

## Modelling Leaktightness in a Sealing System Using Elastomeric Seal

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### 1. INTRODUCTION

Transport regulations limit the release of activity from a radioactive materials package. A leakage test provides global information on the leak and it is necessary to use various models for correlating leakage rates measured during the test and the release of activity in the course of transport.

This study has been initiated to modelize the leakage of gases through elastomeric seal. In order to validate the proposal models, an experimental programme has been achieved using a specific equipment : the "LISE" test ring.

There are two components to gas leakage from elastomeric seals :

- **permeation leakage** : gas passes through the seal itself, which is an intrinsic process for this type of material,
- **by-pass leakage** : gaseous flow through small openings like defects in the seal surface or in the flange surface which are of a random nature.

### 2. EQUIPMENT

The "LISE" test ring (figure 1) is allowed to carry out tests with an O-ring in a dovetail groove (inside diameter between 412 and 330 mm) within a temperature range from -50°C to +300°C.

The leakage rate is measured with a quadrupole mass spectrometer using pure gases or mixture of gases. The control of measurement procedure, data acquisition and analysis, is automated using computers with interfaces to the experiment.

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### 3. PERMEATION LEAKAGE

The permeation of gases is relevant to the solubilisation-diffusion process (e.g., Crank and Park 1968). For gases generally found in package containments like rare gases (helium, krypton, xenon...) and common elastomeric seals (Viton, EPDM, Silicone), the solubilisation may be determined by Henry's law and the diffusion by Fick's law. The time and temperature dependence of the permeation gas flow is well described by theoretical models.

#### 3.1 One dimensional model : slab material

For a flat sample, different relationships can be used to explain the permeation gas flow as a function of time. Using this model, experimental programmes (e.g., Weise et al. 1990) have been developed in order to determine the values of permeation ( $P_K$ ), diffusion ( $D$ ) and solubilisation ( $\alpha$ ) coefficients (with  $P_K = \alpha \times D$ ) and the associated activation energies for the main gas-elastomer couples.

Nevertheless, a realistic estimation of the gas release from typical containment systems using O-ring seals cannot be easily derived from results obtained for flat-geometry permeation.

#### 3.2 Proposal model for a dovetail groove

The similitude between the expression of diffusion (Fick's law) and of conduction (Fourier's law) has been used to develop a 2-D model. Its resolution will be performed by finite elements calculations with the DELFINE code (e.g., Goldstein et al. 1979) used for thermal assessments. This model simulates the dovetail groove designed into the "LISE" test ring. An associated experimental programme is performed with O-ring of 408 mm inside diameter and 8 mm cross section (30 % compression).

Using a model without dimensions, the theoretical calculations give the gas flow through the seal as a function of time at different locations of our models.

A typical permeation process is first studied in which a partial pressure  $P_p$  is applied to the inner side of seal while the outer side is kept under vacuum.

Figure 2 shows the profile concentration on the seal at the permanent state of permeation. It appears that the two cavities at the top of the groove feature an important concentration of gas. For cavity A (near the inner side) :  $P = 2/3 \times P_p$  and for cavity B (near the outer side) :  $P = 1/3 \times P_p$ .

The leakage rate at permanent state is given by :

$$Q_{\max} = 0.468 \times P_K \times P_p \quad (\text{eq. 1}).$$

The first term is relevant to the geometrical parameters of the groove, particularly of the surface area of the inner and outer sides. The leakage rate is proportional to the difference of partial pressure of the gas on the two sides.

Figure 3 shows the gas flow ( $\varphi'$ ) as a function of time ( $t'$ ) (results without dimensions). The gas flow from the outer side which represents the leakage rate, has a typical permeation curve. After a time lag, the gas flow rate rapidly increases and finally saturates for large time. It appears that the initial increase part of the permeation, the filling of cavity A is predominant which delays the progression of gas to the outer side. The same behaviour with smallest amplitude, can be observed for cavity B.

The evolution of the leakage rate relevant to our experimental groove is given by :

$$Q(t) = 0.168 \times \varphi' \times P_K \times P_p \quad (\text{eq. 2})$$

$$t = 3.15 \times 10^{-5} \times \frac{t'}{D} \quad (\text{eq. 3})$$

The permeation gas flow for He through a Viton O-ring seal has been measured for different temperatures. These results are compared with theory (figure 4) and serving as basis to determine the permeation and diffusion coefficients for VITON. These data and the resulting activation energies determined by Arrhenius plot are similar to the data found in the literature (e.g., Weise et al. 1990) which validates our 2-D model.

Using the profile concentration at study state, calculations give an interesting view of the gas release after the permeation process (the inner and outer sides are kept under vacuum). Considering the important concentration of gas in cavities A and B, the outgasing time will be very long and the behaviour of these cavities can be compared to those of punctual sources of gas.

### 3.3 Theoretical prediction

The theoretical results shown in figure 3 can be used directly in order to estimate the leakage rate of various gases under different pressure and temperature conditions. In this view, the data relevant to the gas at the test conditions ( $P_p$ ,  $P_K$  and  $D$ ) must be introduced in equations 2 and 3.

An example is given for a mixture of gases ( $\text{He} + \text{N}_2 + \text{Kr} + \text{Xe}$ ). These gases are representative of an irradiated fuel shipment and the associated partial pressures are chosen in accordance with experimental conditions (sensitivity of the mass spectrometer, test duration...).

The permeation gas flows are calculated for a Viton O-ring at  $150^\circ\text{C}$  and the results are shown in figure 5. Currently, measurements are under way in order to verify this prediction.

The results concern an O-ring with 408 mm inner diameter in a dovetail groove ("LISE" test ring model). In a first approximation, the estimation for other diameters can be made with a correction corresponding to the ratio of inner diameters.

#### 4. BY-PASS LEAKAGE

The gas leakage through small openings depends upon the flow mode and the geometrical aspect of the defects. In order to use the test results, the equations of Poiseuille and Knudsen for viscous and laminar flow may be applied. The gas leakage can therefore be estimated by an empirical relationship (e.g., Blanc et al. 1982).

$$Q = (C_L + Z C_M) (P_u - P_d) \quad (\text{eq. 4})$$

The terms  $C_L$  and  $C_M$  pertain respectively to laminar and molecular flow ; the term  $Z$  (Knudsen's factor) varies between 0.81 and 1. All of them depend upon the pressure ( $P_u$ ,  $P_d$  : upstream and downstream pressures), the temperature, the nature of gas and the geometrical characteristics of the defect.

This equation has a general expression and can be used for most defects (e.g., Henry 1973). Nevertheless, the most representative defects are channels of round and rectangular cross sections (e.g., Rossi 1990).

Currently, the most commonly used model assumes that all leakages occur through a single capillary. Taking into account the fact that the leakage test does not provide any information about the number and the geometrical nature of the defects, it appears that a model which does not use any assumption for these parameters will be more representative.

##### 4.1 Proposal model

For a majority of defects like capillaries and cracks, the equation 4 can be rewritten with a separation of the factors depending upon the geometrical data of the defect and upon the test conditions. This development is explained in (e.g., Langlois 1992). Two new parameters,  $L_u$  and  $V_a$ , have therefore been defined and the resulting expression for a single defect is :

$$Q = \left( Lu \frac{\bar{P}}{\eta} + \frac{Va}{\sqrt{\rho u}} \right) (Pu - Pd) \quad (\text{eq. 5})$$

Where : Lu, Va : two shape factors.

and :  $\eta, \rho u, P$  : experimental data relevant to the test conditions\*.

In case of n leaks, the total leakage rate is given by :

$$Q = \sum_{i=1}^n Q_i = \sum_{i=1}^n \left( (Lu(i) \frac{\bar{P}}{\eta} + \frac{Va(i)}{\sqrt{\rho u}}) (Pu - Pd) \right)$$

$$Q = \left( Lu_g \frac{\bar{P}}{\eta} + \frac{Va_g}{\sqrt{\rho u}} \right) (Pu - Pd) \quad (\text{eq. 6})$$

$$\text{where } Lu_g = \sum_{i=1}^n Lu(i) \text{ and } Va_g = \sum_{i=1}^n Va(i)$$

Therefore, the relationship used for calculating the leakage rate of one or n defects is similar, and all the geometrical characteristics of the defects are represented by shape factors Lu and Va.

With an appropriate test such as measurement of the evolution of pressure or of leakage rate, the two shape factors Lu and Va can be determined by a straight line representation of the first term of the equation 5 as a function of  $\bar{P}$ .

The leakage rate for other conditions is calculated by introduction in the relationship (eq. 6) of these two factors Lu and Va, and of the new data relevant to these conditions ( $\eta, \rho u$  and  $\bar{P}$ ).

#### 4.2 Experimental validation

This model has been validated using a pressurized vessel fitted with metallic gaskets (no permeation leakage). After determining factors Lu and Va by testing, a comparison with experimental results for another gas (argon) and for a mixture of gases (helium + argon + nitrogen) shows a good correlation with predicted results (figure 6).

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\*  $\bar{P} = \frac{Pu + Pd}{2}$  and  $\rho u = \frac{M}{RT}$  (v : viscosity, T : temperature, M : molecular weight, R : universal gas constant)

A similar correlation is performed with the single capillary model (determination of the hole diameter assuming a length of 1 cm). It appears (figure 6) that in this case, the results obtained are more pessimistic.

## 5. CONCLUSION

According to the fact that the permeation is an intrinsic process for elastomeric seal, the results without dimensions obtained with the 2-D model can be used in order to make predictions. Therefore, the release of activity can be determined, and the ratio with by-pass leakage rate during a leakage test using a gas like helium can be estimated.

The new expression given to the leakage rate for by-pass leakage is a general relationship which is more adapted to correlate test measurements with the release of activity during transport. When comparing the two methods, it appears that the shape factor model is quite realistic and less conservative than the single capillary model.

## 6. REFERENCES

BLANC B., HENRY R.P. et LECLERC J. ; Guide de l'étanchéité, vol.2, Les fuites, 1982.

CRANK J. and PARK G.S. ; Diffusion in Polymers, Academic Press, London and New-York, 1968.

GOLDSTEIN S., JOLY J., JUIGNET N., "Some numerical algorithms and applications of the DELFINE computer program, numerical methods in thermal problems". Proceedings of the First International Conference, Swansea, Wales. July, 2<sup>nd</sup> - 6<sup>th</sup>, 1979.

HENRY R.P. ; Cours de Sciences et Techniques du Vide, Livre I, Tome II, 1973.

LANGLOIS B. - Contribution à la qualification des méthodes de contrôle de l'étanchéité des colis de transport de matières radioactives - Thèse en cours.

ROSSI L. ; Défauts d'étanchéité, Approche de caractérisation, La revue des Laboratoires d'Essais, 1990.

WEISE H.P., ECKER K.H., KOWALEWSKY H. and WOLK T. ; Gas permeation through common elastomer sealing materials, Vuoto, Vol. XX, N.2, 1990.

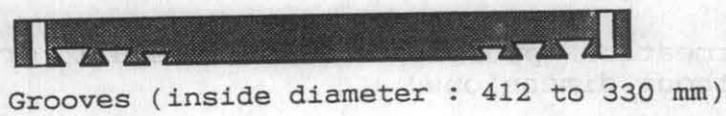
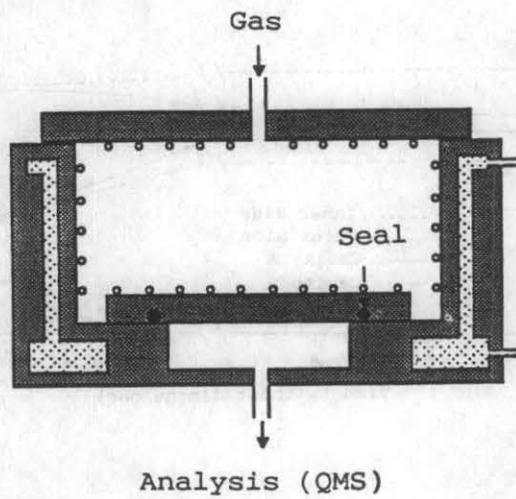


Figure 1 : The "LISE" test ring

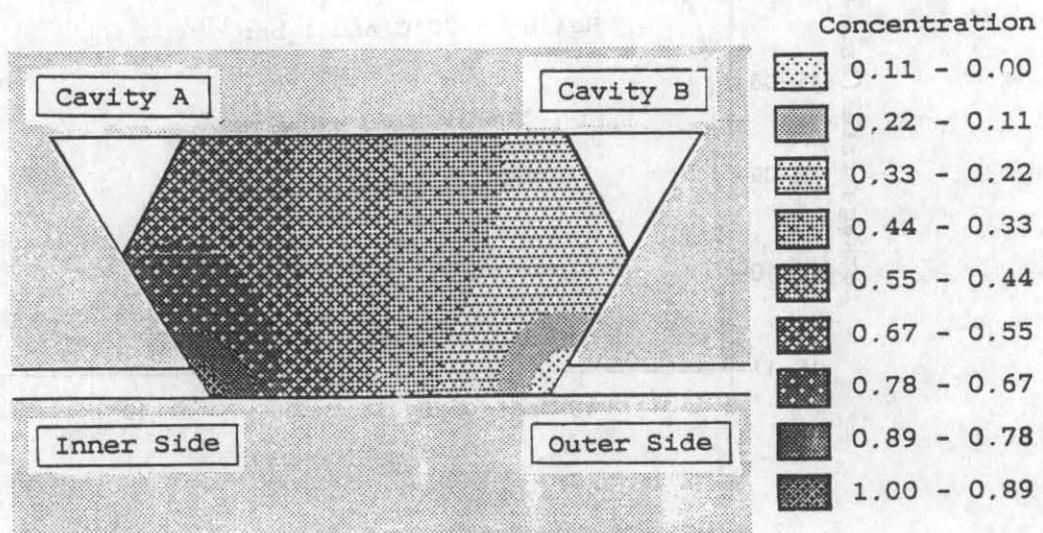


Figure 2 : Concentration profile on the seal at the permanent state of permeation (results without dimensions)

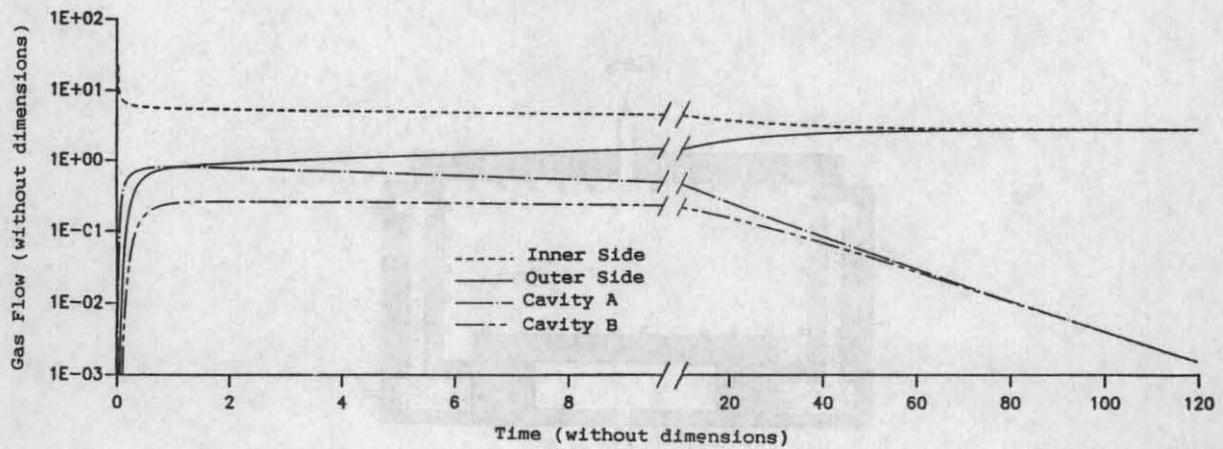


Figure 3 : Permeation profile for a dovetail groove (results without dimensions)

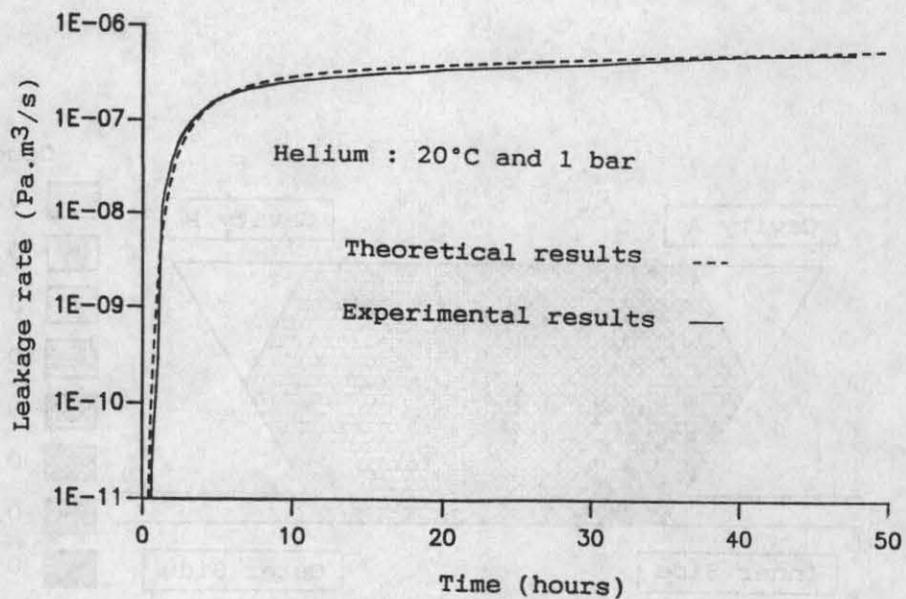


Figure 4 : Correlation between theoretical and experimental results for permeation of helium through VITON at 20°C

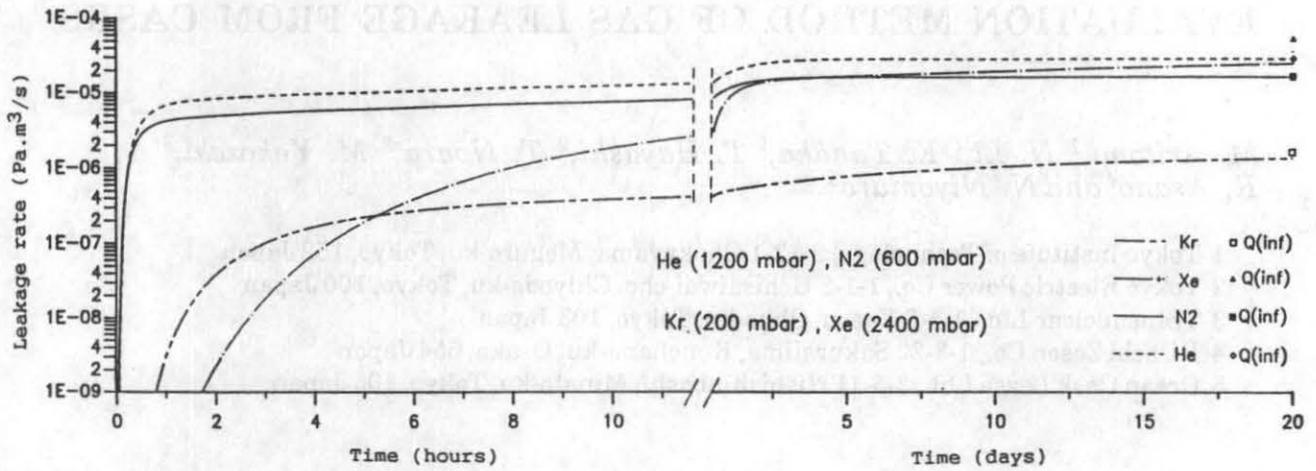


Figure 5 : Predicted profile for permeation of a mixture of gases

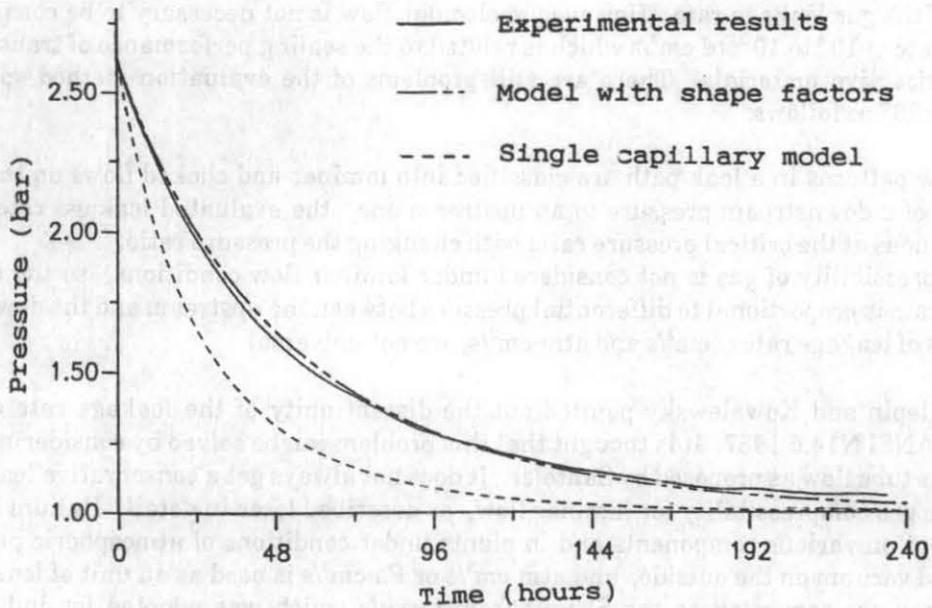


Figure 6 : Theoretical and experimental evolution of pressure for a pressurized vessel