A Review of Information on Flow Equations for the Assessment of Leaks in Radioactive Transport Containers

J. Higson[1], C. Vallepin[2], H. Kowalewsky[3]

[1]*UK Atomic Energy Authority, RTS, Risley, Warrington, United Kingdom*
[2]*CEA, Saclay, Paris, France*
[3]*Bundersanstalt für Materialforschung und -prüfung (BAM), Berlin, Federal Republic of Germany*

Introduction

The International Atomic Energy Agency (IAEA) Regulations establish standards of safety which provide an acceptable level of control of the radiation hazards to persons, property and the environment that are associated with the transport of radioactive material.

The design of packages may be demonstrated as meeting the relevant requirements of the Regulations by subjecting prototypes or samples of a particular design to physical and mechanical environmental tests.

For Type B, designs have to meet the requirements for permitted activity release as shown in Table 1.

Conditions	Type B
After tests for normal conditions	$A_2 \times 10^{-6}$/hour
After tests for accident conditions	A_2 in one week, or $A_2 \times 10$ in one week for krypton 85

NOTE: A_2 values in TBq for radionuclides are given in the Regulations.

Table 1 Permitted Activity Release

Following application of these tests it is necessary to demonstrate that the leaktightness of the containment is maintained to the required degree. This demonstration requires some form of contents leakage test.

To specify activity release in fluid leakage rate terms, a relationship between the activity release and fluid leakage is required. The physical states of the activity must be established ie particulate, liquid, or gaseous. For liquids and gases, leakage calculations can be carried out using established fluid flow formulae. For particulate material, no practical calculation method exists except by assuming that the solid matter behaves as an aerosol in a gaseous environment, or as a homogenous liquid. These assumptions give overestimates of particulate release. If data on particulate size is known, this could be used to determine minimum leakage path size and therefore leakage rate. A paper to this PATRAM Conference (Higson et al) examines this approach.

If we consider a typical Type B package, it would consist of a container with a gasketted lid. A frequently used seal system is shown in Figure 1. A typical leak across the gasketted seal could be in the form of a scratch or a trapped "hair" which would give a leakage path with a relatively long length to diameter ratio (ie a fine capillary). Typical lengths would be of the order of 10 mm.

A further possibility for a leakage path could occur at low temperatures where an elastomeric seal would lose its elasticity and due to the differential expansion rates between the elastomer and the container body, a leakage path in the form of a long narrow gap would develop.

Therefore, this paper considers the leakage through capillaries and gaps. Assuming the shape of the leak geometry it is possible to compare the performance of packages in normal operation and at test conditions. National Codes of Test Practice (ANSI, UKAEA) have been produced to assist operators to meet the requirements of the Regulations. There is a need to understand the flow model and leakage path geometry involved before an agreed interpretation of the leakage rates can be made.

Liquid Flows

For calculating leakage of liquids, Poiseuille's equation for viscous laminar flow through small passages such as capillaries or gaps (Stelzer 1971) is used. Figure 2 shows the application of Poiseuille's equation. In practice, the actual liquid flow rate is likely to be less than calculated from Poiseuille's equation due to the effect of surface tension σ creating a back pressure $P\sigma$.

Water Surface Tension	Capillary $d=1\mu m$ $\ell=1cm$	Gap $h = 1\mu m$ $\ell=w=1cm$
$\sigma(20°C)=7.3 \times 10^{-5}$ bar cm $\sigma(100°C)= 5.9 \times 10^{-5}$ bar cm	$P\sigma$ = 2.9 bar $P\sigma$ = 2.35 bar	$P\sigma$ = 1.45 bar $P\sigma$ = 1.20 bar
Standard Air Flow Rate	$L \approx 10^{-8}$ atm cm^3 sec^{-1}	$\approx 10^{-3}$atm cm^{3}sec^{-1}

Table 2 Examples of back pressure

In reality the pore radii or gap/crack width may have a wide size distribution which could result in no liquid leakage even though gas leakage can be detected.

Gas Flows

In order to calculate leak geometry from measured leakage rates of gas the correct flow regime must be considered. Different basic laws relate leakage rates to pressure differential across the leak, the range of absolute pressure involved, and the nature of the gas passing through the leak.

The various flow regimes are best shown in a figure produced by Santeler (1986) which indicates the flow regimes for both tubes (capillaries) and orifices. The well known semi-empirical Knudsen equation for capillaries covers the whole range from molecular, transition, to viscous laminar flow. There are well established formulae for gas flow rates through circular capillaries as well as through rectangular gaps for both pure molecular flow and pure viscous laminar flow. Blanc et al (1982) have combined these formulae for gaps analogous to the Knudsen equation.

Capillary

$$L = \frac{\pi d^4}{128\,\ell\eta}\,\bar{p}\,\Delta p + \frac{1}{6}\cdot\sqrt{\frac{2\pi RT}{M}}\cdot\frac{d^3}{\ell}\cdot\frac{1+\sqrt{\frac{M}{RT}}\cdot\frac{1}{\eta}\cdot d.\,\bar{p}}{1+1.24\sqrt{\frac{M}{RT}}\cdot\frac{1}{\eta}\cdot d\cdot\bar{p}}\cdot\Delta p$$

which reduces to

$$L\ (\text{air } 25°C) = 134\,\frac{d^4}{\ell}.\ \bar{p}.\ \Delta p + 12.2\,\frac{d^3}{\ell}.\ \frac{1+186\,d\,\bar{p}}{1+231\,d\,\bar{p}}\ .\ \Delta p$$

$$L\ (\text{He } 20°C) = 126\,\frac{d^4}{\ell}.\ \bar{p}\ .\ \Delta p + 32.6\,\frac{d^3}{\ell}.\ \frac{1+65.7\,d\,\bar{p}}{1+81.5\,d\,\bar{p}}\ .\ \Delta p$$

Gaps

$$L = \frac{1}{12\eta}\cdot\frac{h^3 w}{\ell}\,\bar{p}\cdot\Delta p + \sqrt{\frac{RT}{2\pi M}}\cdot\frac{h^2 w}{\ell}\cdot\ln\left(\frac{\ell}{h}\right)\frac{1+\sqrt{\frac{M}{RT}}\,\frac{1}{\eta}\,h\,\bar{p}}{1+1.24\sqrt{\frac{M}{RT}}\,\frac{1}{\eta}\,h\,\bar{p}}\,\Delta p$$

which reduces to

$$L\ (\text{air } 25°\text{C}) = 454\ \frac{h^3 w}{\ell}\ \bar{p}\ \Delta p + 11.7\ \frac{h^2 w}{\ell}\ \ell n \left(\frac{\ell}{h}\right) \frac{1 + 186\ h\ \bar{p}}{1 + 231\ h\ \bar{p}}\ \Delta p$$

$$L\ (\text{He } 20°\text{C}) = 427\ \frac{h^3 w}{\ell}\ \bar{p}\ \Delta p + 31.1\ \frac{h^2 w}{\ell}\ \ell n \left(\frac{\ell}{h}\right) \frac{1 + 65.7\ h\ \bar{p}}{1 + 81.5\ h\ \bar{p}}\ \Delta p$$

d, ℓ, h, w in cm, p in mbar, L in mbar litre $\text{sec}^{-1} \approx \text{atm cm}^3\ \text{sec}^{-1}$

At larger flow rates two effects may occur.

a. With high Reynolds Numbers turbulent flow occurs, but this regime is in practice outside the range of flows we are considering.

b. Gas flow velocity increases and reaches a maximum at the speed of sound. By increasing the upstream pressure with the intention of enhancing the flow rate, the downstream flow velocity will be limited. The effective downstream pressure becomes higher than the gas pressure outside the exit. The pressure differential along the capillary or gap increases slower and slower until only the upstream pressure governs the flow, at which pure orifice choked flow occurs (ASNT 1982; ANSI 1987)

$$L = A\ Pu \left(\frac{2}{k + 1}\right)^{\frac{1}{k - 1}} \sqrt{\frac{2k}{k + 1}\ \frac{RT}{M}}$$

which reduces to

$$L\ (\text{air } 25°\text{C}) = 20.04\ A\ pu = 20.04\ hw\ pu = 15.74\ d^2\ pu$$

$$L\ (\text{He } 20°\text{C}) = 56.6\ A\ pu = 56.6\ hw\ pu = 44.45\ d^2\ pu$$

(using same units as above)

The American National Standards Institute Standard N14.5 (ANSI 1987) "Leakage Tests on Packages for Radioactive Materials" indicates the range of leakage rates for which the flow regimes require consideration. It states that leakage rates up to 10^{-7} atm $\text{cm}^3\ \text{sec}^{-1}$ are considered to represent leaktightness whereas a leakage rate greater than 10 atm $\text{cm}^3\ \text{sec}^{-1}$ is large and does not require testing. Figure 4 indicates the relationship of leakage rate to capillary diameter and gap height between these two levels.

Gas Flow Modes Used by ANSI N14.5

Appendix A of ANSI standard N14.5 uses the equations above relating to a straight circular capillary 1 cm in length modelling a typical leakage path. If the factor

$$\frac{1 + C \bar{p} d}{1 + 1.24 C p d}$$

within the Knudsen equation is slightly rounded up to 1, the Knudsen equation then reduces to

$$L \text{ (air } 25°C) = 1.35 \times 10^{8} \frac{d^{4}}{\ell} \frac{(Pu^{2} - Pd^{2})}{2} + 1.22 \; 10^{4} \frac{d^{3}}{\ell} (Pu - Pd)$$

$$= Lc + Lm$$

d, ℓ in cm, p in atm, L in atm $cm^3 \, sec^{-1}$

Choked flow should occur if

a. $Lc \geq Lm$

b. $\frac{Pd}{Pu} \geq \left(\frac{2}{k + 1}\right)^{\frac{k}{k-1}}$

These two conditions are not strictly accurate, as (a) is an unfounded, arbitrary condition and (b) is a condition valid only for orifices. The physical background becomes evident if the calculated values in Table B2 of N14.5 are shown as a graph (see Figure 5). There is no technical reason why the air flow rate should suddenly increase by a factor of approximately 3600. In our opinion for flow rates from 10^{-7} to 10^{1} atm $cm^3 \, sec^{-1}$ the use of the Knudsen equation for capillaries and slightly modified for gaps can be used without reservation.

Measurements at BAM

For about ten years BAM have been investigating the permeation of gases through elastomeric sealing materials using a quadrupole mass spectrometer (QMS range 1 to 200 atomic mass units) and helium mass spectrometer leak detectors. The calibration of the instruments being based upon the validity of Knudsen's equation for capillaries. The flow of helium at various pressures was measured through selected capillaries and the best fit of length and diameter was found using Knudsen's equation. Using flow of different gases through the capillary allowed a check on the size of the capillary and eventually the calibration and sensitivities of the spectrometers. Reference leaks consisting of compressed sintered stainless steel powder providing pure molecular flow were also used to calibrate the spectrometers. Both types of leaks led to fully compatible results.

Figure 6 shows some of the experimental results, none of which exhibits the sudden large jump in flow predicted in ANS N14.5.

Application with Elastomeric Seals at Low Temperature

All elastomers eventually lose their elastic properties as their temperature is lowered. Considering a VITON O-ring with a 10 mm diameter compressed by 20% between steel flanges, below a critical temperature of eg Tc - 30°C this elastomer becomes stiff and inelastic. If the temperature is then lowered even further to -40°C because of the differing thermal expansions between steel and the now inelastic VITON, a gap will appear between the steel plates and the VITON O-ring.

Material	Linear Thermal Expansion Coefficient ($x\ 10^{-4}$)
VITON at T > TG	1.3 - 1.9
VITON at T < TG	0.4 - 0.5
Stainless steel	0.16

Therefore gap height $h = 1 \times 0.8 \times (0.5 - 0.16) \times 10^{-4} \times 10 \approx 3\ \mu m$

From the measured leakage rates, gap heights could be calculated which at -40°C were of the order of 1 μm.

Measurements of glass transition temperatures TG for some VITON compounds by thermomechanical analysis and of the critical temperature Tc where a gap suddenly appears and leakage occurs have shown (Weise et al 1989) Tc between - 45°C and -30°C and TG to be about 20°C higher.

This effect depends upon the rate of temperature change. Recent work in the USSR (Shtitel'man et al 1988) using vibrations with frequencies between 25 and 0.025 Hz on elastomeric seals at low temperatures found that the temperature of seal failure decreased with lower frequencies. Waiting long enough these gaps might contract and eventually close.

This deterioration in elasticity induced by low temperatures is a physical and reversible effect, the elastomer regaining its original properties when warmed to ambient temperatures.

Helium Leak Testing of Welded Highly Active Waste Canisters

In order to demonstrate the safe disposal of vitrified radioactive liquid waste in a salt mine, the German Ministry of Research and Technology has initiated a test programme managed by Gesellschaft für Strahlen und Umweltforschung (GSF). In this programme 30 steel canisters filled with borosilicate glass each containing up to 5.3 PBq Sr - 90 and/or 7.7 PBq Cs - 137 have been fabricated at the Battelle Pacific Northwest Laboratories. The canister height is 1.20m, diameter 0.30m and weighing 250 kg (Stippler et al, 1989).

To cover transport, handling in the Asse Salt Mine, and retrieval after five years, these canisters are to be licenced as sealed sources. To guarantee leaktightness, a quality assured welding process and an ingenious helium leak test procedure have been developed and successfully applied. Immediately prior to welding the canister lid in position, a capsule with a capillary outlet was pressurised with helium up to 60 atmospheres pressure and placed in the canister. [The fill pressure of the capsule was such that at temperatures up to 300°C the pressure inside the closed canister could not exceed 5.6 atm.]

Helium releases slowly through the capillary and its relation to time is shown in Figure 7. All helium which was released from the capsule during welding was considered lost. The time from emplacement of the capsule to completion of welding usually took one hour but on one occasion took three hours.

The canisters were placed, after welding and further helium release during about a day, in a large vacuum chamber connected to a helium mass spectrometer.

ANS N14.5 considers standard air leakage rates up to 10^{-7} atm cm^3 sec^{-1} as leaktight, this limit being equivalent to a standard helium leakage rate of 2.7×10^{-7} atm cm^3 sec^{-1} for pure molecular flow. By German authorities a standard helium leakage rate of 10^{-7} mbar litre sec^{-1} was set as an upper limit. The average measured helium leakage rate was 2.4×10^{-9}atm cm^3 sec^{-1} and it could be demonstrated that the standard helium leakage rates of all 30 canisters were well below both limits.

Applicable Codes and Standards

The most widely used standard is the American National Standards Institute Standard N14.5 - 1987. This is used as a guide not only in the US but in many countries throughout the world. The United Kingdom Atomic Energy Authority has produced a Code of Practice AECP 1068 - 1988 which has a similar use.

To use these standards requires a knowledge of the leak geometry and flow regime. To assist users, computer codes have been produced nationally but have not been used universally. Examples of these are the CAPIL code in the US (Owczarski et al 1979) and LUSEC in France (CEA).

The International Standard Organisation (ISO) has asked a working group (Tanguy 1989) to produce an International Standard to assist Radioactive Transport Users to successfully leaktest packages.

Conclusion

A large number of papers have been written to assist Radioactive Transport Users to calculate leakage rates through capillaries and orifices. To calculate the flow of a fluid through a hole or gap the flow regime and leak geometry should be known.

The American National Standard Institute Standard N14.5 is a useful starting point as this indicates the range of leakage rates of interest (ie 10^{-7} atm cm^3 sec^{-1} to 10 atm cm^3 sec^{-1}). Within this range it has been shown that for gases the use of the Knudsen Transition Formula for capillaries and with slight amendment for long narrow gaps provides the best correlation.

Examples of the use of this equation with some experimental work indicates the adequacy of this approach.

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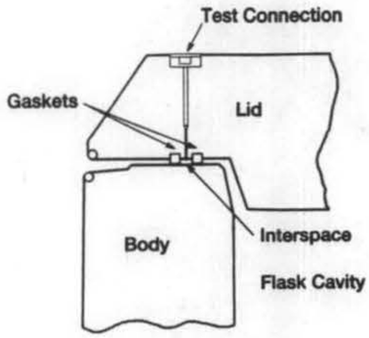
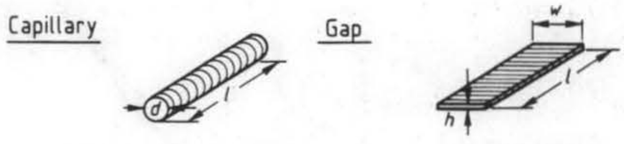


Figure 1. Typical O-ring seal flange joint



$$L = \frac{dV}{dt} = \frac{\pi}{128} \cdot \frac{d^4}{l} \; \frac{(p_u - p_d)}{\eta} \qquad L = \frac{dV}{dt} = \frac{1}{12} \cdot \frac{h^3 w}{l} \; \frac{(p_u - p_d)}{\eta}$$

$$p_a = \frac{4\sigma}{d} = \frac{2\sigma}{r} \qquad p_a = \frac{2\sigma}{h}$$

L = volumetric flow rate
$\frac{dV}{dt}$ = volume of liquid passing per unit time
p_u = upstream pressure
p_d = downstream pressure
η = dynamic viscosity
σ = surface tension
p_σ = blockage pressure
d = diameter = 2r
l = length
h = height
w = width

Figure 2. Volumetric flow rates of liquids (Poiseuille's equation) (Poiseuille's equation)

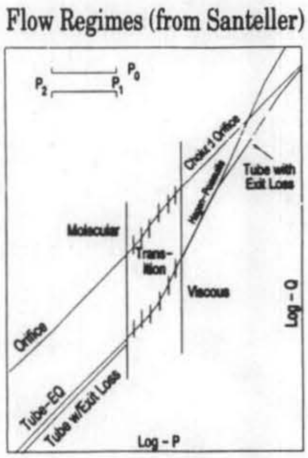


Figure 3. Flow region according to Santeler

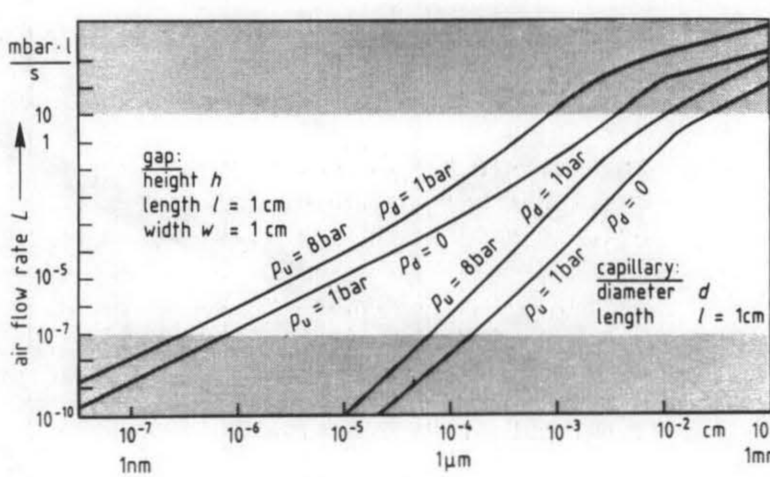


Figure 4. Representative gas flow rates throughout capillaries (Knudsen's equation) and gaps

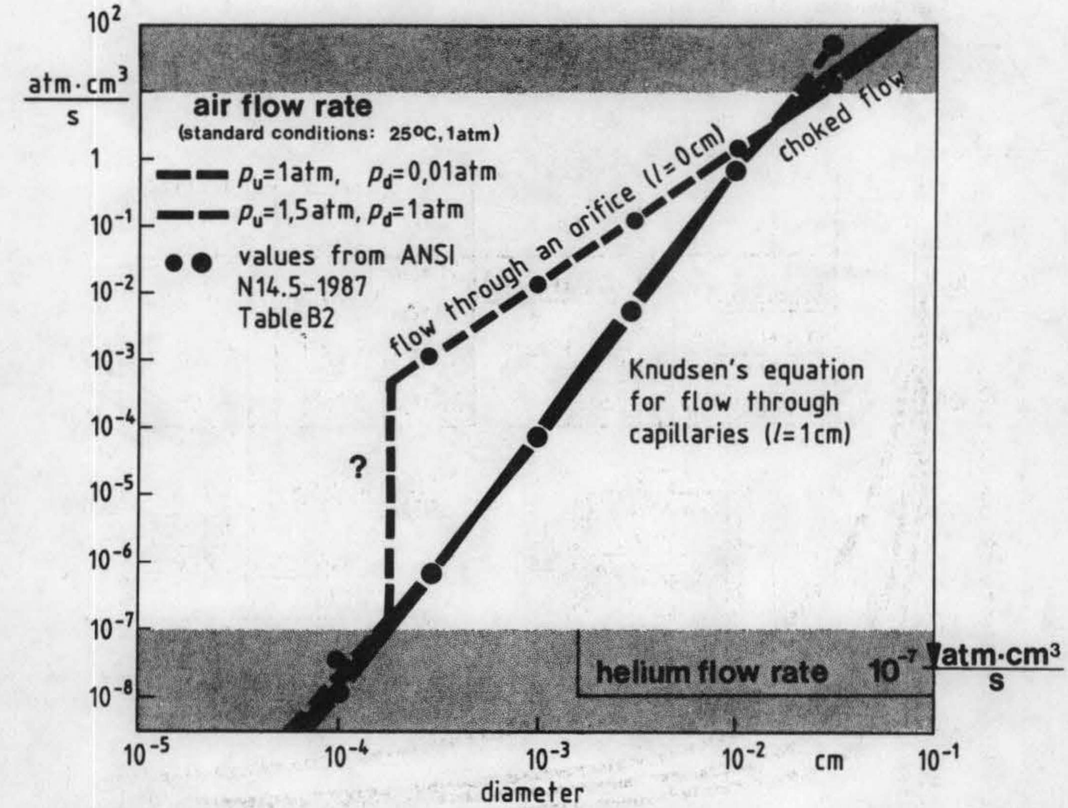


Figure 5. Air flow rates according to Appendix 3 of ANSI N14.5 - 1987 of ANSI N14.5 - 1987

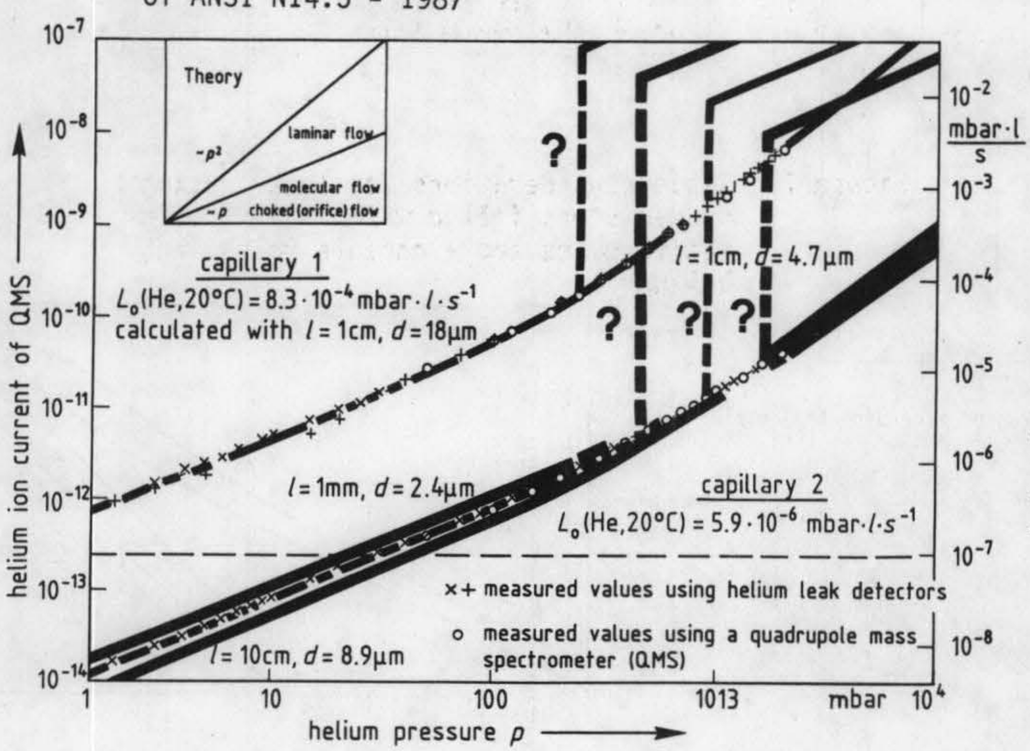


Figure 6. Measured helium flow rates through capillaries

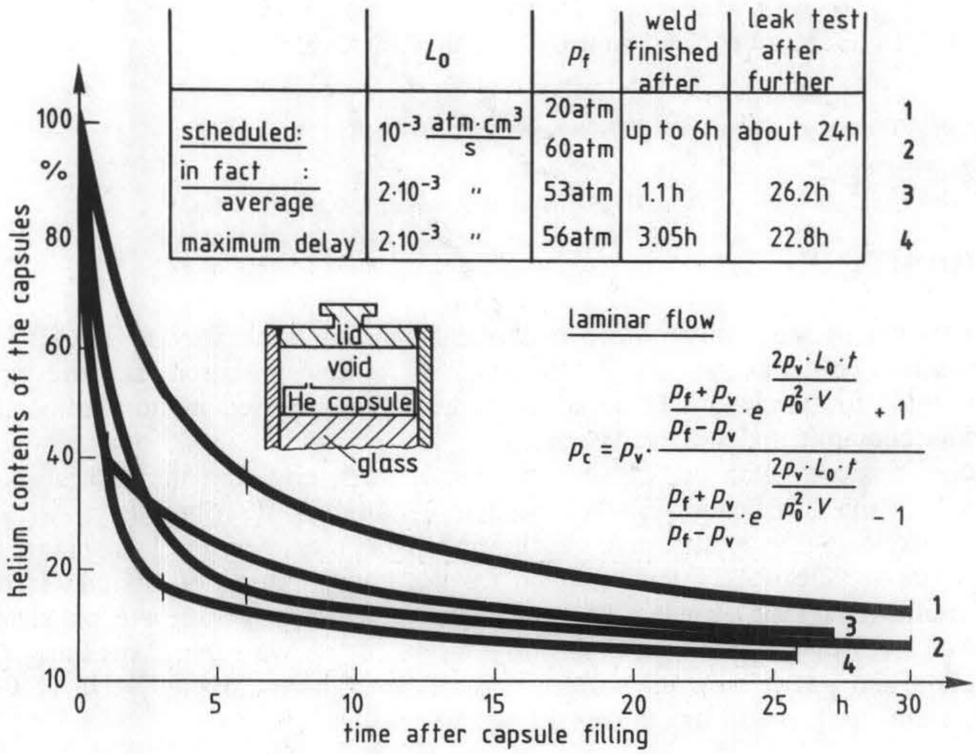


Figure 7. Basic considerations for leak testing of canisters filled with highly radioactive glass and a capsule releasing helium