Ageing of Helicoflex[®] metallic gasket for spent fuel cask: results and analysis of sealing performances of a 75,000h campaign

A.Béziat^a, F.Ledrappier^b, <u>K.Vulliez^a</u>, B.Deschamps^a, J.F.Juliaa^b, L.Mirabel^a, M.Wataru^c, K.Shirai^c, H-P.Winkler^d

^a CEA, DEN, SDTC, Laboratoire d'Etanchéité, 30207, Bagnols Sur Cèze France.

^b TECHNETICS Group France, Laboratoire d'Etanchéité, 2 rue James Watt 26700 Pierrelatte.

^c Central Research Institute of Electrical Power Industry, 1646 Abiko, Abiko-shi, Chiba-ken 270-1194 Japan

^d Gesellschaft für Nuclear-Service GmbH, Hollestrasse 7A, 45127 Essen

Corresponding author: K.VULLIEZ (<u>karl.vulliez@cea.fr</u>)

ABSTRACT

In the framework of CEA, TECHNETICS GROUP France, GNS and CRIEPI collaboration an experimental program is being carried out to assess the long-term use of HELICOFLEX® metallic seals in spent nuclear fuel storage casks. The harvested data on more than 60 mockups after 75,000h of ageing provide a consistent database to extrapolate the long-term sealing performances to lifetime exceeding 100 years. After a description of the HELICOFLEX[®] gasket and of the ageing program, the results of all the measurement campaign are discussed using an analytical model to interpret them from a statistical point of view. The paper then focuses on the finite element approach implemented to consolidate the experimental protocol and the statistical analysis hypothesis. A 3D-model of the mockup is first used to check and understand the mechanical behavior of the assembly (two bolted thick stainless steel blind flanges). Based on the comforting results of this analysis, a detailed 2Daxisymmetric model of the seal is then developed to assess qualitatively the time extrapolation method. In the numerical simulation time-dependent loads are applied on the seal to replicate the different loading cycles inherent to the experimental protocol. The non-linear behaviors of the seal materials are also considered (plasticity of spring and inner layer, plasticity and viscoplasticity of silver outer lining) to describe as precisely as possible the gasket load relaxation along the ageing. The detailed description of the most relevant features and outcomes of these calculations are presented. In the light of the statistical analysis of the experimental data and prediction methods, providing a maximum temperature of 'safe' use, the sealing performances of the HELICOFLEX[®] over duration exceeding a century can be assessed.

INTRODUCTION

Long-term dry storage of spent fuels in metal casks is a very commonly used solution. This technical choice requires the use of leak-tight casks and therefore seals to ensure a containment criterion [1]. Among a large panel of existing sealing systems, the expected lifetime of the casks, that can exceed one hundred year, combined with thermal constraints favor the use of metallic seal based technologies [2]. The optimal seal arrangement to efficiently ensure the long-term tightness between the cask closure flanges relies on a Metal To Metal (MTM) contact between them [3][4], a mechanical junction where the gasket load relaxation is one of the key parameters regarding sealing performances over time [5].

To investigate the detrimental loss of leak-tightness linked to the time-evolution of the seal materials, a long-term ageing experimental campaign is conducted on a large panel of mockups that consist in two symmetric flanges holding tightened HELICOFEX[®] gasket. The works aims at evaluating the minimum residual linear load that can be guaranteed for a seal after a given time of relaxation. Taking benefit of the influence of temperature on creep kinetics and thus on seal relaxation, the ageing of the mockups in furnace is made at different constant temperatures: Room Temperature (RT), 100 and 200°C. The residual load and 'useful' spring-back have been measured after the holds under an instrumented press. The measurement were made after 10,000, 25,000 [6], 50,000 and 75,000 hours. The harvested results provide a large database that can be interpreted according to two methods based on a time-temperature equivalence Larson & Miller Parameter (LMP) [7] and a time extrapolation. Both methods relies on assessed linear relationships through statistical analyses (calculation of scatter applied to the large database) also used to estimate the minimum guaranteed residual load for a given period of time. Finite element model are implemented to validate the experimental protocol and the hypothesis considered in the statistical analyses.

Outer Lining Inner Lining Helicoidal spring

THE HELICOFLEX GASKET

Figure 1. HELICOFLEX[®] metallic gasket.

The HELICOFLEX[®] is a metallic seal (Fig.1) patented and manufactured by TECHNETICS Group which consists in a helicoidal spring covered by several linings. With a proper combination of materials for spring and linings, this mechanical structure can provide a very interesting sealing system tolerant to the flanges surfaces imperfections (roughness, waviness), delivering a significant spring back able to withstand important deformations. The HELICOFLEX[®] is therefore well suited for long-term use in nuclear spent fuel casks.

The spring of the gaskets tested in this study are made of Nimonic 90, and the 0.3 mm thick outer lining is in pure silver. Two different cross-sectional diameters are studied 6.2mm and 8.4 mm with 304L Stainless Steel (SS) inner lining respectively of 0.3 mm and 0.4 mm. The choices of suitable material for the outer lining is driven by considerations about hardness, creep resistance and corrosion properties. Thus, preliminary 10,000 hours ageing tests reveal that silver is suitable for long-term use. For the study the seal mean-diameter is 250 mm, to be compared with the real application diameters ranging from 1 to 2 meters.

The gasket mechanical behavior can be described by the set of parameters summarized in Table 1 and Fig.2. Among them, the most important is the Y_2 which represents the recommended linear load in Newton/mm required to tighten the seal and e_2 the corresponding compression of the seal (height difference between an untightened and tightened seal).



Figure 2. Loading-unloading curve of a seal

Table 1. Nomenclature

Y	Linear load of a seal (reaction force of the seal on its flanges, divided by the seal perimeter).
Y_0	Linear load of seal required to obtain the tightness during the first seal tightening.
Y_1	Linear load below which the leak rate exceeds a target value, upon decompression of a seal.
Y_{1R}	Linear load below which the leak rate exceeds a target value, upon decompression of a seal after relaxation
<i>Y</i> ₂	Linear load of a seal (just after tightening at room temperature) when the metal-to-metal contact is achieved. The compression of the seal is then the initial height of the seal minus the height of the groove. This compression is e_2 .
Y_{2R}	Residual linear load (after relaxation due to a temperature hold).
r_t	Total recovery.
r_{tR}	Total recovery after relaxation.
r_u	Useful recovery (no leak)
r_{uR}	Useful recovery after relaxation (no leak)

EXPERIMENTAL PROCEDURE

The seal is mounted in a groove between two rigid blinded flanges. The experimental protocol begins with the first compression of the seal between the flanges with a hydraulic instrumented press to achieve the MTM contact. During the process the load F and the seal compression Δe are measured (Fig.3). The Y_2 and the corresponding seal compression e_2 are determined when the MTM is obtained. The bolts of the mockup, kept under the press, are then screwed up to a given torque. Once released from the press, mockups are piled up inside an oven maintained at a constant temperature. In this program about 40 mockups are used for each of the three selected temperatures: RT, 100°C and 200°C. As a reminder, in a real-life cask the maximal expected temperature at the seal location will be in the range of 150°C and will decay with time down to 50°C.



Figure 3. Test principle: Left side - mockup under the press. Right side - mockups piled up inside a furnace

After a given ageing period, the mockups are removed one by one from the oven and once at RT placed back under the instrumented press. Then, they are loaded up to the Y_2 load before removing bolts. The load applied by the press is then decreased down to an opening of the flanges of 50 µm, while the load and the distance between the flanges are measured to obtain the unloading curve. It is estimated that when the recovery reaches 25 μ m all the direct contact between flanges is lost, due to their bending. The measured data for recovery values ranging from 25 to 50 µm are used to evaluate a best fitting second order polynomial curve. This range is chosen because the first part of the decompression curve (from 0 to 10 μ m) can be affected by the mockup stiffness and is not representative of the gasket properties. The upper limit of 50 µm is fixed by the experimental protocol, to avoid any influence of contact lost between seals and flanges on its sealing performances. The Y_{2R} is then derived by an extrapolation as shown on Fig.4. During the decompression, the leak rate is monitored with a helium spectrometer to evaluate the Y_{IR} corresponding to the linear load under which the leak rate exceeds an accepted limit (10⁻⁸ atm.cm³.s⁻¹). For ageing time exceeding few thousands of hours even at RT, the Y_{IR} can be equal to zero, meaning that the mockups remain vacuumtight with leak rate below the limit. That point is linked to a very close contact between seals and flanges roughness related to the outer lining creep.



Figure 4. Example of *Y*_{2*R*} determination: mockup#137, Ø6.2 mm section silver lining seal held at 200°C for 75,000 h. Blue curve: leak rate measurement. Red curve: residual seal loading.

DATA ANALYSIS



Figure 5. Seal with \emptyset 6.2 mm cross section: Y_{2R} values determined by linear interpolation for 3 temperatures (blue 20°C, black 100°C, red 200°C). Two points connected by a dotted line correspond to values measured on the same seal.

Cross section Ø6.2mm								
	RT 20°C		100°C		200°C			
	75 000 h	100 000 h	50 000 h	75 000 h	50 000 h	75 000 h		
Mean value of Y _{2R} [N.mm ⁻¹]	340	339	270	250	141	-		
Cross section Ø8.4mm								
	RT 20°C		100°C		200°C			
	75 000 h	100 000 h	50 000 h	75 000 h	50 000 h	75 000 h		
Mean value of Y _{2R} [N.mm ⁻¹]	421	420	318	298	172	-		

Table 2. Mean values of Y_{2R} for Ø6.2 and 8.4mm cross-section (Analyses of the 75000h@200°C mockups are still on-going).

The Fig.5 plotting the Y_{2R} parameter as a function of the ageing time shows a strong influence of the temperature that can only be explained by a creep regime of the seal material, assumed to be a dislocation climb of the silver outer lining. On the basis of this assumption, the data collected throughout this very long-term experimental program are analyzed using two different methods.

A time extrapolation method, based on the assumption that the mean value $y^*(T,t)$ of Y_{2R} at a given time and temperature behaves as:

$$y^{*}(T,t) = K(T) - C \log_{10}(t) \{Eq.1\}$$

K and C are two parameters interpolated using experimental data. Thanks to the large database, a statistical data processing can then be made to estimate long-term evolution of the Y_{2R} parameter. This analysis assumed that the scatter of Y_{2R} is a Gaussian normal stochastic variable with a variance independent of hold time. By mean of a probabilistic approach based on Student law, the probability of failure can then be estimated. It can be established that a seal can be safely used provided its Y_{2R} remain superior to a limit value Y_{2Rlim} that will depend on the most severe accidental case the cask has to withstand, such as drop or shocks. Therefore, with proper definition of this limiting value, based on *ad-hoc* studies, seal lifetime can be estimated associated with a failure probability. The second method implemented considers a Larson Miller Parameter (LMP) as a time-temperature equivalence improving the statistical consistence for the long-term extrapolation. Indeed this method allows us to increase significantly the number of available results used to define the long-term seal reliability. In this method the LMP is given by:

$$LMP(T,t) = T(C + \log_{10}(t)) \{Eq.2\}$$

C independent of the temperature is characteristic of the metal and seals designs and is defined such as Eq.3 and Eq.4 match at best the experimental data:

$$Y_{2R}(T,t) = A_1 - B_1 LMP(T,t) \{Eq.3\}$$

The same kind of probabilistic prevision can be implemented with this method. This method also presents the great advantage to enable the prediction for temperatures at which no data are available.

The statistical analyses are described and discussed with much more details in [6]. This paper also presents more predictive results based on the analyses made on 25,000 hours measurements, such maximum seal temperature ensuring a given Y_{2R} after hold time of 100 and 300 years with a probability greater than 0.999999.

FINITE ELEMENTS ANALYSES

Finite Elements Analyses (FEA) are used to understand how the experimental protocol can affects seals behaviors, to support the experimental program and the statistical processing, comforting the validation of the creep mechanisms hypothesis made to implement the statistical methods described above.



Figure 6. 3D FEA model used for preliminary analysis

A preliminary simulation of the experimental setup is done with a 3D FEA model (Fig.6) using the commercial software ANSYS® v6.12. It allows the evaluation of the mockup bending mechanisms and the amplitude of any scalloping effects related to bolts distribution. The numerical model is meshed with quadratic tetrahedrons elements. The gasket itself is described by mean of "gasket elements", which reproduces the seal non-linear characteristic curves (Fig.2 & Fig.6) measured on real seals. To simplify the model, some artifacts are used: symmetry planes between neighbour bolts (only a 15° sector of mockup is represented), materials definitions that do not consider creep or ageing effects nor flange plasticity. The complete experimental protocol with the press loading and unloading processes is described in the model. The calculation is divided into 3 main Load Steps (LS) to reproduce the press assisted bolts tightening procedure. The first step LS₁ correspond to the bottom flange blocked while a load 20% above the Y_2 value is applied on the top flange to obtain MTM contact. During the second step, LS₂, the bolt tension is increased up to the nominal bolts tension of the ageing test. In the final step LS₃, the load on the top flange is removed with the bolt tension maintained.



Figure 7. Flanges deformations (in mm) throughout the experimental protocol.

The Fig.7 reveals that scalloping effect amplitude is negligible, thus compression could be considered as constant along the seal perimeter. For further analyses this effect can be considered as negligible. Three kinds of mockups deformations that can affect the seals ageing test can also be identified: in "X" during LS_1 due to deflection of the flange greater at

the center than at the mockup outer diameter, followed by a return to parallel flanges during LS_2 when the press load and bolts tension are balanced, and finally a deflection in "O" during LS_3 when the bolts on the outer periphery apply a torque leading to a bending in an opposite direction compared to LS1. These simulations allow the evaluation of the seal total deflection during the different steps and the appraisal of the influence of flanges bending mechanisms on real seal deflection in comparison with the target value of 0.9 mm.



Figure 8. Seal deflection (in mm) as a function of operating time.

The Fig.8 reveals that as expected the MTM contact is obtained before the end of LS1, with a load corresponding to 80% of max load. The seal deflection reaches its maximum value of 0.897 mm, 3 μ m below the targeted value of 0.9mm at the end of LS2, with the help of the addition of press loading and bolts tension. This deflection decreases to a 23 μ m gap when the press load is removed at the end of LS3. This analysis provide the followings useful information regarding the flanges bending influence on seals ageing tests: the seals are aged with a deflection smaller than expected and the deflection of the seal during ageing test is 20 μ m smaller than the deflection achieved after the initial compression. These considerations, as the fact that the deflection gap is related to the seal reaction force will decrease with time due to seal relaxation, demonstrate the need of further numerical simulation to evaluate seal long-term behavior.

As any scalloping is negligible, a 2D axisymmetric model of the assembly can be considered to simplify the calculation. In this type of model, the spring complex structure can be described with an equivalent toroidal tube with a wall thickness adjusted to reproduce seals maximum linear loads Y_2 . These major simplifications allow us to improve the materials definition. The elastic-plasticity of spring and inner layer and temperature dependent viscoplasticity for the silver outer lining can be implemented. For all the materials, a Krupkowski law is used to describe the strain hardening:

$$\sigma = K.(\varepsilon_{\text{plastic}} + \varepsilon_0)^n \{\text{Eq.4}\}$$

K, ε_0 and n are derived from tensile tests performed at RT, 100 and 200°C. The reliability of these parameters was demonstrated with a comparison between the experimental tests and FEA simulations. The creep influence in the seal mostly concern the silver outer lining, far much softer that the spring and inner lining. Thickness measurements on crept seals comfort this hypothesis showing that the inner lining and spring creeps are negligible compared to what is observed on the outer lining. For the need of the study an additional and more conservative law is also implemented. The influence of silver creep on seals ageing has been evaluated using FEA, with a strain hardening creep law, detailed in the equation 5.

$$d \varepsilon_{\text{creep}}/dt = C_1 \cdot \sigma^{-C2} \cdot \varepsilon^{-C3} \cdot e^{-C4/T} \cdot \{\text{Eq.5}\}.$$

Two set of coefficients (C_1 to C_4) derived from tensile creep tests are used to frame the seals behavior. The LS of the model include the same 3 steps LS₁ to LS₃ as previously, followed by a heating phase, an ageing period and finally the press loading, the bolts untightening and the press unloading required for the Y_{2R} measurement. During all the phases, the thermal expansion coefficients influence and the thermal dependence of materials characteristic are implemented in the model. The attention is focused on the residual maximum linear load determination in order to compare FEA with experimental results. The three ageing temperature (RT, 100°C and 200°C) for both silver creep laws and nine ageing times are computed.



Figure 9. Comparison of FEA and experimental results for Ø8.4 mm silver layered seals.

The Fig. 9 compares experimental results and their scattering with numerical simulation results. In this Figure, the theoretical values of Y_{2R} are plotted as a function of LMP in order to take into account the influence of temperature. The Figure reveals a good fit with experimental results obtained with FEA silver creep law. These simulations also reveal a small gap between different ageing temperatures, particularly for 200°C, with Y_{2R} values up to 50 N.mm⁻¹ greater than 100°C for a given LMP value. This trend is not observed with the experimental results.

The FEA has also proved to be a very useful tool to counter-check the influence of the measurements made on the mockup regarding their ageing evolution. Indeed, during the experimental protocol the thermal transient (the mockup must be cooled to RT to be placed under the press), the applied loads and the flanges deformations change the constraints applied on seals. Even if the duration of the Y_2 measurement is short compared to the ageing period, the effects of this interruption in the mock-up ageing phase can be questioned. A numerical model is therefore implemented, that reveals a negligible difference for RT ageing tests (Fig.10). For the 200°C continuous ageing, a decrease of the seals spring back of about 5µm corresponding to a loss of 18N.mm⁻¹ (10% loss) is estimated. The comparison between the RT and 200°C cases suggests that the most significant parameter which affects seals

behavior is the thermal expansion of the materials during cooling and heating steps. Nevertheless, it is important to point out that information resulting from interrupted tests underestimate the residual linear load by less than 10% (depending of the ageing temperature) and are conservative from the sealing performance point of view. Even if an Y_{2R} decreasing for interrupted tests reduce slightly silver creep strain rate, seals spring back and residual load are still underestimated by interrupted tests.



Figure 10. Ø8.4mm silver layered seals, mockup at RT and 200°C. Comparison of the unloading curves after 50,000h between continuous ageing tests (continuous lines) and interrupted tests (dash lines) at 25,000h for Y_{2R} evaluation.

DISCUSSION

The statistical analyses based on the 75,000 hours experimental program confirm the ability of HELICOFLEX[®] seals to preserve a significant residual maximum linear load over a century, the increased database size used for the statistical analysis improve significantly the forecast reliability. The evaluation of seals spring back for a given residual load, which allows the prediction of the maximum temperature imposed to preserve a given spring back (Table 3) has however to be considered with caution. Indeed the suggested method is quite too pessimistic as it considers a constant linear load for sealing performances losses, when experiences demonstrate a significant decrease of this key parameter.

Regarding the numerical simulations, the results of the FEA provided very useful information to support the statistical analyses. The understanding of the mockup bending deformation mechanisms and the estimates of the large deflections provide valuable inputs for the creep simulations. Indeed due to the precision required with this type of simulation, the hypothesis of perfectly rigid and planar flanges is not relevant. The combination of these two numerical approaches allows the implementation of a reliable 2D axisymetric model of seals and mockup. Even if this model strongly relies for ageing studies on the silver creep laws precisions, it provides with the use of two different laws a reliable framing of seals behaviors. The comparison of FEA results with experimental data demonstrates an acceptable concordance of initial silver creep law from the Y_{2R} estimation point of view, bolstering the FEA model reliability. The spring-back analysis is far more tedious with the 2D axisymetric model, which reliability has to be demonstrated.

CONCLUSION: OUTLOOKS AND PERSPECTIVES

The experimental campaign is pursed up to 100,000 hours (125,000 hours for RT mockups), with additional data that will increase the forecast reliability. As part of this program, several actions will be launched to improve the data processing and analyses of this rather unique experimental campaign.

As suggested by some abnormal results observed on the preliminary analysis of the 75,000h/200°C mockups, the time-temperature method should be adapted to describe more precisely the creep deformation mechanisms. Indeed, as observed during the ageing tests, seal stress is decreasing due to creep-relaxation as their own deformation affects their residual linear load. A promising solution might be to modify the LMP analysis in order to take into account the RT ageing results and to predict horizontal asymptote of residual maximum linear load.

The improvement of the silver creep law is also a key point for the precision of the ageing numerical simulations. A specific effort is engaged based on further creep tests results to improve our understanding of this mechanism. Metallurgy analyses of aged seals, machined in the equatorial and poloidal plane are also planned on RT/100,000h and 200°C/75,000h mockups. The measurement of the outer/inner lining thickness and hardness evolution of the material, added to a characterization of the aged metallic structures compared to the one of unused seals, will provide very interesting inputs to better understand the creep regime. The measurement of the penetration of the spring wires inside the inner lining will also give elements to benchmark and enhance the numerical model.

Finally as discussed above, the 2D axisymetric model of the seal, with a spring represented by an equivalent pipe, even if it give realistic results (benchmarked by experimental data), has to be assessed regarding the long-term determination of the seal spring-back. The development of a 3D high fidelity model of the complex HELICOFLEX[®] seal structure is on its way to improve the spring model precision.

ACKNOWLEDGEMENT

These works were performed in the joint research program among CEA, Technetics Group France, GNS and CRIEPI.

REFERENCES

[1] Yamamoto, T., 2001. Dual-purpose metal cask integrity after long-term interim storage (activities for the verification test of a dual-purpose metal cask). In: Proceedings of the PATRAM 2001, Chicago, IL, USA, September 7–9.

[2] Droste, B., 1988. *Safety evaluation of dry spent fuel storage casks*. In: Soviet-West German Seminar, USSR, Leningrad, June 27–July 1.

Benda, B.J., Langland, R.T., 1980. *Methods for Evaluating the Leak Tightness of Spent Fuel Container Closures*. Lawrence Livermore Lab.

[3] Rouaud, C., Lefrancois, M., Caplain, P., 1999. *Static sealing: technologies of the future*. In: Proceedings of the First International Conference on Sealing Technology and Plant Leakage Reduction, Charlotte, NC, USA, November 15–19.

[4] Birembaut, Y., Ledauphin, T., Vignaud, J.C., 2001. *Mechanical and sealing performances of gaskets specifically manufactured for use in metal to metal conditions*. In: PVP Conference 2001.

[5] Benda, B.J., Langland, R.T., 1980. Methods for Evaluating the Leak Tightness of Spent Fuel Container Closures. Lawrence Livermore Lab.

[6] H. Sassoulas, L. Morice, P. Caplain, C. Rouaud, L. Mirabel, F. Beal, *Ageing of metallic gaskets for spent fuel casks: Century-long life forecast from 25,000-h-long experiments*. In Nuclear Engineering and Design 236 (2006), p 2411-2417.

[7] Larson, F.R., Miller, J., 1954. A time-temperature relationship for rupture and creep stresses. Trans. ASME 174, 765–775.