Development of a Technique for the Leak Testing of Elastomeric Seal Materials at High Temperature*

D.R. Bronowski, K.B. Sorenson
Sandia National Laboratories

INTRODUCTION

Performance requirements for radioactive material (RAM) packages are specified in Title 10, Code of Federal Regulations, Part 71 (NRC 1993). Package components that form the containment boundary must function in both high and low temperature environments which are characteristic of the hypothetical fire accident and the -40°F (-40°C) normal transport condition, respectively. Seals that provide the containment system interface between the packaging body and closure(s) are a source of special consideration when designing, testing, and licensing a RAM package. These seals are usually elastomeric O-rings or gaskets and are tested frequently during development and operational use.

Typically, a helium mass spectrometer leak detector (MSLD) or pressure change type of system is used to test the leaktightness of package seals. The type of system used is determined primarily by required test sensitivity. In the MSLD system, helium is used as the tracer gas due to its small molecular size and high activity, which results in higher sensitivity. Such tests are conducted for fabrication or preshipment assembly verification and are conducted at or near ambient temperature conditions.

While there is little reason to perform a leakage test on operational packages while at elevated temperatures, such a test may be required during a package development program or as a component verification test. For these tests, the commonly used helium MSLD system encounters problems. As temperatures increase, elastomeric seals become more permeable, and the helium tracer gas more easily permeates through the material. This permeation can effectively mask real leakage around the seal, rendering the measurement useless.

---

* This work was performed by Sandia National Laboratories, Albuquerque, New Mexico, supported by the United States Department of Energy under Contract No. DE-AC04-94AL85000.

Residual gas analyzers (RGAs) have been used as an alternative to a helium MSLD in seal material leak testing work performed by Sandia National Laboratories. Unlike a common MSLD which is tuned to a single gas, an RGA can measure either an entire gas spectrum or any selected gas. This allows for the use of alternate tracer gases. A tracer gas with a larger molecule size (higher molecular weight) can be selected that slows permeation while still maintaining the required sensitivity. Possible candidates for tracer gases include most noble gases and halogen compounds. During the course of a test program on elastomeric seal materials, the use of an RGA with alternate tracer gases has been developed and refined.

BACKGROUND

When monitoring a leak detector, two types of leakage may be observed: bypass leakage and permeation leakage. For bypass leakage, tracer gas flows across the seal at seal/fixture interfaces and is identified by a near immediate (less than 5 seconds) signal increase with a rapid rise time to an equilibrium rate. Permeation leakage, which is tracer gas passing through the seal material, is identified by a time-delayed signal with a slower rise time. For permeation leakage, time delay, rise time, and equilibrium rate are dependent on the tracer gas, pressure, temperature, and seal material. Figure 1 illustrates helium permeation through silicone material.

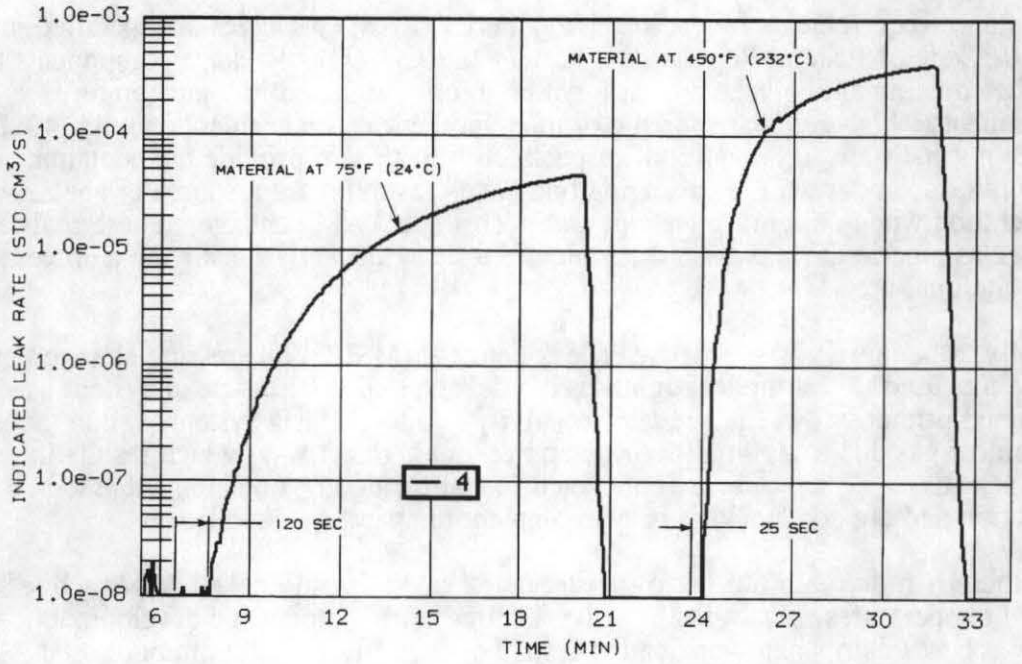

Figure 1. Helium Permeation Through Silicone at 75°F (24°C) and 450°F (232°)

The leak test system developed for the test program is illustrated in Figure 2. The leak detector consisted of a residual gas analyzer integrated into a cart-mounted pumping system. The RGA is a quadrapole type system with a 1 to 100 AMU (atomic mass unit) range. Data collection was made by a Hewlett Packard Series 9000 computer in conjunction with an HP3852 data acquisition unit. Software was developed for valve control, pressure monitoring, and automatic system logs for quality assurance purposes. An environmental chamber was used to heat the fixtures.

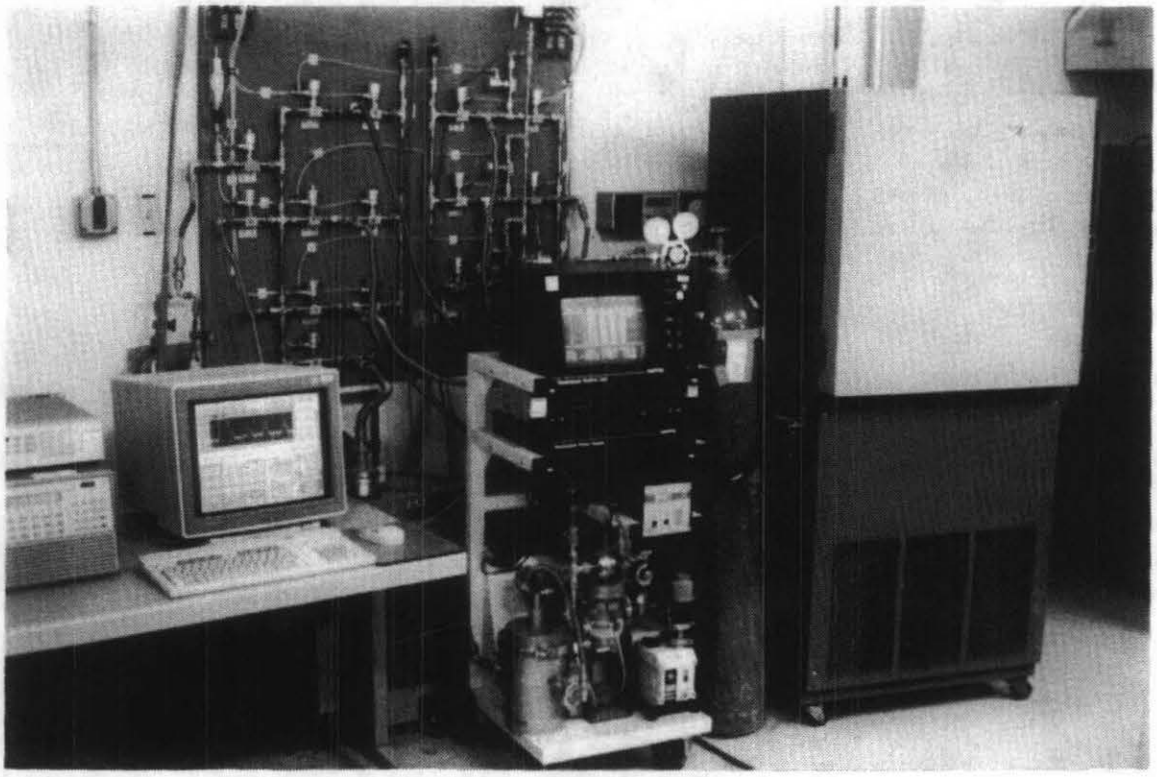

Figure 2. Leak Test System

The test fixture configuration used for the program is illustrated in Figure 3. This design is a face seal configuration in which the compressive force is applied across the O-ring thickness. The leak detector was connected to the central cavity of the fixture, and the area between the two seals was used for the tracer gas cavity. This reduced the amount of seal surface area exposed to the detector, reducing outgassing rates. This configuration also eliminated the possibility of atmospheric gases permeating through the outer seal and raising the test system background. As scaling laws for leakage rates do not exist, all measurements from this program were intended to be used as guidance rather than as a quantitative measure of seal performance.

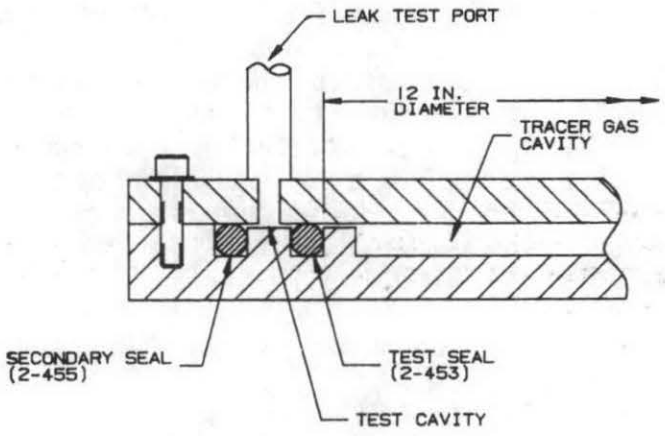

Figure 3. Seal Test Fixture

An RGA measures the partial pressures of the remaining atmosphere constituents in an evacuated system. Like an MSLD, the RGA output signal is linear with the amount of gas in the system. As shown in Figure 4, linearity is maintained so long as backgrounds remain stable (no other significant changes other than tracer gas) and total system load (real leakage plus outgassing) remains within the pumping capacity of the leak test system. Whereas the actual RGA output is proportional to partial pressure, most RGAs have a multiplier function that can relate the instrumental output directly to leakage rate units.

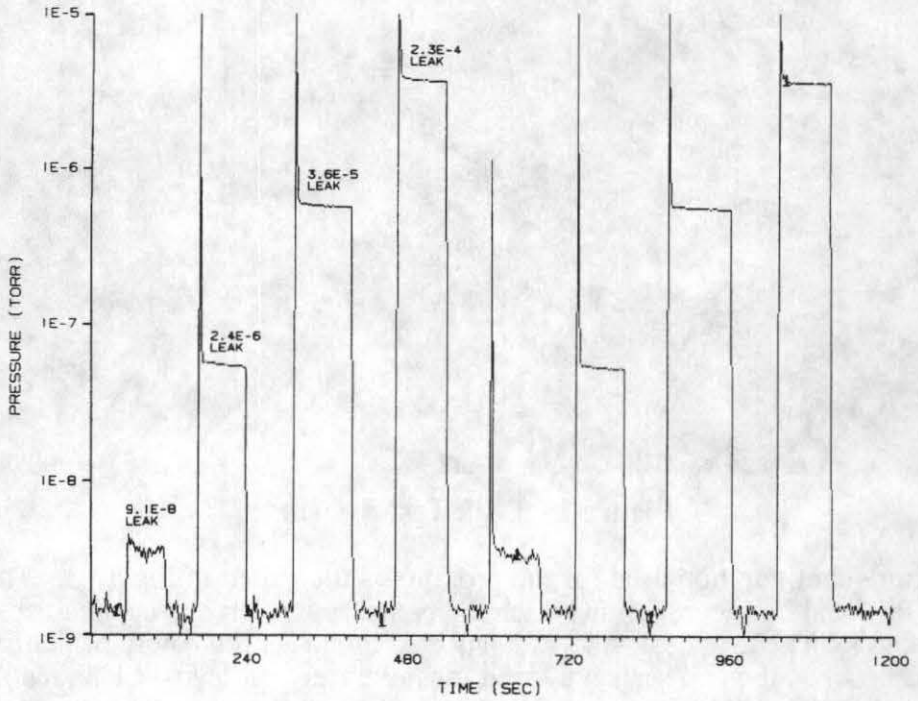

Figure 4. Linearity Demonstration Using Various Leak Standards

TEST METHOD DEVELOPMENT

As previously mentioned, possible tracer gases might include most noble gases and halogens. A primary consideration is to select a gas with a mass peak or peaks that are not hidden by normal system backgrounds. Figure 5 illustrates the background which exists in a normal low-vacuum system. This would be considered "dirty" by most vacuum standards but is indicative of a system which has been recently vented to the atmosphere and has had limited pump-down time. Extra effort to reduce these levels is of little use when the background due to an elastomeric seal material is added. Figure 6 shows the background resulting from a silicone O-ring at room temperature. Very limited regions exist in this spectra for selection of a tracer gas. Neon at mass 20 and argon at mass 40 fall in high background areas, as does sulfur hexafluoride (SF_6) with major mass peaks at 89, 54, and 35.

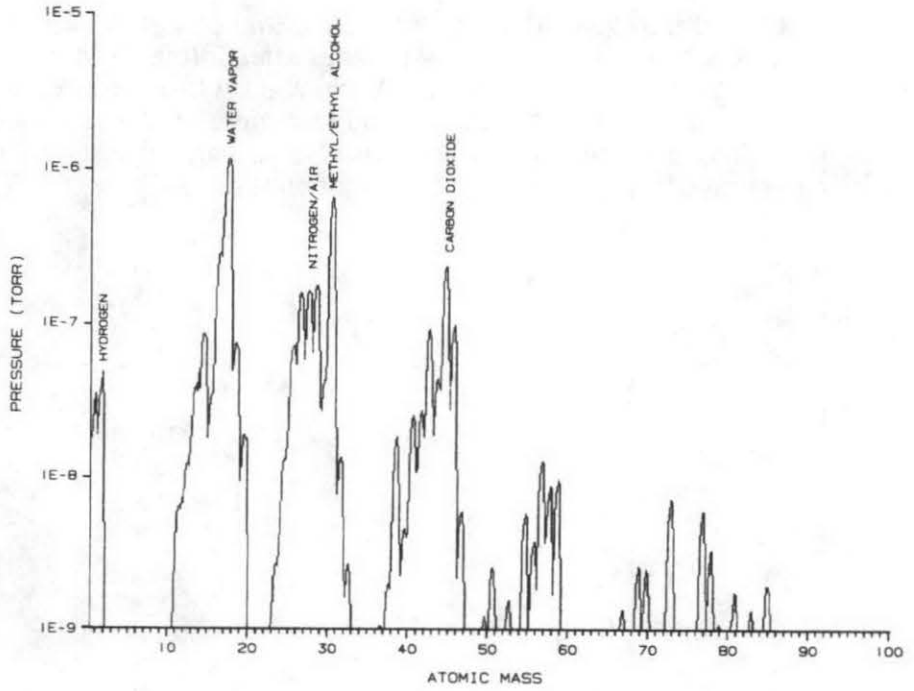

Figure 5. Leak Test System Background

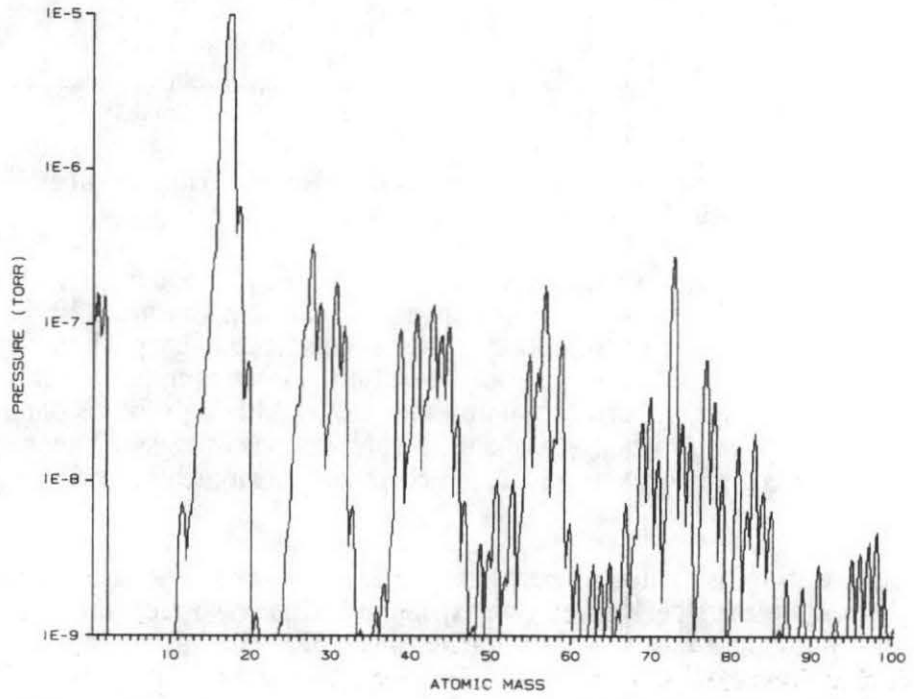

Figure 6. Spectrum of Silicone Material at Room Temperature

This spectra became even worse when the seal material was heated. Outgassing rates increased dramatically with the material temperature. It was also found that most seal materials, when heated, produce constituents which contaminate and desensitize the RGA. Parallel pumping systems were tried with little improvement. At this point, it became obvious that a trapping system was required to protect the RGA.

A simple, low-cost liquid nitrogen (LN) trap was added to the system. Figure 7 shows the spectra of the same silicone material a few minutes after filling an inline LN trap. The trap proved acceptable in protecting the RGA and significantly reduced overall system pressures, but with a drawback. Many candidate gases, particularly halogens, were completely condensed in the trap. Argon was also partially affected by the trap, presumably being entrained by other condensing compounds.

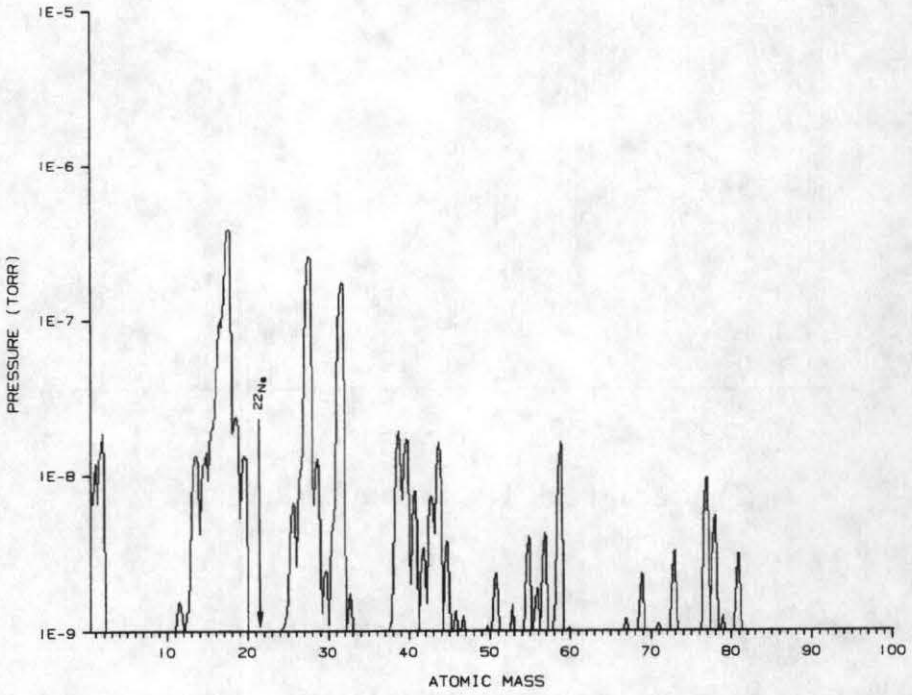

Figure 7. Spectrum of Silicone Material at Room Temperature With a Liquid Nitrogen Cold Trap in the System

These problems encountered with argon, halogens, and SF_6, made neon a primary candidate. While unaffected by the cold trap, its primary peak at mass 20 lies in a relatively contaminated area of the spectra. Neon has a secondary peak that shows up at mass 22. This peak is a combination of the naturally occurring neon isotope and the fragmentation or cracking pattern created by the RGA. Although the secondary peak is only 10% of the primary peak, seal material backgrounds at mass 22 averaged only 1 to 3% of that at mass 20. This provided an acceptably clean area for tracer gas measurements.

In preliminary testing using neon tracer gas, the RGA showed leak rate sensitivity in the E-7 cubic centimeters per second (cm^3/s) range. This was acceptable though 2 orders of magnitude less sensitive than a helium MSLD. The larger mass of neon slowed permeation breakthough time by a factor of 2 versus helium. This follows classic kinetic gas theory which shows that diffusion time varies directly with the square root of the molecular weight, other factors being equal. The square root of the molecular weight ratio of neon and helium is 2.2, consistent with observations. This doubling of permeation breakthough time was sufficient to obtain reliable measurements in many seal compounds with the exception of silicone compounds. While permeation and real leakage data show different overall shapes, the initial rise time was still so rapid for silicones that differentiating between initial permeation and a small leak was difficult and not easily proved. An additional problem with

defensible data interpretation was the somewhat slow sampling rate by the data acquisition system. System programming caused difficulty in relating the precise time of tracer gas backfill (i.e. the start of the test) to the acquired data which was delayed in its display to the operator.

The final tracer gas selected was a 50/50 mixture of helium and neon. The mixture served two functions: 1) the helium signal provided the primary leak rate measurement since it has the highest signal-to-noise ratio and thus the highest sensitivity; and 2) the mass 22 isotope of neon could be simultaneously monitored for response. A simultaneous rise in both signals indicated a real leak, whereas a delayed response between the two signals denoted permeation (Figure 8). This gas mixture was also unaffected by the use of a liquid nitrogen cold trap.

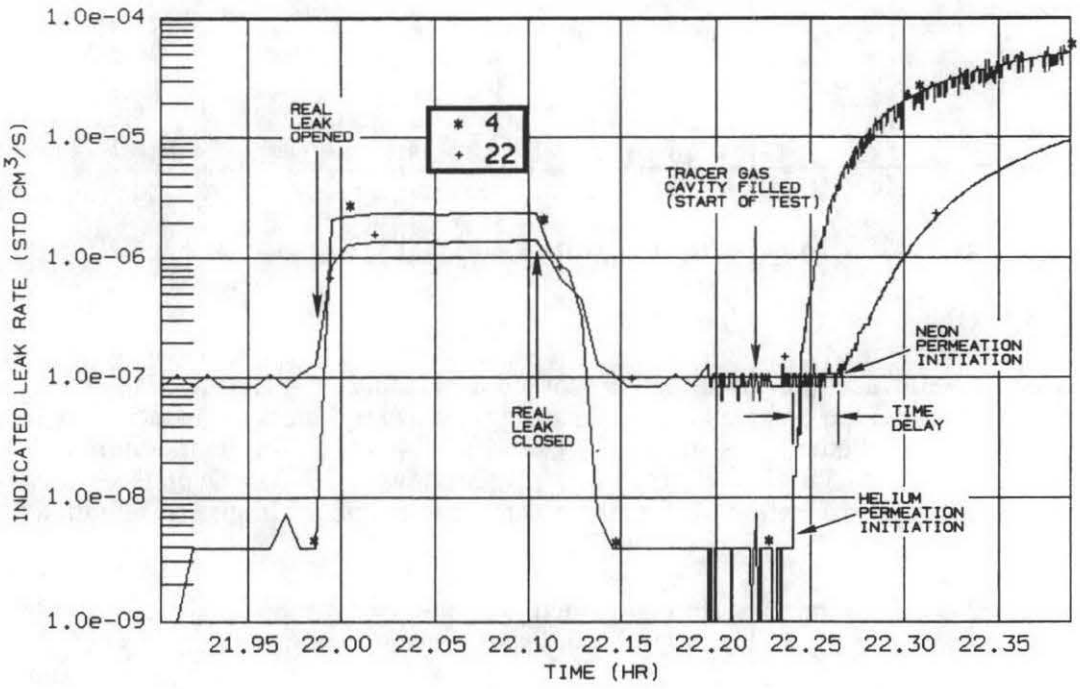

Figure 8. Bypass Versus Permeation Leakage Using Helium/Neon Mixture

In practice, the data were easily interpreted. Figure 9 shows a test of three fixtures containing various seals. After an initial calibration check to verify system operation, each fixture was sequentially valved into the system, allowed to stabilize, and then backfilled with tracer gas to test. Signals were monitored for either a simultaneous or delayed rise. Each of these three fixtures were demonstrated to have no detectable leakage as there were the expected delays in permeation breakthough. All leak data were obtained from the helium signal; the neon 22 signal was used for interpretational and backup purposes.

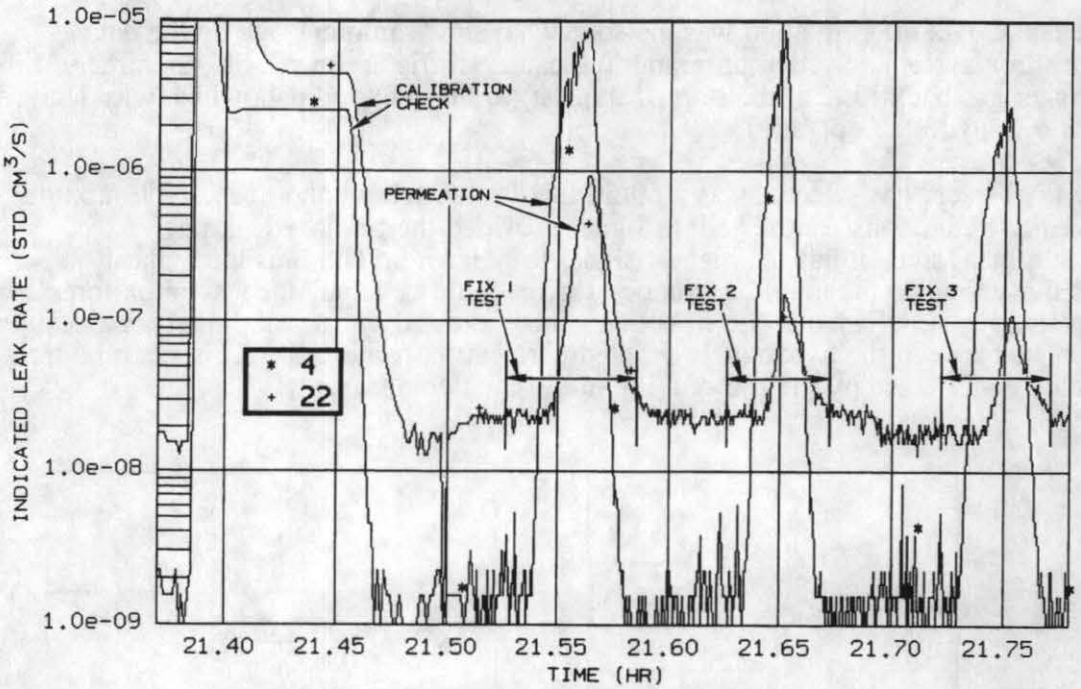

Figure 9. Example Leak Test Data

CONCLUSIONS

The system/mixed tracer gas combination worked acceptably well for the designed purpose. It provided the defensible data regarding the leaktightness of materials in this test series. The method was successfully used for 240 tests with temperatures ranging from 300°F to 550°F (149°C to 288°C) (Bronowski 1995). Overall sensitivity was very good at approximately 5E-9 cm^3/s for most tests due to the use of helium as the primary indicator.

The system requires a moderate amount of maintenance (daily trap cleaning, quadrapole cleaning every 50 hours). Step-by-step procedures must be strictly followed to prevent miscues such as inadvertent venting to atmosphere or allowing the LN trap to warm while pumping, both of which quickly contaminate the RGA.

An RGA system such as this is not recommended for routine leakage rate measurements. RGAs can be temperamental and require a higher level of understanding and maintenance and should only be considered when a system cannot utilize helium as tracer gas.

REFERENCES

Bronowski, D.R., *Performance Testing of Elastomeric Seal Materials Under Low- and High-Temperature Conditions - Final Report,* SAND 94-2207, Sandia National Laboratories, Albuquerque, NM (1995).

U.S. Nuclear Regulatory Commision, Title 10, Code of Federal Regulations, Part 71, Washington, DC (1993)