SAFETY ASSESSMENT OF LEAKTIGHTNESS CRITERIA FOR RADIOACTIVE MATERIALS TRANSPORT PACKAGES

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SUMMARY

Preliminary results of a running research project, funded by the European Commission, that will be finished at the beginning of 1998 are presented.
The aspects described are focussed on the high and low temperature performance of elastomeric O-ring seals, consideration of seal ageing, and studies of aerosol leakage phenomena.

INTRODUCTION

Initiated by the IAEA, from 1987 a group of experts from Belgium, Canada, France, Germany, Japan, the UK and the USA developed ISO 12807 "Safe transport of radioactive materials - Leakage testing on packages" published in Sept. 1996. It was based on a similar ANSI Standard N14.5 from 1977 and revised in 1987. The next revision should bring both standards together. To support this need, to fill some gaps of knowledge, and to promote acceptance of radioactive materials transport a research programme on the assessment of seals and leaktightness, supported by the European Commission (contract no. 4.1020/D 196-004), has been performed by BAM, Germany and AEA Technology, UK.

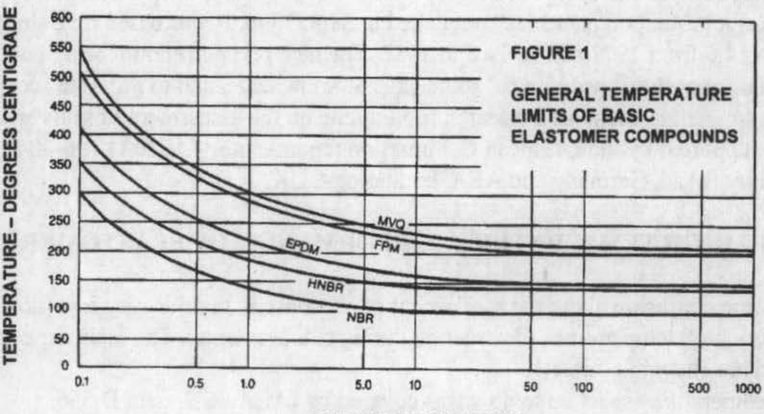
LOW AND HIGH TEMPERATURE PERFORMANCE OF ELASTOMERIC SEALS

There is some confusion about the assessment of elastomeric seals at very low (down to - 40 °C) and very high temperatures (destruction during fire accidents). The main aspects to be considered are shown in Table 1.
The low temperature aspect up to now was covered by IAEA SS 7, para E-556:
"The lower temperature is important because of pressure increases from materials which expand upon freezing (e.g. water), because of possible brittle fracture of many metals (including some steels) at reduced temperature and because of possible loss of resilience of seal materials."
We have proposed to IAEA an amended version to clarify the situation:
"Of these effects, only fracture of materials could lead to irreversible damage. Some elastomers which provide good high temperature performance (e.g. fluorocarbons, such as Viton compounds) lose their resilience at temperatures of - 20 °C or less. This can lead to narrow gaps of some μm width arising from differential thermal expansion between the metal components and the elastomer. This effect is fully reversible. In addition, freezing of any humid contents and internal pressure drop at the low temperatures could prevent leakage from the containment. Therefore in certain cases the use of such elastomeric seals could be accepted."

parameters	at low temperature	at high temperature
elastomeric seals	inelastic thermal contraction (reversible effect)	destruction (irreversible effect)
permeability of the elastomer (enables permeation of gaseous contents)	low	high
leakage path geometry - for the design	uniform narrow gap	porosity, cracks
- in case of defects (e.g. scratches in the sealing face, ...)	capillaries, ...	
driving pressure	below atmospheric	high
permeation and leakage rates	low	high
consideration of radiological consequences by dispersion of released activity	slight	strong (typical: fire accident)

Table 1 : Leakage Considerations for the Usage of Elastomeric Seals

For the region of high temperatures a seal manufacturer (Parker 1994 and earlier) has published a typical graphical representation (fig. 1) which recently has been verified for some examples of practical importance (Bronowski 1995).



Some years ago BAM had performed some research work which now has been continued. A special Viton compound FPM MK 634 had been developed by a German seal manufacturer which shows leaktightness down to about - 40 °C (Kowalewsky 1995), see fig. 2, being more suitable than Viton E60 C, see fig. 7.

Following the cooling period and after reaching a critical temperature which is below the temperature where the compression set becomes 100 % - representing transition to inelasticity - the 0-ring detaches discontinuously from the metallic sealing face. Because the thermal expansion/contraction coefficients for elastomers are smaller than those for metals, and because there is at first some residual adhesion between elastomer and flange surfaces, gradually a gap between elastomeric seal and metallic sealing face develops. With increasing temperature fully open gaps close steadily and reversible, there is no further evidence and memory of this previous effect. From any pressure rise in the vacuum system air leakage rates can be derived and corresponding gap heights can be calculated shown in fig. 2 and being in the range of only a few μm for the temperatures of interest down to - 40 °C.

Some other experiments have been performed at fire accident conditions. Permeation through and failure of elastomeric 0-rings (152 mm x 5.3 mm)had been investigated at high temperatures in a mixed helium/air atmosphere, (Weise et. al. 1986). Our experimental heating rate in a furnace was very similar to a calculated temperature rise in the outer lid sealing region of a transport and storage cask for spent fuel exposed 4 hours to an ambient fire temperature of 800 °C. Helium permeation rises with increasing time/temperature until elastomer destruction causes high leakage for the EPDM compound at about 380 °C, for the Viton compound at about 390 °C, and for the silicone compound at about 400 °C. These 0-rings were irreversibly destroyed.

The same test apparatus and heating rates have been used for some further experiments (Weise et. al.1993). Helium permeation rates during repeated thermal cycling with increasing maximum temperatures are shown in fig. 3 - 5. The upper temperatures used for these experiments (about 305 °C for the EPDM compound, about 300 and 350 °C for the Viton compounds, are not comparable, there might be different safety margins. Fig.3 shows the last two of four cycles: During the last period of constant temperature permeation even decreases which might be due to the observed permanent deformation of the O-ring to the dovetail groove geometry. At any case they are not far below the measured temperatures of destruction and higher than temperatures recommended for current use, especially concerning EPDM.

ELASTOMERIC O-RING-SEAL PERFORMANCE: SEAL AGEING (AEA Harwell)

Low temperature effects in seals for transport containers have previously been studied also at AEA Harwell for a number of different elastomers. These studies have looked at changes in the leakage rate, sealing force or seal gap as a function of seal temperature to identify the lower limits of practical use of typical seal materials. Most of this work has been on new seals with little work on the sealing behaviour of aged seals.

The test data to be presented examines the low temperature sealing behaviour of several Viton and EPDM seal materials after ageing, covering both thermal and radiation ageing of the seals. Measurements of sealing force and leakage rate will be presented over the temperature range down to - 40 °C, comparing the behaviour of both new seals and seals aged to different levels of degradation.

Test rig

Low temperature seal tests have been carried out using a test rig containing a single O-ring groove as described in Burnay, 1991. The rig is cooled via refrigerant fluid circulated through coolant channels in the flanges. The refrigerant temperature is controlled by a Neslab model LT50-DD refrigerated cooling bath. This enables the flange temperature to be controlled to ±0.2°C down to -40°C.

Measurements of the sealing force exerted by the seal on its sealing face are obtained using a pressure transducer (Entran EPX M5 35) mounted directly over the O-ring seal. This gives a relative measurement of the changes in sealing force as the temperature changes. Leakage rate measurements are made via the pressure drop method, using a high sensitivity pressure transducer (Entran EPV 835X) mounted adjacent to the central well.

The materials which have been tested are Viton E60C fluoroelastomer, silicone (Dowty 8866) and EPDM, all in the form of O-rings of 5.3 mm cord diameter and 73 mm internal diameter.

Sealing force measurements

In the Viton material tested, the measured sealing force decreases linearly with decreasing temperature down to about -20°C, then decreases very rapidly until it reaches zero at about -30°C (fig. 6). When the temperature is increased, the sealing force starts to increase again, regaining its initial value as the temperature reaches near ambient conditions. However, there is a marked hysteresis in the behaviour between -25°C and -20°C, with the sealing force increasing rapidly and then decreasing. The thermal cycle shown in fig. 6 and 7 for a temperature change rate of 2°C/hr.

Leakage rate measurements

In the Viton seal material, leakage rates remain constant at $(5.2 \pm 0.8) \cdot 10^{-6}$ Pa $\cdot$ $m^3 \cdot s^{-1}$ for temperatures down to -25°C and then increase very rapidly as the temperature decreases (fig. 7). This rapid increase corresponds to the sealing force decreasing to 15% of its initial value. When the temperature increases again, the leakagerate starts to decrease at around -30°C but does not return to its initial value until the temperature has increased to -17°C.

A limited amount of work has also been carried out on the effects of ageing on the sealing performance of the EPDM seals. These have been thermally aged at 200°C, which is a temperature which would be expected to cause severe degradation of the seal material. Leakage rate measurements have only been made at 20°C so far but the effects of ageing on low temperature performance are currently undergoing investigation. Thermal ageing at 200°C increases the leakage rate significantly in the first few hours of ageing and then rises more gradually. Even after 200 hours of ageing, the leakage rate has only increased by an order of magnitude over the initial value for the new seal. Examination of the seal after ageing showed that, although the surface of the seal was cracked, the bulk of the material remained flexible and leakage was confined to bypass leakage through the surface cracks.

Discussion

As temperatures are decreased, the sealing force exerted by an elastomeric seal on its sealing face will decrease due to the combined effects of the Joule-Gough effect and differential thermal expansion. Leakage rates past an elastomeric seal are usually independent of the sealing force exerted, provided it is above a minimum level, therefore leakage rates do not change significantly as the temperature decreases until this limit is reached. If the temperature range of interest includes the region near the glass transition temperature T_g then large changes in the coefficient of thermal expansion occur and the observed sealing force decreases very rapidly. This is the type of behaviour which is seen in the Viton seals examined.

In the silicone and EPDM seal materials, the data from leakage rate and sealing force measurements indicate that the glass transition temperature is well below -40°C.

In the Viton E 60 C material, which goes through T_g during the thermal cycle from +20°C to -40°C and back, leakage rates increase dramatically when the sealing force decreases below 15% of the initial value and does not fully recover until the transition is completed and sealing force has returned to 60% of its initial value. These specific values refer to a sealing face with a surface finish of 1.5 to 2 µm. For other surface roughnesses, these values may change.

MATERIAL LEAKAGE FROM TRANSPORT PACKAGES (AEA Winfrith)

Previous studies of aerosol leakage from transport containers at AEA Winfrith have concentrated on idealised geometries, in particular short, circular capillaries (10-50µm internal diameter and 5-50 mm in length) challenged by micron-sized particles. When a pressure differential of 100 kPa was applied across a capillary, the aerosol penetration rate was directly proportional to the air leakage rate for a given upstream aerosol concentration, and there was little evidence of leakage-path plugging (Ball and Morton, 1996). Some limited tests, on capillaries of length 30 mm and bore 20 µm, showed that aerosol penetration was attenuated at lower differential pressures (<60 kPa), indicating plugging of the leakage-path (Morton and Mitchell, 1995). The results of further experiments on pressure effects (funded by CEC Contract FI3S-CT92-0007 and by the UK department of Trade and Industry) are reported here.

Experimental Details

The aerosol leakage test apparatus consisted of a 9 litre cylindrical vessel with a fluidised bed aerosol generator located at the base (Morton and Mitchell 1995). The pressure in the vessel was maintained between 10-100 kPa. The capillary on test was mounted vertically between the pressurised vessel and the particle collection vessel at ambient pressure. The aerosol consisted of well-characterised micron-sized glass microspheres. Particles that passed through the capillary were swept to the inlet a near real-time single particle counter which allowed sudden events such as capillary blockage to be detected as they happened.

Results and Discussion

The first set of tests were performed with fixed driving pressures, using capillaries of 30 µm bore and 20 mm in length, in order to confirm the results of the earlier tests. Capillaries exposed at 15, 20 and 40 kPa blocked within 30 minutes, as was observed in the previous work. The plugs consisted of large, well-developed hemispheres which covered the capillary entrance. The data from the particle counter indicated that capillaries exposed to 60 and 80 kPa did not block, while post-test examination found a small plug at the capillary entrance in both cases. The test conducted at 100 kPa driving pressure did not plug, but its penetration efficiency decreased through the test. Post-test examination showed that a build-up of particles had occurred around the rim of the capillary entrance. Air leakage testing of the capillaries before and after the aerosol tests found that large plugs often caused no substantial restriction to the air flow. The model developed to describe aerosol penetration through capillary-like leakage-paths (Clement 1995) predicts that aerosol will deposit on capillary walls near the entrance due to inertial impaction, as found in these experiments. Clement also noted that it is more appropriate to relate blocking to the total number of particles leaked rather than to time.

Analysing the experimental data showed that the total number of particles penetrating the capillary before plugging occurred, increased with the driving pressure.

The second set of tests examined the stability of the plugs formed. The capillaries were exposed to aerosol at a driving pressure of 20 kPa until blocked and then the pressure was rapidly increased at various rates (0.4 to 0.8 kPa s^{-1}). Unblocking consistently occurred at about 70 kPa, as shown by a burst of particles penetrating the capillary. However, this was followed in all cases by rapid re-blocking, and the re-formed plug was stable up to the maximum applied pressure of 100 kPa. Hence, once a plug has formed, it may be affected by pressure increase, but always reforms and is then stable to higher pressure. The third set of tests examined to examine the scaling effect of increasing the capillary length and diameter. It was intended to see if these capillaries, which had the same nominal standard leakage rate (SLR) as the capillaries used in the first tests, would behave in the same way, or if a larger opening was less likely to block. The results showed that there was no increased leakage for increased capillary diameter at a given SLR and pressure difference. In fact, since the actual SLR values of the capillaries covered a range around the nominal value, it could be seen that the number of particles penetrating before blocking occurred increased with SLR for a given driving pressure.

Testing with more realistic leak geometries is required to demonstrate that the results of the capillary tests represent the 'worst-case'. Hence, the aerosol leakage facility has been modified so that the capillary is replaced by a single Viton 0-ring-to-metal seal, suspended horizontally within the challenge aerosol, and containing deliberately created faults. The aim is also to describe the aerosol penetration by extending the existing model for straight capillaries (Clement 1995). Clement noted that, prior to the onset of plug formation, there is no significant deposition of aerosol in the capillary. Violation of the expected relationship between gas and particle leakagerates indicates the onset of plug formation. Once plugging begins, experience shows that it quickly becomes complete. For inertial deposition of particles at the entrance to the leakage path, Clement has shown that, in terms of the upstream and downstream pressures, p_u and P_d, plugging scales as, X, where

$X = (p^2_u - p^2_d)/Lp_u$ and L is the length of the leakage path. (1)

He used this result to show that, in previous experiments, plugging was indeed dominated by this mechanism. However, if the entrance is more streamlined than for the capillaries, the rate of impaction at the entrance may be significantly reduced. If, in addition, the flow path is horizontal, gravitational settling may become important, or even dominant. Different scaling relations for penetration cut-off are found for different deposition mechanisms. A general dependence on path radius (R) of the form R^n/L is supported with n = 1 - 3, for circular capillaries. Hence, it is important that the test matrix includes variations in the radius to length ratio of the flow path and the ratio X defined by equation 1, in order to determine the dominant aerosol deposition mechanism(s) and derive a correlation for particle leakage. In addition, the test matrix should also include variation of leakage path-geometry (e.g. corresponding to scratched O-rings or entrapment of dust particles or hair etc.) and possibly material properties.

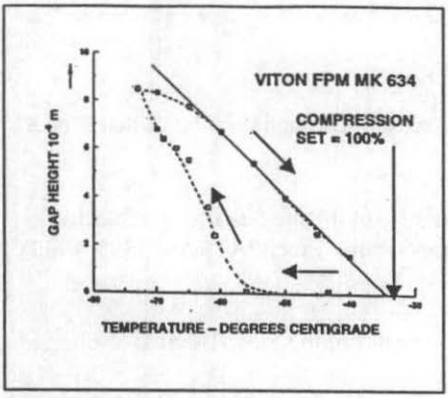


Figure 2

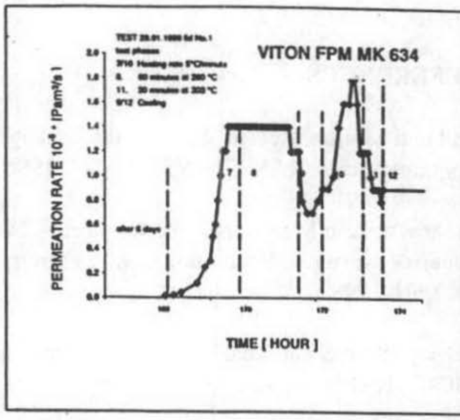


Figure 3

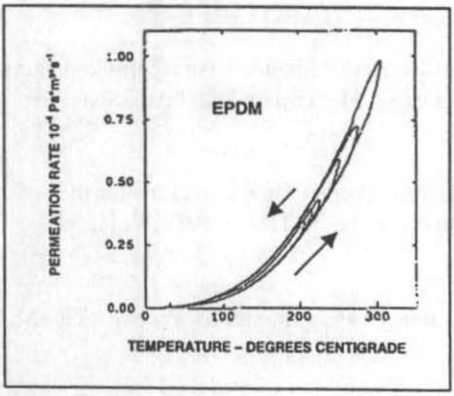


Figure 4

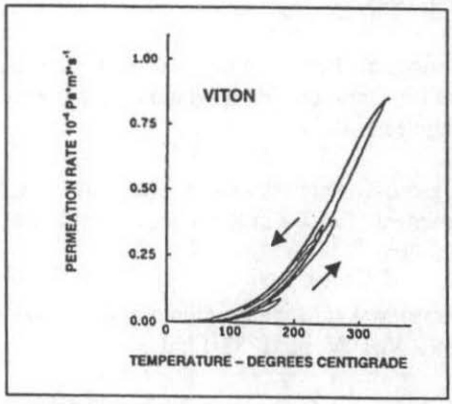


Figure 5

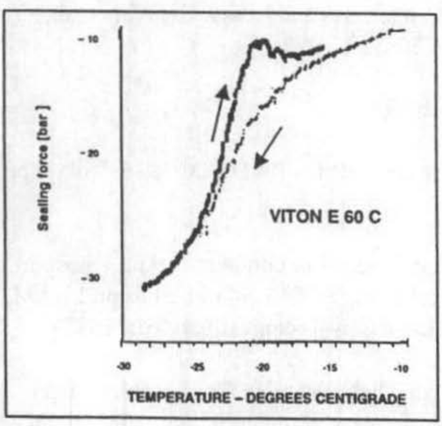


Figure 6

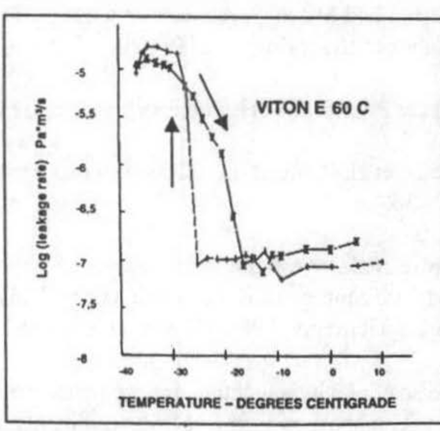


Figure 7

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