LA-UR-17-25093

Approved for public release; distribution is unlimited.

Title:	Technical basis for inner container leak detection sensitivity goals in 3013 DE surveillance
Author(s):	Berg, John M.
Intended for:	Report
Issued:	2017-08-11 (rev.1)

Disclaimer:
Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the Los Alamos National Security, LLC for the National Nuclear Security Administration of the U.S. Department of Energy under contract DE-AC52-06NA25396. By approving this article, the publisher recognizes that the U.S. Government retains nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

Technical basis for inner container leak detection sensitivity goals in 3013 DE surveillance

White paper prepared for MIS Working Group

Revised August 2017

John M. Berg, Los Alamos National Laboratory

Executive Summary

Helium leak checking of 3013 inner container lids is under consideration for addition to DE Surveillance tasks as an improved means to detect any through-wall flaws that may have formed during storage. This white paper evaluates whether leak checking at DE could replace and improve upon the current method of comparing gas compositions and pressures within the inner and outer containers. We have used viscous and molecular flow equations in ANSI N14.5 to calculate what the measured standard helium leak rate would be for hypothetical leaks of three different sizes. For comparison, we have also calculated the effects on gas composition and pressure differences as a function of pre-DE storage time for the same three leak sizes, using molecular and viscous flow equations as well as diffusion equations to predict the relevant gas transport. For a hypothetical leak that would be measured at 1×10^{-7} std cc/sec, likely an achievable sensitivity using helium leak checking at DE, the calculations predict no measurable effect on pressure difference or gas composition as measured by DE gas analysis. We also calculate that it would take over 200 years for water vapor to diffuse through a 10^{-7} std cc/sec leak enough to raise the RH outer container to half the RH value in the inner container. A leak 100 times larger, which would be measured at 1×10^{-5} std cc/sec, the same water vapor diffusion would take at least 14 years. Our conclusion is that helium leak checking will be useful even at a sensitivity of 1×10^{-5} std cc/sec, and a significant improvement over current DE methods at a sensitivity of 1×10^{-7} std cc/sec.

Introduction

Helium leak checking of 3013 inner container lids is being evaluated as an option for inclusion in destructive examination (DE) surveillance to confirm that the inner container has maintained its integrity to gas leaks throughout the storage period. Performance criteria need to be established and compared with achievable sensitivity before a decision can be made on whether to go forward with this option. To enable this comparison, we will use standard gas transport models incorporating viscous and molecular flow, as well as diffusion, to predict the detectability of several leak sizes using both standard helium leak checking and using the current method of comparing gas pressure and composition differences between the inner container headspace (IC) and outer container headspace (OI) at DE. To clarify, the term OI is taken from DE gas analysis sampling and reporting. It is an abbreviation of outer-inner, meaning the gas space between the outer and inner container walls.

A secondary goal of this paper is to consider what size of through-wall flaw might be considered problematic for overall 3013 integrity. We consider defining such a flaw as one large enough to allow leakage of corrosive gases to the outer container wall in sufficient quantity during the storage period that the outer container environment could become even slightly corrosive. We will look at water vapor transport from the IC as one key limitation on corrosivity, since some data exists on the internal humidity that can be used in these calculations. We will also look at the partial pressures of chlorine-containing gases that would be needed in the inner container headspace to transport corrosion-enabling quantities of chlorine to the outer container headspace, where it would be available to deposit on the outer container surface.

Potential leak test sensitivity goals:

We consider the following possible drivers for a leak test sensitivity criterion for the inner container at DE:

1. The leak test could target performing as well or better than the current method of using DE gas sampling and analysis to detect a leak. It should be able to detect:
    a. Minimum leak that would allow the observed differences in gas concentrations between OI and IC to have persisted through the storage period prior to DE.
    b. Minimum leak rate and leak duration combinations that could support observed pressure differences at DE.
2. The leak test could target detecting the minimum leak size through which H_2O vapor could diffuse in sufficient quantity during the storage period raise the outer container to a potentially corrosion-enabling RH.
    a. One advantage of this approach is that one could more reasonably pick a threshold problematic value for the total H_2O transfer through the leak based on how much H_2O would be needed to reach a problematic RH in the outer container, say 20% to be reasonably conservative.
    b. One could also make assumptions about the partial pressure range of H_2O in the inner container based on packaging glovebox limits and DE measurements.
3. The leak test could target detecting a leak that could pass sufficient chlorine-containing gas to enable corrosion.
    a. Total Cl leak that exceeds the surface coverage needed for localized corrosion on the outer container weld. I think one needs the Cl-containing gas concentration in the inner to get its leak rate relative to the measured leak rate for He at 1 std atm.
    b. Cl gases are reactive, so they have to leak out somewhat quickly or they will be consumed by reaction with the convenience and inner container walls. I don't have a good lifetime to use in this calculation to turn it into a quantitative adjustment to the acceptable leak rate.

Assumptions and simplifications used throughout these calculations

1. When there is an overall gas pressure difference between the inner and outer containers, the leak rate will be well represented by Equation B.5 of ANSI 14.5. This

formulation covers both viscous and molecular flow regimes. The flow rate depends on the properties of the gas mixture, but there is no fractionation of the mixture flowing through the leak from the high-pressure side to the low-pressure side.

2. Where the pressure difference between the inner and outer container is negligible, the leak rate of individual gas components of the inner container will be described by the diffusion alone, driven by differences in the upstream (IC) and downstream (OI) partial pressures of individual components of the gas mixtures. Fick's first law applies to the diffusion, so the concentration gradient through the hypothetical cylindrical leak is linear with distance through the wall. Mixing is assumed to be fast in the IC and OI volumes outside the leak path, so time-dependent concentrations at each end of the leak path are pinned at the average concentrations in the inner and outer container volumes at each time.
3. The effects of temperature differences between the leak tests and storage conditions are ignored.

Calculation methods for gas transport through idealized cylindrical leaks

Continuum and molecular flow calculation method

When there is a sufficient difference in pressure between IC and OI to drive either continuum or free molecular flow through the leak, Equations B.1-B.5 in Appendix B of ANSI N14.5 [ANSI 2014] can be used to calculate PV flow as functions leak diameter and length, upstream and downstream pressures, and molecular mass and gas viscosity. They are reproduced below as Equations 1-5. The mass flow in PV units, designated Q in Eq. 1, is obtained by multiplying volume flow, *L*, by the pressure, *P*. In these equations, the subscript *a* designates an average and the subscripts *u* and *d* designate that the subscripted parameter applies to the upstream or downstream end of the leak path.

$$Q = L \times P \tag{1}$$

$$L_a = (F_c + F_m)(P_u - P_d)\,cm/s \tag{2}$$

$$F_c = [2.49x10^3\ diam^3]/(a \times \mu)\ cm\ /atm \times s \tag{3}$$

$$F_m = [3.81x10^3\ diam^3\ (T/M)^{0.5}]/(a \times P_a)\ cm^3/atm \times s \tag{4}$$

$$L_u = (F_c + F_m)(P_u - P_d)(P_a/P_u)\ cm^3/s \tag{5}$$

F_c and F_m are the coefficients of continuum flow and free molecular flow conductance, *T* is the temperature, *M* is the molar mass, μ is the viscosity, and *a* and *diam* are the length and diameter of a cylindrical leakage hole. See ANSI N14.5 for full definitions of parameters, explanations of the equations, and sample calculations.

Diffusion calculation method

When the pressure difference is small enough, transport predicted by the continuum and molecular flow equations will be exceeded by diffusion of individual component gases driven by concentration gradients across the leak. Diffusion transport can be calculated using Fick's first law, Eq. 6, which gives the diffusion rate of component gas 1 in a mixture as a function of concentrations of the 1 along the leak path through the container wall.

$$J_1 = -AD\frac{\partial c_1}{\partial z} \tag{6}$$

In Eq. 6, J_1 is the flux of component gas 1 through a cross-sectional area A. That flux is the product of A, the diffusion coefficient, *D*, and the concentration gradient of component *1* in the direction of the flux, $\partial c_1/\partial z$.

Tabulated experimental values of *D* are available for many binary mixtures of common gases at 1 bar and near room temperature. For example, for a N_2-He mixture *D* is 0.794 cm^2/sec at 44 °C, for H_2O-He *D* is 0.908 cm^2/s at 25 °C, and for H_2O-N_2 *D* is 0.293 cm^2/s at 24 °C [Cussler 2009]. Multicomponent mixtures introduce complications because in principle the diffusion of each gas depends on the all other components in the mixture, but in many cases the binary diffusion coefficients can be used to get reasonable estimates of flux. Of particular relevance to us, this simplification applies if one is interested in diffusion of trace gases where a single gas forms the bulk of the mixture, or if two gases make up the bulk of a the mixture and one is only interested in the flux of one of those major gases. It is also helpful that temperature dependences of *D* values are weak [Cussler 2009], so reasonable estimates for diffusion rates at storage temperatures can be made without correcting *D* from tabulated values.

Using partial pressure as the concentration unit, the flux is expressed in PV units and Eq. 6 becomes

$$J_{1,\mathrm{PV}} = -AD\frac{\partial p_1}{\partial z} \tag{7}$$

For our case of slow leakage over a long leak time (years), the gas volumes outside the leak path can be assumed to be well mixed at all times, so the only non-zero partial pressure gradient is within the leak path. The partial pressure gradient along the leak path is essentially constant at any time and is determined by the difference between the partial pressures at the upstream and downstream ends of the leak divided by the leak path length. This allows a further simplification of Eq. 6 to

$$J_{1,\mathrm{PV}} = -AD\frac{p_{1d} - p_{1u}}{a} \tag{8}$$

For the case where the only leakage removes or adds gas, *i.e.* where the diffusing gas is neither consumed nor produced in either the upstream or downstream reservoirs, Eqs. 9 and 10 are the rate equations for the partial pressures of the diffusing gas as functions of time.

$$p_{1d}'[t] = \frac{J_{1,\mathrm{PV}}}{V_d} = -\frac{A\,D}{a\,V_d} \times (p_{1d}[t] - p_{1u}[t]) \tag{9}$$

$$p_{1u}'[t] = -\frac{J_{1,\mathrm{PV}}}{V_u} = \frac{A\,D}{a\,V_u} \times (p_{1d}[t] - p_{1u}[t]) \tag{10}$$

Under the initial condition of no diffusing gas in the OI, $p_{1d}[0] = 0$, the analytical solutions are

$$p_{1d}[t] = -\frac{\left(-1 + e^{-\frac{ADt(V_d+V_u)}{zV_dV_u}}\right)p_{\mathrm{u}0}V_u}{V_d + V_u} \tag{11}$$

$$p_{1u}[t] = \frac{p_{1\mathrm{u}0}\left(e^{-\frac{ADt(V_d+V_u)}{zV_dV_u}}V_d + V_u\right)}{V_d + V_u} \tag{12}$$

If the upstream partial pressure is held constant at its initial value during leakage, for example by replenishment from a condensed phase source term in the inner container, then Eq. 13 replaces Eq. 12.

$$p_{1u}'[t] = 0 \tag{13}$$

and the solutions simplify to

$$p_{1d}[t] = (1 - e^{-\frac{ADt}{zV_d}})p_{1\mathrm{u}0} \tag{14}$$

$$p_{1u}[t] = p_{1\mathrm{u}0} \tag{15}$$

These results will be used below to calculate the predicted pressure evolution in the IC and OI when diffusion is the primary flow mechanism through a leak.

Leakage and Pressure Calculation Results

Duffey placed the observed IC-OI pressure differences at DE in context by calculating that it would take over 13 years to equalize the typical 20 kPa IC-OI pressure difference in a Rocky Flats 3013 containers at a constant gas leak rate of 1x10^{-7} std cc/sec [Duffey 2014]. Here we expand on that idea by assuming leaks of various sizes as defined by their measured standard helium leak rates and explicitly calculating the decay of IC-OI pressure difference as a function of time.

We begin by defining three different cylindrical through-wall flaws as reference points, sized to produce predicted standard helium leak rates of 1x10^{-5}, 1x10^{-6} and 1x10^{-7} std cm^3/sec. To calculate their diameters we substitute the hypothetical standard leak rates, the wall thickness of 1.5 mm, the molar mass and viscosity of helium, and the standard test pressure of 1 atm into Equation 5 and solve for the diameters that would produce these standard leak rates. The diameters are 3.0, 1.5 and 0.74 microns respectively.

Pressure difference observed at DE compared with a standard leak test

Having defined the three reference leaks, we use Equations 1-5 to get expressions for continuum flow leak rates and pressure changes with time in storage for each of the three leaks sizes as functions of upstream and downstream gas conditions. The results can be used to judge the ability to detect different leak sizes using IC and OI pressure measurements at DE.

Figure 1 shows the predicted pressure evolution from hypothetical initial conditions of 100 kPa of helium in the IC and 90 kPa of helium in the OI. The half-lives for the decay of the initial 10 kPa pressure difference are 0.42, 4.5, and 46 years for the three different hole sizes. Figure 2 shows the same calculated pressure evolution using air instead of helium as the fill gas in both the IC and OI. The air results are qualitatively similar to helium but with somewhat slower pressure difference decays.

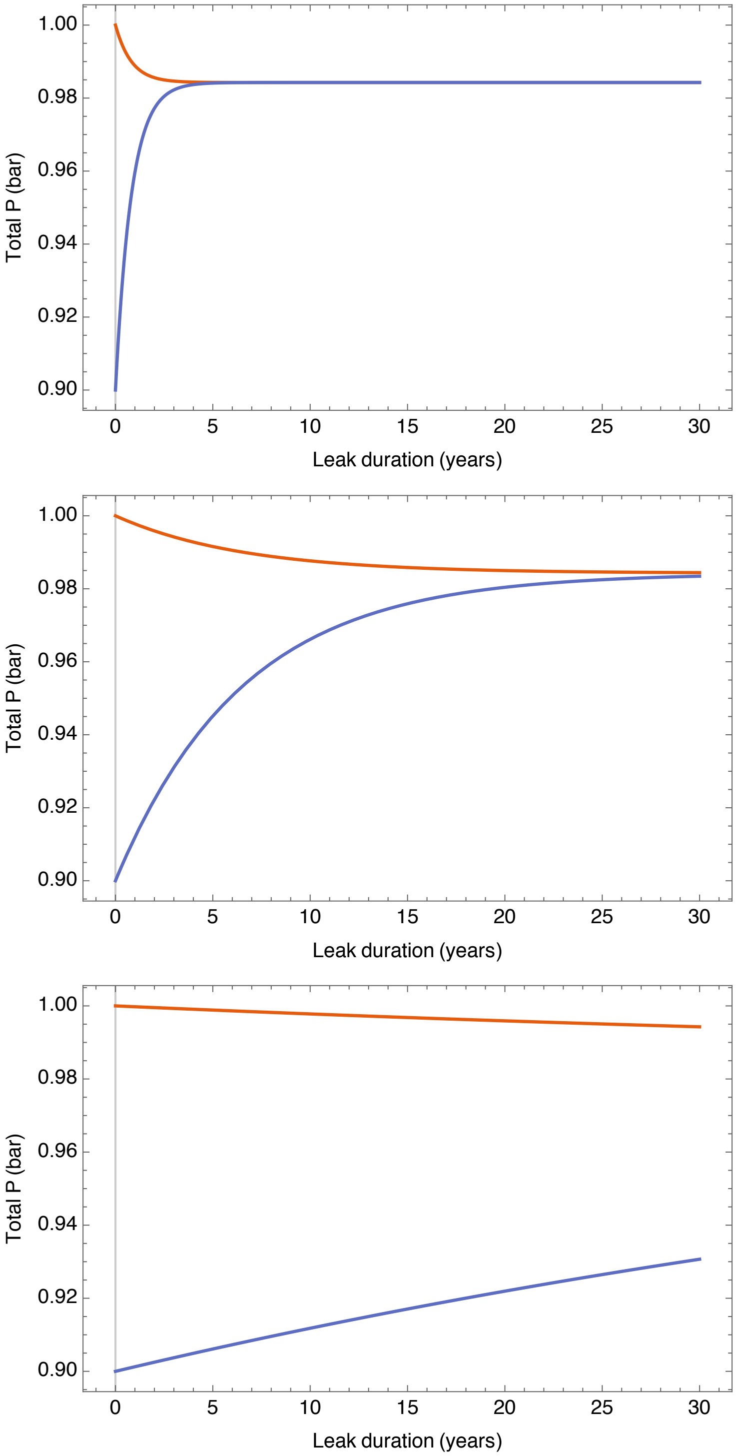

Figure 1: Predicted evolution of IC pressure (top/orange) and OI pressure (bottom/blue) as a function of time for ***helium*** *leakage through cylindrical holes with diameters of 3.0 microns (top), 1.5 microns (middle), and 0.74 microns (bottom). The initial conditions are set at 1.0 bar IC pressure and 0.9 bar OI pressure.*

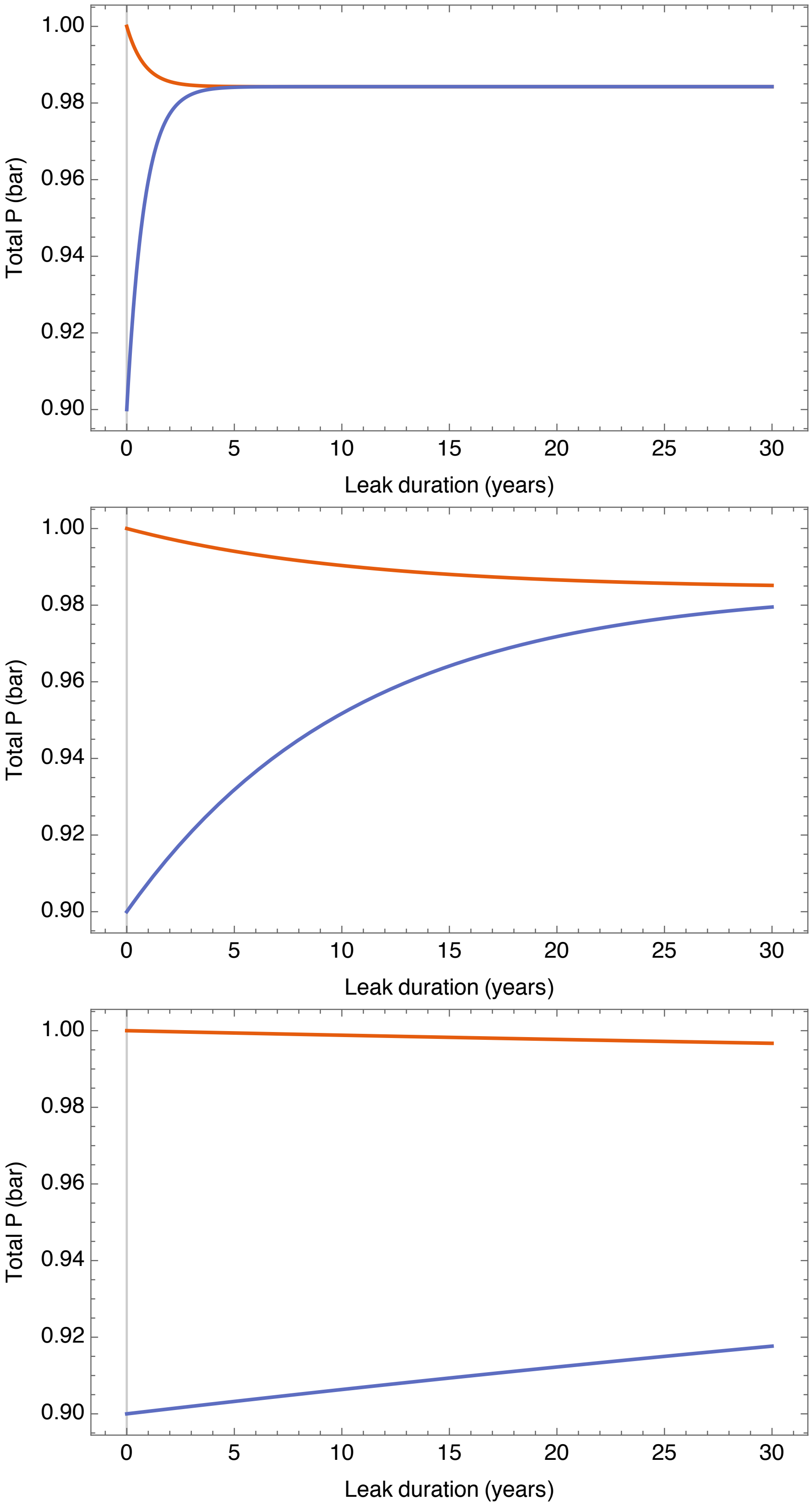

Figure 2: Predicted evolution of IC pressure (top/orange) and OI pressure (bottom/blue) as a function of time for ***air*** *leakage through cylindrical holes with diameters of 3.0 microns (top), 1.5*

microns (middle), and 0.74 microns (bottom). The initial conditions are set at 1.0 bar IC pressure and 0.9 bar OI pressure.

From the top graphs in Figures 1 and 2 we see that a leak that would exhibit a standard leak rate of 10^{-5} std cc/sec would allow any credible initial IC/OI pressure difference to equalize in less than five years of leakage, so a detectable pressure difference measured at DE after 5 years is a reasonably good indicator of that such a leak had not formed and persisted over most of the storage period. The bottom graphs in Figs. 1 and 2, in contrast, show that a 10^{-7} std cc/sec leak rate gives only small dissipation of the pressure difference even after 30 years. Such a leak could not be ruled out by measuring a pressure difference at DE unless one had very good information about the initial pressure difference at packaging and confidence that no significant gas generation or consumption occurred during storage. Practically speaking, these calculations suggest that a standard helium leak check at the 10^{-6} std cc/sec sensitivity level would probably be as good or better than using the DE pressure difference to detect a leak in the IC.

Predicted DE gas composition manifestations of leakage

About 30-35% mole % N_2 is typically found at DE in IC gas samples from Hanford-packaged containers in the pressure and corrosion surveillance bin. This has been inferred to result from incorporation of about 40% air in the IC at packaging, so N_2 is assumed to be present in the IC throughout the storage period. The OI gas samples taken from these containers at DE usually show about 2 kPa of N_2, which is presumed to have been incorporated during packaging and sealing. This background N_2 in the OI limits the sensitivity of using the N_2 concentration in the OI at DE to detect a small IC leak. For this discussion, we will assume that to be detectable by measuring N2 in the OI, a leak would have to result in at least a 2 kPa increase in N_2, as anything smaller would be indistinguishable from the background.

Entrainment of N_2 gas in pressure-driven continuum flow from IC to OI

Consider the effect of continuum flow driven by a difference in total pressure across a leak. If we assume 30 kPa N_2 in the IC initially and 100 and 90 kPa initial IC and OI total pressures, about 10% of the gas in the much smaller OI volume will have come from the IC if the pressure equalizes through a leak. Therefore the N_2 partial pressure in OI will reach about 10% of that in the IC, or 3 kPa. The time scale of this increase in N_2 will depend on leak size in the same way as the pressure difference equilibration times shown in Figs. 1 and 2. The OI would reach 2 kPa of N_2 after about 7 years of leakage through a leak measured at 10^{-6} std cc/sec, according to the middle graph of Fig. 1.

Diffusion of N_2 gas from inner to outer compared with DE gas analysis results

For comparison, we also calculate the expected change in N_2 partial pressure in the OI volume resulting from diffusion through several different leak diameters. The results of the calculations are shown in Figure 6 for diffusion through a 1.5-micron diameter hole, where the results show it would take about 6 years for the N_2 level to increase by 2 kPa. In other words, measuring N_2 concentrations at DE would be about as sensitive at detecting a leak that had persisted for 6 years as a He leak check with a detection threshold of 1 x 10^{-6} std cc/sec. Gas analysis performance improves in proportion to the duration of the leak, but it is not likely outperform an achievable standard leak test sensitivity. For example, a 0.74-micron diameter leak would have to persist for over 20 years for gas analysis to show a 2 kPa increase in the N_2 partial pressure in the OI, but this leak should be detectable with a helium leak test having a sensitivity of 1x10^{-7} std cc/sec.

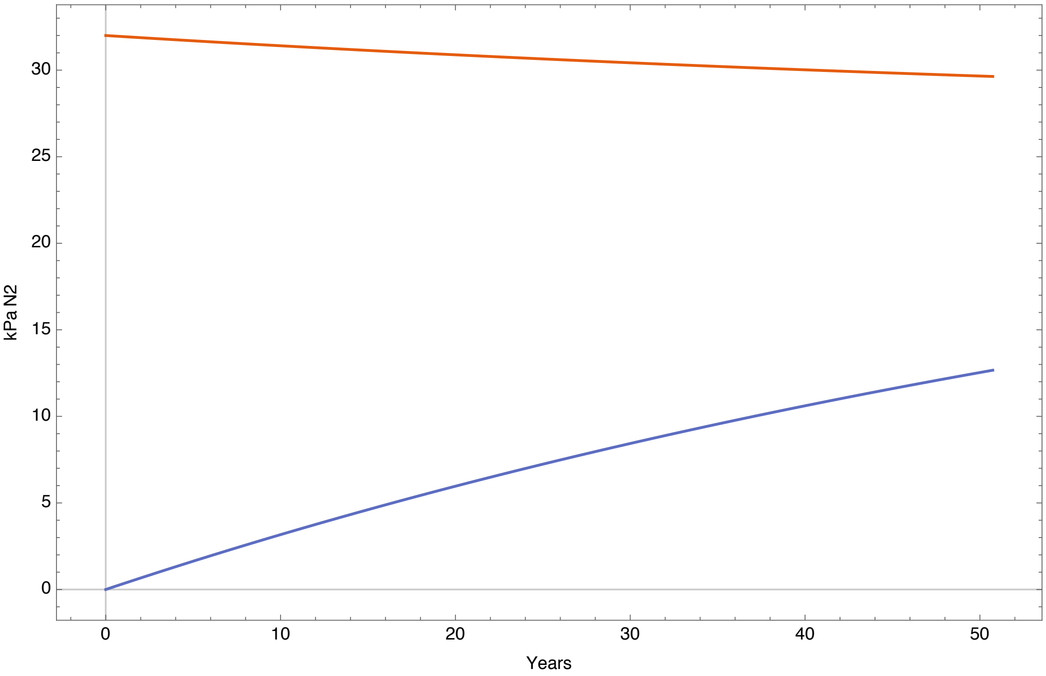

Figure 3: Predicted evolution of upstream N_2 partial pressure (top/orange) in the IC and downstream N_2 partial pressure (bottom/blue) in the OI as a function of time (seconds) for N_2 diffusion through a 1.5 micron diameter cylindrical leak. The IC (1.5 liter free volume) is assumed to contain in 100 kPa total pressure made up of 32 kPa of N_2 and the balance He at the time of sealing. The OI (0.28 liter free volume) is assumed to contain only 100 kPa helium initially.

Possible detection criteria based on transport of corrosion-enabling gases through a leak

Water vapor entrainment in pressure-driven leak flow

Consider water transport by entrainment in the gas flow driven a total pressure difference, with no fractionation of the gas mixture during leakage. Assume 100 kPa initial total pressure in the IC and 90 kPa in the OI. Further assume a constant 50% RH at 25 °C in the IC, corresponding to an H_2O partial pressure of 1.6 kPa, and no initial water vapor in the OI. If the gas pressure were to equalize completely by continuum flow through a leak, approximately 10% of the final gas in OI would consist of gas that had flowed through the leak from the IC. In that case the H2O partial pressure would be 10% of its partial pressure in the IC, or 0.16 kPa, giving 5% RH in the OI. Therefore, pressure-drive water vapor transport is not regarded as a critical factor in setting an acceptable leak size or leak test sensitivity criterion.

Diffusion of H_2O

Consider diffusion of H_2O through a cylindrical leak. Assume initial conditions of air in the IC at 100 kPa total pressure that includes 1.5 kPa of water vapor (~50% RH at 25 °C). Assume that the water vapor in the IC is in equilibrium with a condensed phase that maintains this 1.5 kPa partial pressure regardless of loss through the leak. Assume 100 kPa total pressure of air and zero H_2O partial pressure initially in the OI space.

We use the tabulated H_2O-He diffusion constant in Eq. 6 to model the diffusion of water vapor. This ignores the complication that the actual gas in the IC is only about 65% helium and contains over 30% nitrogen. Use of the H_2O-He diffusion constant will result in an overestimation of the water vapor diffusion rate in the actual system because the H_2O-N_2 diffusion constant is smaller. This is an acceptable result for this exercise because it skews the results toward conservative, i.e. shorter time, estimates of how quickly problematic quantities could move to the OI through holes of various sizes.

Figure 4 shows the predicted evolution of H_2O partial pressure as a function of time due to diffusion though a 3-micron diameter cylindrical leak. The diffusion calculation gives the initial rate of increase of the H_2O partial pressure in the OI space as 2.3×10^{-7} kPa/s, and shows that it would take about 14 years for the H_2O partial pressure in the OI space to reach half that in the IC, or about 25% RH. In short, a leak test at DE with a sensitivity of 1×10^{-5} std cc/sec would be able to detect a leak large enough to pass enough water vapor to raise the RH in the OI to 25% over 14 years.

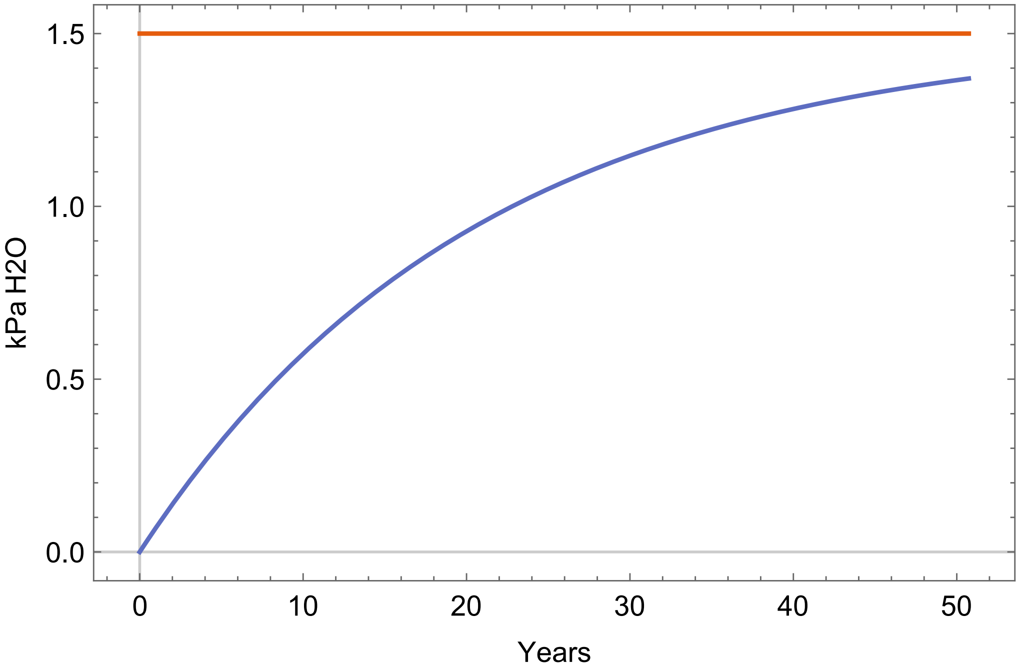

Figure 4: Predicted evolution of upstream H_2O pressure (top/orange) in kPa and downstream H_2O pressure (bottom/blue) as a function of time (seconds) for H_2O diffusion through a 3.0 micron diameter x 1.5 mm long hole from a 1.5 liter volume (the IC) at a steady p_{H2O} = 1.5 kPa in He at 100 kPa total pressure into a 0.28 liter volume (the OI) starting at 100 kPa He and 0.00 kPa H_2O. The standard helium leak rate measurement predicted for such a leak is 1×10^{-5} std cm^3/sec.

Figure 5 shows the predicted evolution of H_2O partial pressure by diffusion though a 1.5-micron diameter cylindrical leak, which would be measured to have a standard leak rate of 1×10^{-6} std cc/sec. It would take at least 57 years for to diffuse enough H_2O into the OI space to reach 25% RH.

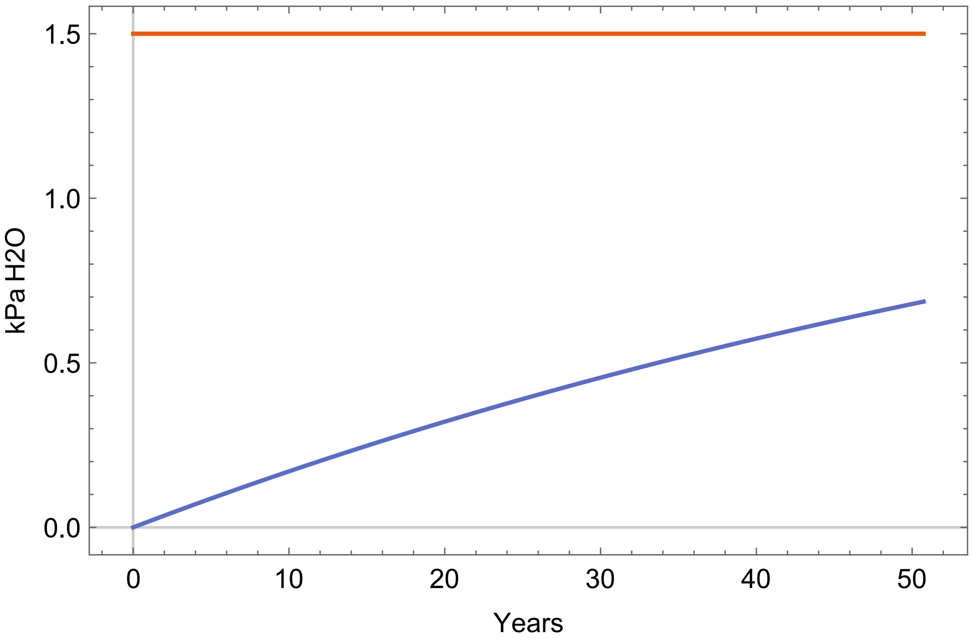

Figure 5: Predicted evolution of upstream H_2O pressure (top/orange) in kPa and downstream H_2O pressure (bottom/blue) as a function of time (seconds) for H_2O diffusion through a 1.5 micron diameter x 1.5 mm long hole from a 1.5 liter volume (the IC) at a steady p_{H2O} = 1.5 kPa in He at 100 kPa total pressure into a 0.28 liter volume (the OI) starting at 100 kPa He and 0.00 kPa H_2O. The standard helium leak rate measurement predicted for such a leak is $1x10^{-6}$ std cm^3/sec.

HCl entrainment in pressure-driven leak flow

Consider HCl transport through a leak from an IC having a hypothetical steady HCl partial pressure of 0.02 kPa. This is the HCl partial pressure at equilibrium over a 6 M HCl aqueous solution, which has been shown to cause corrosion of 304L stainless steel samples. Assume the OI initially contains no HCl. As with the water vapor example above, assume 100 kPa IC initial total pressure and 90 kPa OI total pressure. The final OI partial pressure of HCl after equalization of an initial 10 kPa total pressure difference is 10% of 0.02 – 0.00 kPa = 0.002 kPa. Given the OI volume of 0.28 liters and applying the ideal gas law at 298 K, there will be 0.23 micromoles of HCl transported through the leak to the OI after the 10 kPa pressure rise is complete. This is less than 25% of the 1.1 micromoles of chloride found in citric acid washes of just the ICCWR portion of one DE. Given that deposition on the outer is likely to be somewhat dispersed to other areas in addition to the weld, we conclude that pressure equalization transport of HCl would not move enough chlorine to the outer container to cause much corrosion. Looking at it another way, the HCl partial pressure in the OI resulting from pressure equilibration will always be much lower than its partial pressure in the IC, no matter how large the leak. For leak flow driven only by a total pressure difference, the corrosion impact on the outer container will always be less than the corrosion impact on the inner container.

It is clear from the above discussion that leakage flow of gases driven by small initial pressure differences cannot not drive the gas environment in the outer container to approach HCl concentrations as corrosive as those within the inner container. The pressure difference would go to zero before enough corrosive gas could be transported. However, diffusion could move more of these minor gases because it is driven by partial pressure gradients rather than total pressure gradients, and partial pressure gradients would persist in the presence of a leak.

Summary

We have used formulae in ANSI N14.5 to determine dimensions for three hypothetical inner container leaks that would exhibit standard helium leak rates of $1x10^{-5}$, $1x10^{-6}$, and $1x10^{-7}$ std cc/sec. For these three leaks, we have calculated gas pressure and composition changes predicted during storage via both continuum flow driven by a pressure difference and via diffusion. This enables comparison of leak detection ability of DE gas analysis with that achievable by standard helium leak tests at various levels of sensitivity. Finally, we have calculated the expected transport of potentially corrosive gases to the OI if a leak were to occur.

Table 1 summarizes the results of the calculations in this paper for quick comparison between helium leak test detectability and leak manifestations in DE pressure and gas composition measurements for our three hypothetical reference leak sizes. The half-life of the pressure difference gives a rough indication of how long a leak of each size would have to persist to be detectable using DE pressure difference. The next two columns give an idea of the leak durations needed to show measurable increase in the N_2 partial pressure in the OI, which is the most consistently useful indicator of leakage in the DE gas composition analysis [Duffey 2014]. The final column is intended to give an idea of the leakage time needed to transport a minimum amount of corrosion enabling gas to the OI for the reference leak diameters.

The results show that for the minimum leak detectable at a standard test sensitivity of $1x10^{-7}$ std cc/sec, there would be no conclusive manifestations in the pressure difference or gas composition at DE unless the leak persisted for several decades. For leaks persisting for five to ten years, DE measurements would have comparable detection ability to standard helium leak tests at the $1x10^{-6}$ std cc/sec sensitivity level. Neither of these sizes of leaks would allow sufficient water vapor to diffuse into the OI to raise its RH to a credibly corrosive level during the projected storage time of 50 years or less, even assuming no depletion mechanisms for the water vapor in the system over that period.

Table 1: Summary of calculated gas transport manifestations of three hypothetical cylindrical leaks through the IC wall (1.5 mm).

Standard helium leak rate (atm cc/sec)	**$1x10^{-5}$**	**$1x10^{-6}$**	**$1x10^{-7}$**
Diameter of idealized cylindrical leak path (microns)	3	1.5	0.74
Half-life of an initial 10 kPa IC/OI pressure difference (years) [i]	0.42	4.5	46
Time for 2 kPa of N_2 to diffuse into OI (years) [ii]	1.5	6.1	25
Half-life for N_2 partial pressure in OI approach to IC value (years) [iii]	16	66	270
Half-life for RH in OI approach to that in IC by diffusion in He atmosphere[iv]	14	58	240

---

[i] Initial conditions for pressure leak-up simulation are: IC contains 100 kPa helium, OI contains 90 kPa helium.

[ii] Initial conditions at container closure assumed to be OI containing 100 kPa helium and IC container 68 kPa helium and 32 kPa nitrogen at 25 °C. Time is from appearance of leak, assumed to be a step function from no leak to the full leak size.

[iii] Same conditions as note ii.

[iv] IC assumed to be maintained at a steady state H_2O partial pressure of 1.5 kPa, which is about 50% RH at 25 °C. IC has initial H_2O partial pressure of 0.0 kPa.

References

ANSI (2014). American National Standard for Radioactive Materials - Leakage Tests on Packages for Shipment. New York, NY, USA, American National Standards Institute. **ANSI N14.5-2014**.

E. L. Cussler (2009). Diffusion. Cambridge, UK, Cambridge University Press.

J. M. Duffey (2014). "A Comparison of Gas Composition and Pressure of Inner and Outer 3013 DE Containers." SRNL-STI-2014-00439.