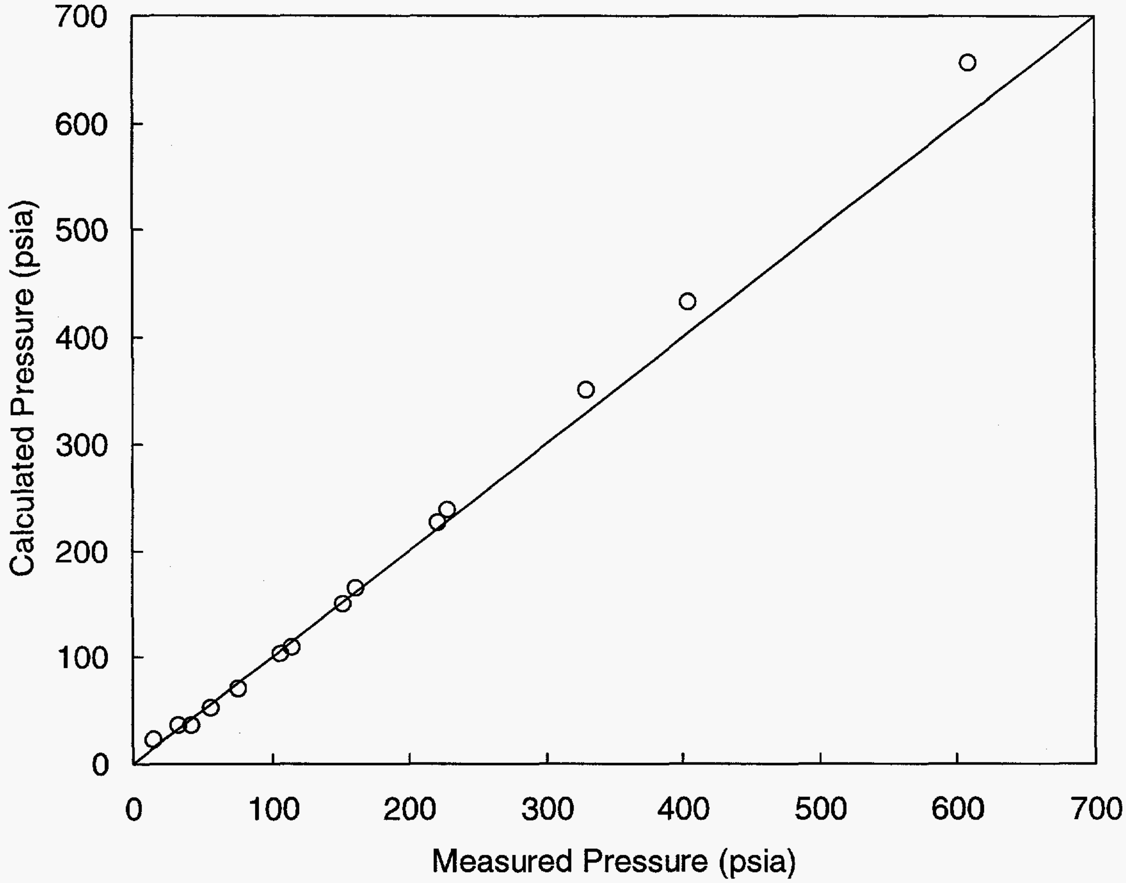
Table I. Saturated vapor pressure and saturated vapor density calculated from various sources for a dew point temperature of $T_D = 37.859$ K.

Source	P_D (psia)	ρ_D (mol/L)
Friedman, *et al.*[a]	226.3	----
Young and Souers[b]	226.6	8.873[c]
NIST-12	236.8[c]	15.12[c]

[a]Calculation based upon a four-constant equation derived from data taken between 29 K to 38.34 K. [b]Calculated from approximate relations based upon the 1967 NBS EOS. [c]Uncertainties of calculated parameters can be large near the critical point.

Table II. Reported D_2 fill temperature T_0 and pressure P_0 for each of the BE31 target series shells. The acoustically determined fill pressure P_A (at T_0) is given along with the standard deviation of measurements associated with a given shell σP_A and the number of acoustic modes N used in the calculation.

	BE31-1	BE31-2	BE31-3	BE31-4	BE31-5	BE31-6
T_0 (K)	292.5	296.0	295.4	295.8	~296	296.1
P_0 (psia)	4066	4084	4031	4092	~4050	4090
P_A (psia)	4407	4569	4553	4419	1996	5225
σP_A (psia)	7	9	545	290	3	18
N	5	7	3	3	3	5



Calculated Pressure (psia)
Measured Pressure (psia)
0
100
200
300
400
500
600
700

Figure 1

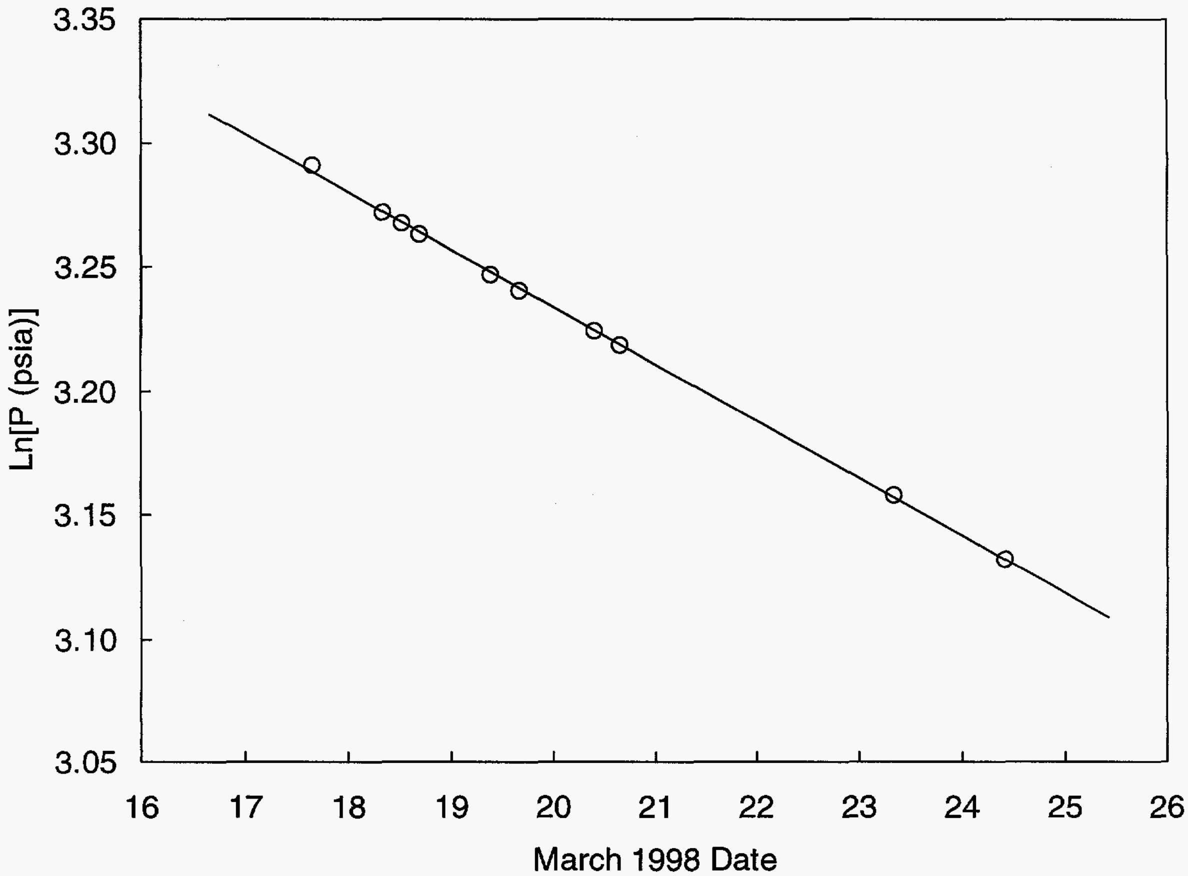

3.35
3.30
3.25
3.20
3.15
3.10
3.05
Ln[P (psia)]
16
17
18
19
20
21
22
23
24
25
26
March 1998 Date

Figure 2