Modeling Contents and Leaks—Advantages and Limitations

H. Kowalewsky
Bundesanstalt für Materialforschung und -prüfung

INTRODUCTION

During 11 meetings between Sept. 1987 and Sept. 1992 the Draft International Standard ISO/DIS 12807 "Leakage testing on packages for the safe transport of radioactive materials" has been developed and was published Nov. 1993. In July 1994 ISO/TC 85 "Nuclear Energy"/SC 5 "Nuclear Fuel Technology"/WG 10 "Leakage Testing ..." under its chairman (Louis Tanguy, France) met again to resolve last questions and comments. End of March 1995 the final version (draft P) was submitted to the ISO Central Secretariat; ISO 12807 shall be issued May 1996. This International Standard should supersede the American National Standard N14.5 for "Leakage testing on packages for shipment of radioactive materials" of 1977 (chairman William R. Taylor, Canada) and its revision of 1987 (chairman Larry Fischer, USA); both chairmen were active members of this new ISO working group. One negative tendency: ANSI N14.5-1977 included 5 "Calculation Examples", ANSI N.14.5-1987 34 (!!) "Example Problems", and ISO 12807 will include only 13 "Worked Examples"; among others a new one about permeation (helium during leakage testing, and radioactive krypton-85 release at normal transport and accident conditions) as a German contribution. Further you will find comprehensive "Explanatory notes" on gas flow prepared by UK and German delegates. Within all three standards the "one capillary" leak model with laminar-viscous gas flow predominates. For a later revision of ISO 12807 we need some further worked examples! Supplementary now it seems useful to explain some features used in German licensing practice to achieve a more general acceptance and application.

FLOW MODES

Leakage and permeation rates Q depend on the upstream pressure p_u and the downstream pressure p_d across a leak or a permeable wall according to the relationship $Q = f \cdot (p_u^x - p_d^y)^z$ where f depends on the leak geometry, the flowing medium, or - in case of permeation - on the geometry and permeability of the permeable material for the permeating gas. p is either the total pressure p_t or the partial pressure p_i as indicated by + in the table where also the numerical values of the exponents x, y and z are listed:

$Q = f \cdot (p_u^x - p_d^y)^z$		p_t	p_i	x	y	z
permeation of diatomic molecules through metals			+	0.5	0.5	1
permeation of any molecules or atoms through elastomers			+	1	1	1
leakage of gases, molecular flow*)	through		+	1	1	1
" " ", viscous flow*)	capillaries	+		2	2	1
" " ", turbulent flow	and	+		2	2	4/7
" " ", choked flow	gaps	+		1	-∞	1
leakage of liquids, viscous flow		+		1	1	1

*) both flow regimes described by the semiempirical Knudsen equation (Higson et al. 1989), which was devised in 1909 to represent experimental data.

Figure 1 shows the flow modes for air through single capillaries characterized by their Standardized Leakage Rates (SLR) between 10(-8) Pa · m³/s SLR (= 10 (-7) mbar · l/s or ≈ 10 (-7) std cm³/s) and 1 Pa · m³/s SLR. A Type B(U) package shall not have a maximum normal operating pressure (MNOP) in excess of a gauge pressure of 700 kPa (7 bar) indicated by a vertical dashed line. The air flow rates through capillaries A and B within the envelope (up to 7 bar) are covered by the Knudsen equation for molecular and viscous-laminar flow; the air flow rate through capillary C passes into the turbulent region and through capillary D even into the choked flow region, but in both cases the use of the Knudsen equation would overestimate the leakage rate at higher gauge pressures thus being conservative.

What means 10(-8)Pa · m³/s SLR? We have to consider and to imagine orders of magnitude:
3.1 std cm³ per year or 2.3 · 10(12) molecules of air per second.

Figure 2 shows the most important flow modes for our work: viscous-laminar flow for liquids according to the Hagen-Poiseuille equation and the similar viscous-laminar flow for gases according to the Knudsen equation. Because any gas is compressible, we have to add a factor consisting of the average pressure $\bar{p}$. It is an important fact that in these cases the composition of the flowing medium does not change, therefore the term "medium flow" is used equal to "pure viscous-laminar flow". The other part of the Knudsen equation - here to the left - describes molecular flow which generally consists of two opposite diffusion flows driven independently of each other by the different partial pressures. Looking at Figure 2, addition of both air flows results in an air flow into the cask, but we will have Kr-85 leakage out of the cask against a higher partial pressure. Similarly, permeation has to be considered.

Performing one leakage test, we cannot decide if one or several or even a great many leaks or capillaries contribute to the total leakage rate. **Figure 3** shows a variety of combinations related to 10 (-4) Pa · m³/s SLR. The leakage may be caused by one leak or by many very small leaks and would be limited at minimum by assuming pure molecular flow. The latter model covers also very thin gaps. An unknown number of leaks will provide leakage rates at higher pressures only between the curves for pure viscous-laminar flow and pure molecular flow. Being aware of these limitations, we can derive a pressure rise range within evacuated casks, see the following equations and **Figure 4** with an example: at 0.5 bar total pressure after loading and at temperature equilibrium 10 (-3) Pa · m³/s SLR and a cavity volume of 100 l, it will take at least 20 weeks until a total pressure of 950 mbar will be reached, that means that in any case cavity total pressure will be lower than the atmospheric pressure, but nevertheless we might have molecular leakage out of the cask. Narrow natural capillaries could be found in gasket materials made of asbestos fibers mixed with rubber compounds. In a German thesis (Bierl. 1978) the pressure dependence of leakage rates has been measured and can be described by the Knudsen equation applied to a very large number of similar small capillaries. Because of the narrow leak paths around the asbestos fibers with their diameters in the μm range - responsible for lung cancer risk after any inhalation like PuO_2 powder - these gaskets provided blockage against liquid penetration.
At PATRAM '89 we introduced the gap model (Higson et al. 1989), which in many cases is more appropriate than the capillary model: mechanical or thermal stresses may cause gaps but no capillaries. Reversible gaps may occur at elastomeric sealing systems at low temperatures.

Figure 5 shows the result of an experiment at BAM: If an O-ring made of low-temperature VITON is cooled down, it will adhere to the grooves of the sealing surface - this adhesion will be better the higher the previous temperature was. From a certain low temperature downwards there grows an ideal gap which can be closed continuously by heating up the sealing system. IAEA Safety Series No 6 requires for Type A packages (= packaging + radioactive contents) "to take into account temperatures ranging from -40°C to +70°C for the components of the packaging" and for Type B packages "to be designed for an ambient temperature range from -40°C to +38°C". We feel that VITON is a very useful sealing material as well for rough handling as for high-temperature scenarios. High-activity contents involves heating of the sealing system. If there is no heat, this involves a low specific activity, may be also freezing humidity, and therefore we can agree to a small gap. From a realistic view, high temperature occurrence e. g. caused by an accident is more likely than any low temperature conditions near -40°C, and any activity release will be higher and more dangerous for people at higher temperature. Testing cask materials at -40°C means looking for irreversible destruction effects, but any freezing of an elastomeric material is a reversible process. In any case we must have information about the contents; e. g. transport of radioactive gases will require another seal quality.

CONCLUSIONS

The Knudsen equation and the "one capillary" model assuming a leak to be a smooth and straight tube is conservative in most cases, looking for gaseous, liquid, and particle contents as well for demonstrating the predominating flow mechanism as for taking into account any blockage mechanisms. Only a little more conservative we can consider "one capillary" allowing only pure viscous-laminar flow thus validating the medium or continuum flow methodology preferred by ISO 12807. But this model could hide molecular leakage against a higher total pressure.

The gap model (Higson et al. 1989) sometimes is more realistic, has a strong tendency to molecular leakage effects allowing some gas release, but provides additional arguments for blockage effects in case of liquid or powder contents.

There is some progress to validate these simple models and calculations by experiments (in Japan, UK, USA) which will help us to understand these mechanisms and to convince the public of our earnest efforts to achieve the best technical standard to prevent inadmissible release of radioactivity.

Today a problem seems to be solved only if we come down from a high level of understanding to a convincing simplification.

REFERENCES

Bierl, A. *Untersuchung der Leckraten von Dichtungen in Flanschverbindungen*, Thesis, University of Bochum, Germany (1978)

Higson, J., Kowalewsky, H., Vallepin, C. *A Review of Information on Flow Equations for the Assessment of Leaks in Radioactive Transport Containers*, PATRAM 1989, Washington, USA

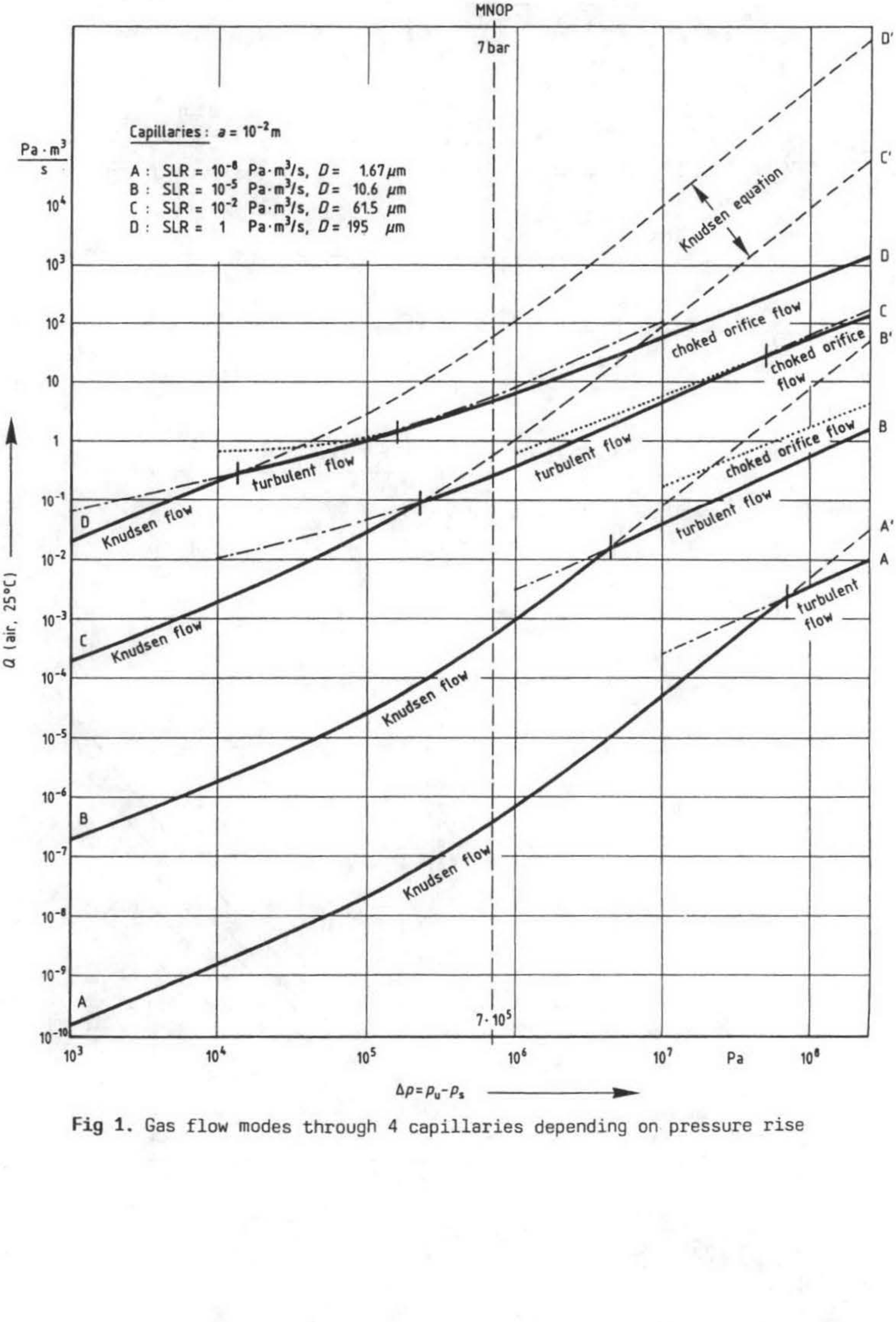


Fig 1. Gas flow modes through 4 capillaries depending on pressure rise

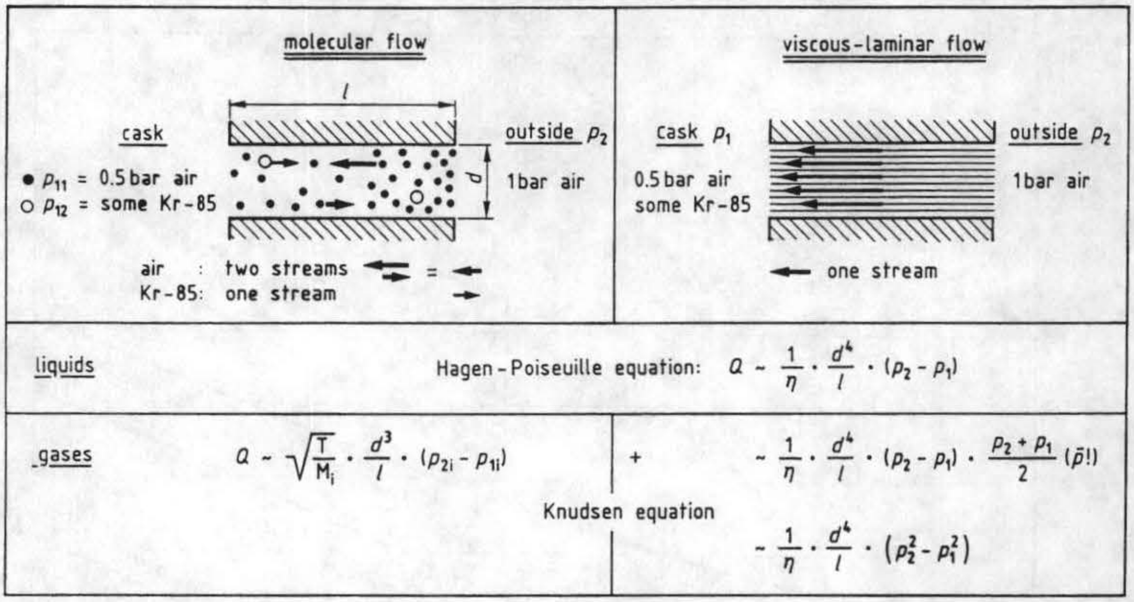


Fig 2. Flow modes of liquids and gases

FIG 3. Air leakage rates Q at pressure differences Δp relative to 1 atm based on an integral standardized air leakage rate $Q_0 = 10^{-4}$ $\frac{Pa \cdot m^3}{s}$ SLR provided by n capillaries of length 5 mm and diameters d'

n		$Q_0'\left[\frac{Pa \cdot m^3}{s}\right]$		$Q_0\left[\frac{Pa \cdot m^3}{s}\right]$	d'[µm]
1	×	10^{-4}	=	10^{-4}	16.0
10		10^{-5}		10^{-4}	8.9
10^2		10^{-6}		10^{-4}	4.9
10^3		10^{-7}		10^{-4}	2.6
10^4		10^{-8}		10^{-4}	1.4
10^5		10^{-9}		10^{-4}	0.69
10^6		10^{-10}		10^{-4}	0.34
10^7		10^{-11}		10^{-4}	0.16
10^8		10^{-12}		10^{-4}	0.075

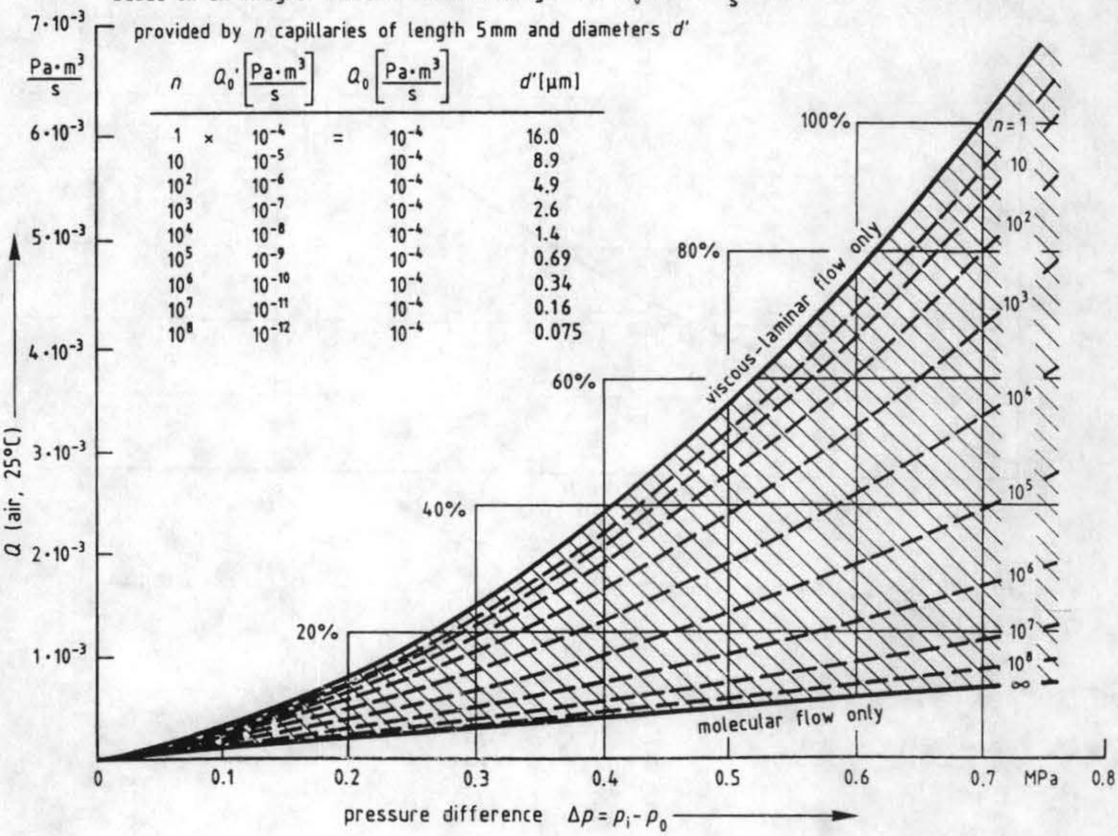
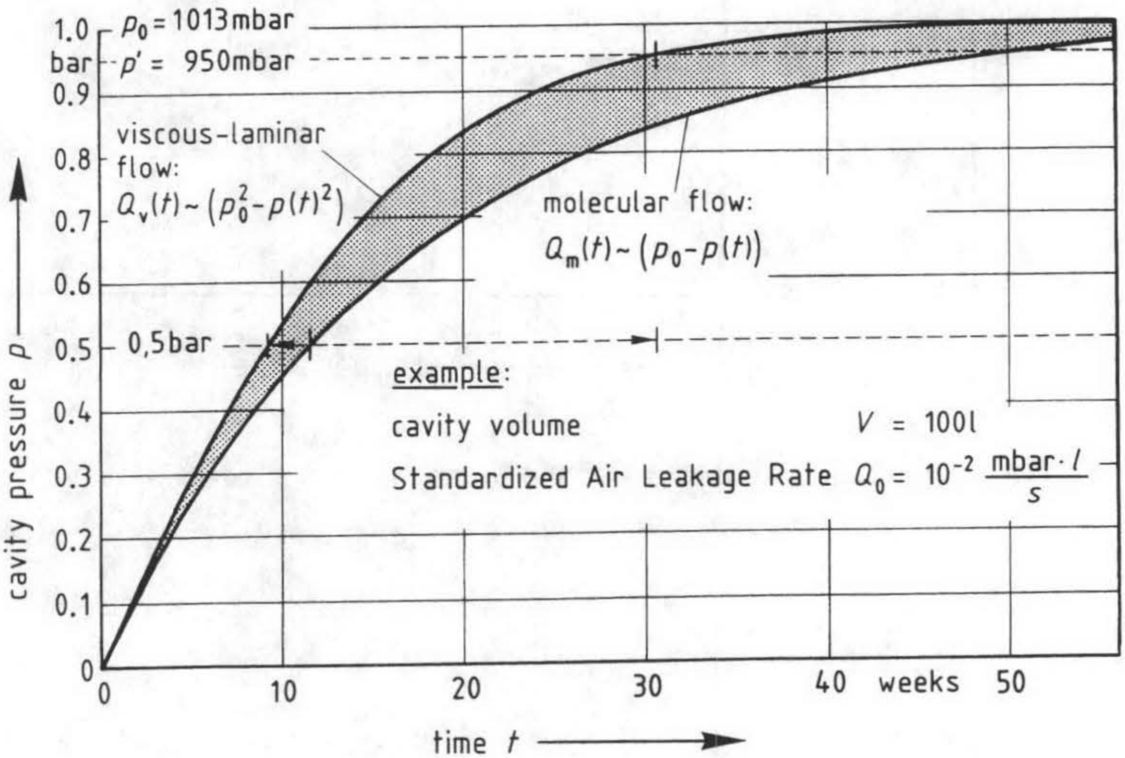


Fig 4.

Pressure Rise by Leakage into an Evacuated Volume

molecular flow	viscous-laminar flow
$p(t) = p_0 \cdot \left(1 - e^{-\frac{Q_0 \cdot t}{p_0 \cdot V}}\right)$	$p(t) = p_0 \cdot \left(\dfrac{1 - e^{-2\frac{Q_0 \cdot t}{p_0 \cdot V}}}{1 + e^{-2\frac{Q_0 \cdot t}{p_0 \cdot V}}}\right)$

short time t: $p(t) \approx \dfrac{Q_0}{V} \cdot t$



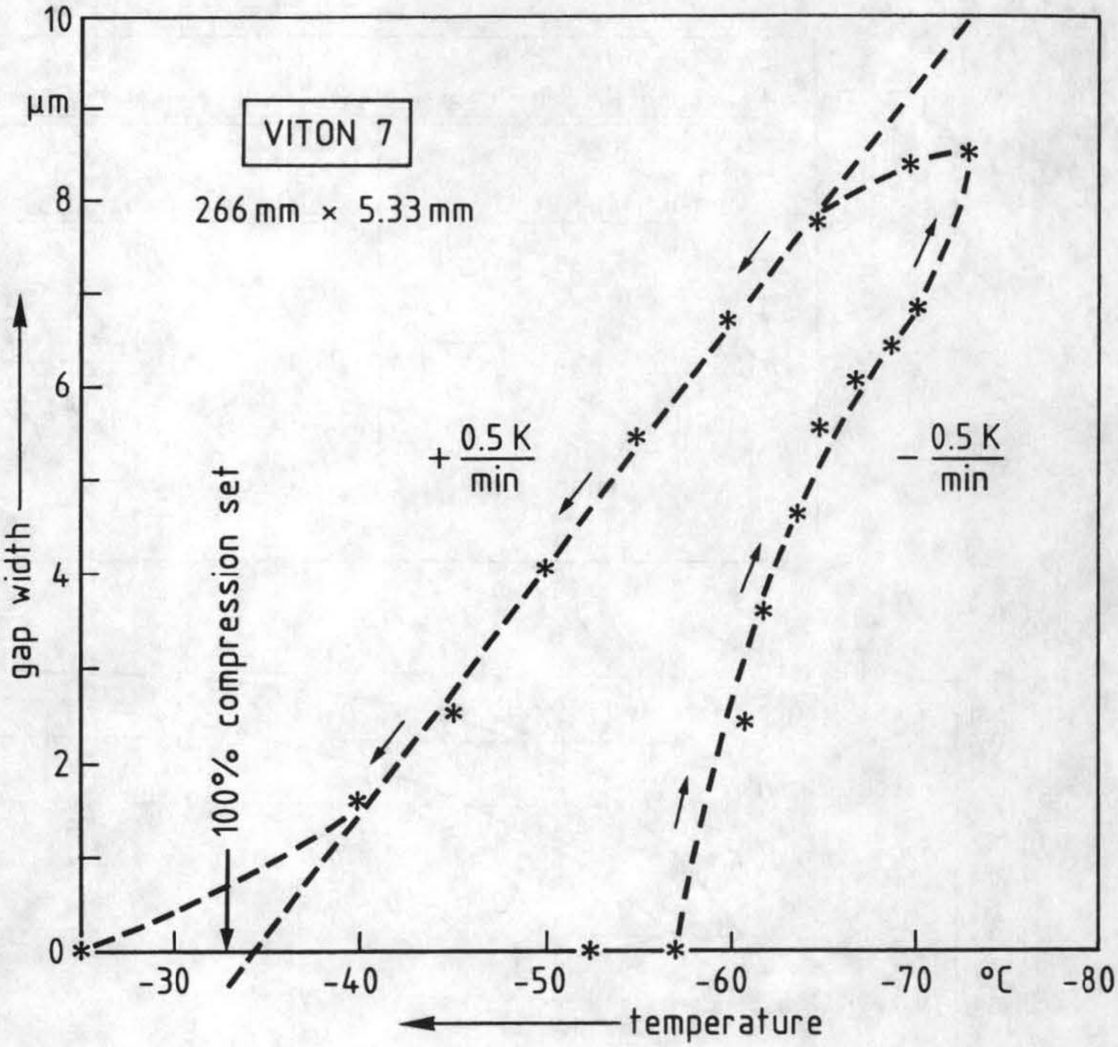


Fig 5. Cooling and heating behaviour of an elastomeric O-ring at low temperature.