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LONG-TERM PERFORMANCE OF METAL SEALS FOR TRANSPORT AND STORAGE CASKS

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ABSTRACT

Dual purpose casks for the transportation and storage of spent nuclear fuel and other radioactive materials require very high leak-tightness of lid closure systems under accident conditions as well as in the long-term to prevent activity release. For that purpose metal seals of specific types with an inner helical spring and outer metal liners are widely used and have shown their excellent performance if certain quality assurance requirements for fabrication and assembling are satisfied. Well defined surface roughness, clean and dry inert conditions are therefore essential. No seal failure in a loaded cask happened under these conditions until today.

Nevertheless, the considered and licensed operation period is limited and all safety assessments have been performed and approved for this period of time which is 40 years in Germany so far. But in the meantime longer storage periods might be necessary for the future and therefore additional material data will be required. BAM is involved in the qualification and evaluation procedures of those seals from the early beginning. Because long-term tests are always time consuming BAM has early decided to perform additional tests with specific test seal configurations to gain a better understanding of the long-term behavior with regard to seal pressure force, leakage rate and useable resilience which is safety relevant mainly in case of accidental mechanical loads inside a storage facility or during a subsequent transport. Main test parameters are the material of the outer seal jacket (silver or aluminum) and the temperature. This paper presents the BAM test program including an innovative test mock-up and most recent test results. Based on these data extrapolation models to extended time periods are discussed, and also future plans to continue tests and to investigate seal behavior for additional test parameters are explained.

INTRODUCTION

At the end of 2012 nearly 1.000 metal casks have been in dry storage of spent nuclear fuel from German nuclear power reactors and high active waste from reprocessing and the number of casks still increases. The German concept for the dry interim storage of spent nuclear fuel is defined by the *Guidelines for Dry Cask Storage of Spent Fuel and Heat-generating Waste (revised version of 29.11.2012)* issued by the German Waste Management Commission (ESK) and requires thick-walled dual purpose casks for transportation and interim storage with a continuously monitored double barrier lid system. All casks have to demonstrate their accident safe design during transportation, handling, and storage operations concerning the main safety goals: safe enclosure, shielding, subcriticality and decay heat removal.

The applied and licensed interim storage period for all interim storage facilities whether centralized or on-site is limited to 40 years so far mainly due to administrative reasons. All safety demonstrations have to be performed allowing for that period of time under consideration of ageing and degradation mechanisms for materials and components concerning the ambient operation conditions of any particular storage facility.

With regard to the long-term safe enclosure of the radioactive inventory metal seals of the Helicoflex® type HN 200 with outer aluminum or silver jackets manufactured by Technetics Group France are in use with the casks. Up to now several thousand of those seals have demonstrated their expected proper function with all bolted cask lid systems for up to 30 years of operation without any failure.

As mentioned all storage licenses are limited to 40 years so far because it was expected to have a final deep geological repository available after that period of time. In the meantime the German siting procedure focusing on the Gorleben salt dome so far has been changed for political reasons and a new law on repository selection has been confirmed by the German Bundestag and the Bundesrat representing the 16 Federal states by mid of 2013. The scheduled timeframe expects a decision about the location for the repository not until 2031 followed by exploration and construction of that facility. In the unlikely event of no further delays by lawsuits during the various stages of that procedure a repository might be available by around 2045 at the earliest. At that time all current storage licenses are going to expire or have already been expired and it’s obvious that extensions for some more decades will become necessary during the repository operation period which is expected to last at least 30 years until all spent fuel and high active waste will be disposed of.

In case of applications for lifetime extensions for interim storage facilities beyond 40 years additional data will be required to demonstrate the long-term safe enclosure including the proper seal function as an essential aspect. For that reason BAM has been starting long-term investigation programs and laboratory tests with representative seals under various temperatures looking for potential changes in mechanical behavior and seal function.

SEAL TEST CONFIGURATION

Figure 1 shows an appropriate schematic representation of a casks common lid and sealing system. The presented metal seals actually have the main sealing function and have to fulfill the high requirements on the leakage rate. The elastomeric seals are auxiliary seals to create a cavity which is necessary to measure and validate the leakage rate of the metal seals after installation.

The bolts fixing the lids onto the cask body have to be provided with a suitable pre-tension for compressing the metal seals to their proper assembly situation. The lids are equipped with grooves to carry the seals. In case of metal seals there has to be a specific groove depth, which corresponds with the given operation point and optimal pressure force of the seals. By screwing the primary or the secondary lid to the contact position onto the cask body the correct seal compression is given. For the safe long-term operation of such cask closure systems it is important to understand time dependent degradation mechanisms like loss of seal pressure forces and screw pre-stresses due to creeping and relaxation effects. For that reason systematic investigations of seal function under consideration of installation conditions, material properties, operation temperatures and periods of time gain necessary information for further long-term safety assessments.

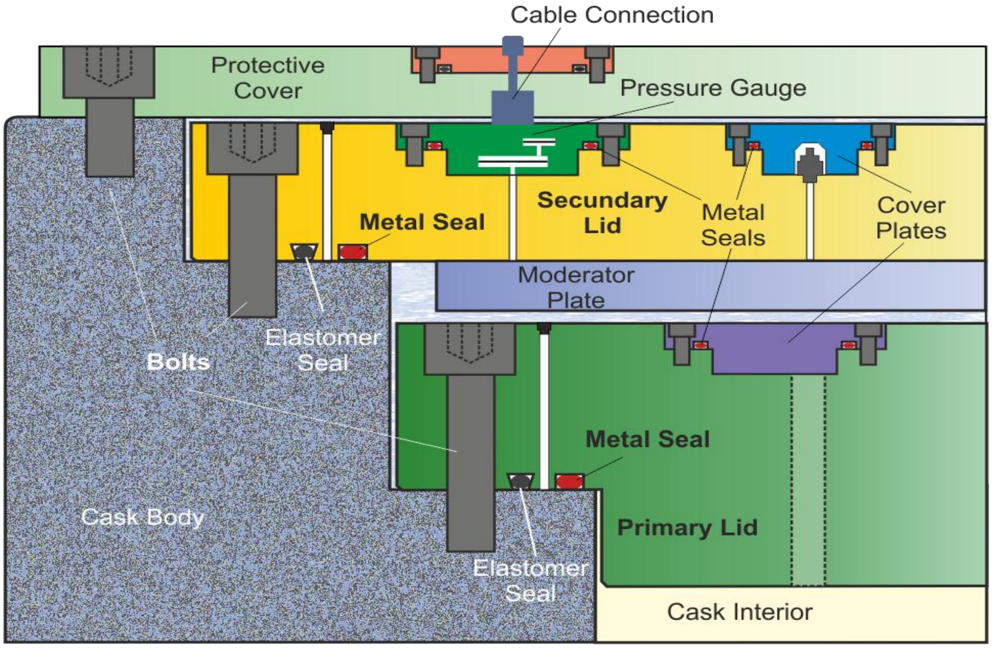


Figure 1. Lid area and sealing system of a transport and storage cask

Metal seals of the Helicoflex® type as illustrated in Figure 2, consist of an inner helical metal spring and two metal jackets. The outer metal jacket is made of flexible materials as aluminum (Al-seals) or silver (Ag-seals) to achieve and maintain tight contact between seal and lid or cask body surfaces. The main criteria for sufficient leak tightness of such seals after assembly is the standard helium leakage rate $Q_{He/St} < 10^{-8}$ Pa·m^3/s. This value has to be demonstrated for each metal seal used in casks and it also sets up the evaluation level for experimental investigations.

In the BAM test series seals with a smaller overall diameter compared to full-scale cask lid seal diameters are used to allow appropriate dimensions of the test setup. However, its cross section diameter of about 10 mm as well as materials and dimensions of spring and jackets are identical to the ones of cask seals to yield representative test results.

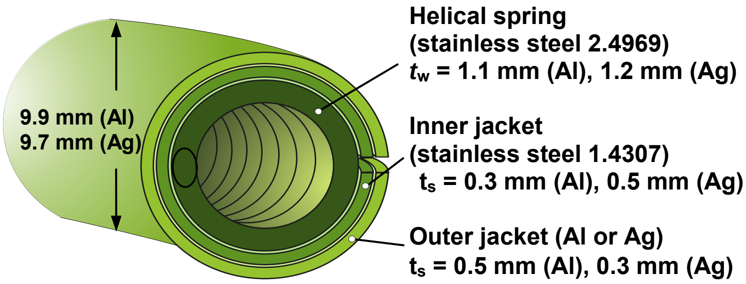


Figure 2. Helicoflex® seal type applied in test series typical for applications in dry storage casks

BAM has developed special test flange systems for its specific investigation program concerning long-term test conditions. In real cask situation, the seal deformation at operation point is given by the depth of the seal groove, which assures well defined geometric deformation after screw tightening until cask body and lid come in contact. To perform representative assembling conditions test flanges for investigations under static long-term conditions are also equipped with according seal groove geometry. The test flange systems are screwed with installed seals and can be stored outside a testing machine over longer time periods, e. g. at room temperature or in a heating chamber at higher temperatures. Figure 3 illustrates the BAM test flange system including the screw connection and groove geometry principle for seal compression up to the operating point at the flange surfaces contact position.

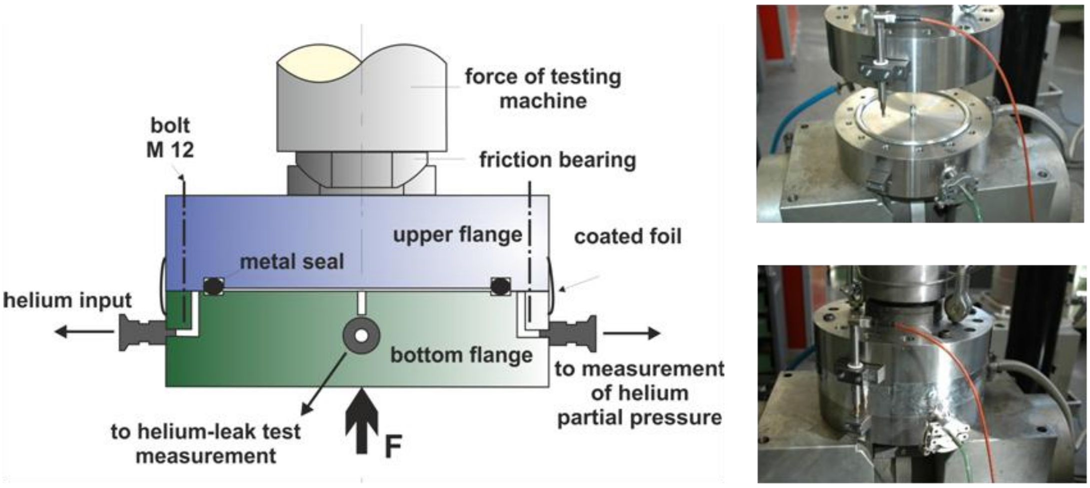
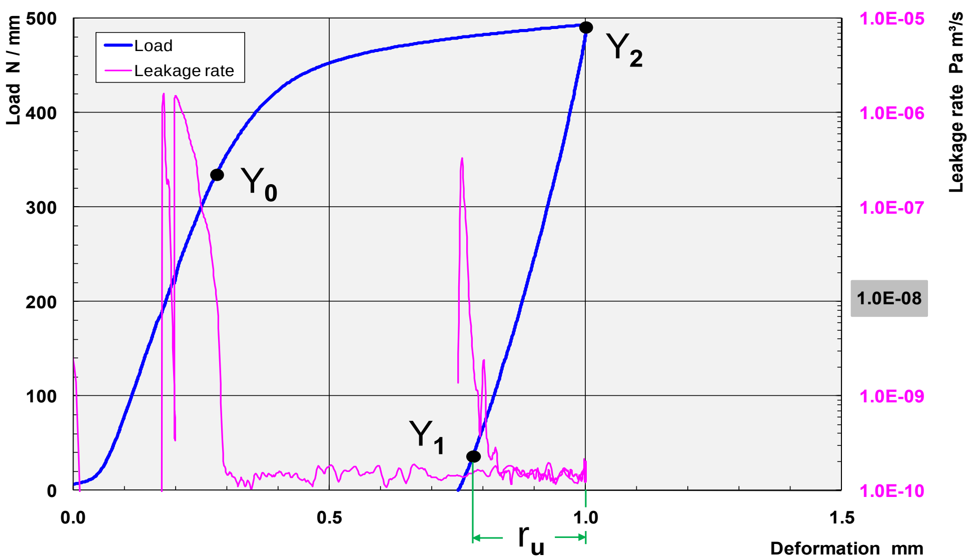


Figure 3. BAM test flange system. Groove depth designed for flange contact position. Right: Open and closed flange configuration with masked flange gap prepared for helium leakage test.



- $Q_{He/St} \leq 10^{-8}$ Pa m^3/s
- Y_0 = Initial achievement of $Q_{He/St}$ during compression
- Y_1 = Exceeding $Q_{He/St}$ during load relieving
- Y_2 = Operation point. Deformation path according to manufacturer specification
- r_u = useable resilience up to exceeding $Q_{He/St}$

Figure 4. Characteristic load - deformation relationship of a Helicoflex® seal with outer silver jacket with respect to standard helium leakage rate

The characteristic mechanical behavior of Helicoflex® seals can be illustrated by their load-deformation relationship during compression and relieving procedures (Figure 4).

The load-deformation curve including the characteristic points Y_0, Y_1 and Y_2 is given with respect to the standard helium leakage rate during compression and relieving process. The typical load-deformation course in Figure 4 results from a 9.7 mm Helicoflex® HN 200 seal with outer silver jacket.

The distance between Y_2 and Y_1 in terms of the deformation represents the seal's ability of elastic recovery and is denoted as "useable resilience" r_u, which is the deformation path between the operation point Y_2 and the point Y_1, where $Q_{He/St} = 10^{-8}$ Pa·m³/s is exceeded during load relieving. In case of real casks, an exceeding of $Q_{He/St}$ may be caused either by mechanical loads under accident scenarios or by reduction of the restoring seal force (F_r) due to time depending creeping processes of seals and/or screws.

Maximum temperatures reach about 110°C in the seal area of cask lid systems at the beginning of storage depending on cask design and spent fuel decay heat. Knowing that higher temperatures can accelerate ageing mechanisms, BAM decided to perform the tests at three different temperature levels: 20°C (ambient temperature), 100°C and 150°C for the time being. For those test series, Ag- and Al-seals were applied. Table 1 shows the test parameters at a glance.

Table 1. Metal seal test configurations (Helicoflex® HN 200)

Seal temperature	***20°C (ambient temp.)***	***100°C***	***150°C***
Seal types	Al + Ag	Al + Ag	Al + Ag
Beginning of test series	02/2009	11/2010	02/2009

At the beginning, each seal is assembled and compressed in the test flange hold by a testing machine. The seal groove depth is equivalent to the required compression path and the operation point Y_2 is reached upon contact of the flange surfaces. To identify the initial point Y_1, a first load relieving was done. After recurrence of compression, the test flanges were screwed together to keep the seals at the operation point after removal out of the testing machine for further long-term storage periods. Heating chambers were used when tempering the flanges at 100°C and 150°C.

So far, the following test configurations are under investigation by BAM: one Al-seal and one Ag-seal each at 20°C and 150°C since February 2009, one Al-seal and one Ag-seal at 100°C since November 2010. After definite time periods, the seal behavior is investigated periodically as follows:

Each test flange is reinstalled in the testing machine and compressed applying loads higher than the initial operation point force. Then all flange screws are loosened and removed completely. Now, test flange and seal are relieved by continuous reduction of the testing machine pressure force. The restoring seal force (F_r) at the operating point is determined when the flange surfaces start to lift off. By pressure force reduction to point Y_1, the remaining useable resilience r_u is determined. Figure 5 shows test results of an Al-seal and an Ag-seal for holding times of up to 48 months at 150°C. It can be easily found that the r_u reduction is much more significant in case of Al-seals.

A permanent seal deformation occurs already in the course of the initial compression. Depending on holding time and temperature, an increase of the plastic deformation mainly of the outer jacket occurs due to creeping processes (Figure 5, right) and accordingly the torus diameter reduces. The groove geometry and thus the deformation path remain constant. Therefore, the seal relaxation correlates to the reduction of the jacket thickness. This, in turn, causes a reduction of F_r. With reduction of F_r, r_u generally reduces as well. After comparatively short holding times (during BAM test series at 150°C approximately one week) the test seals, subject to compliance with $Q_{He/St}$, could be relieved nearly completely. Once this condition is met, r_u reduces in the same ratio as F_r.

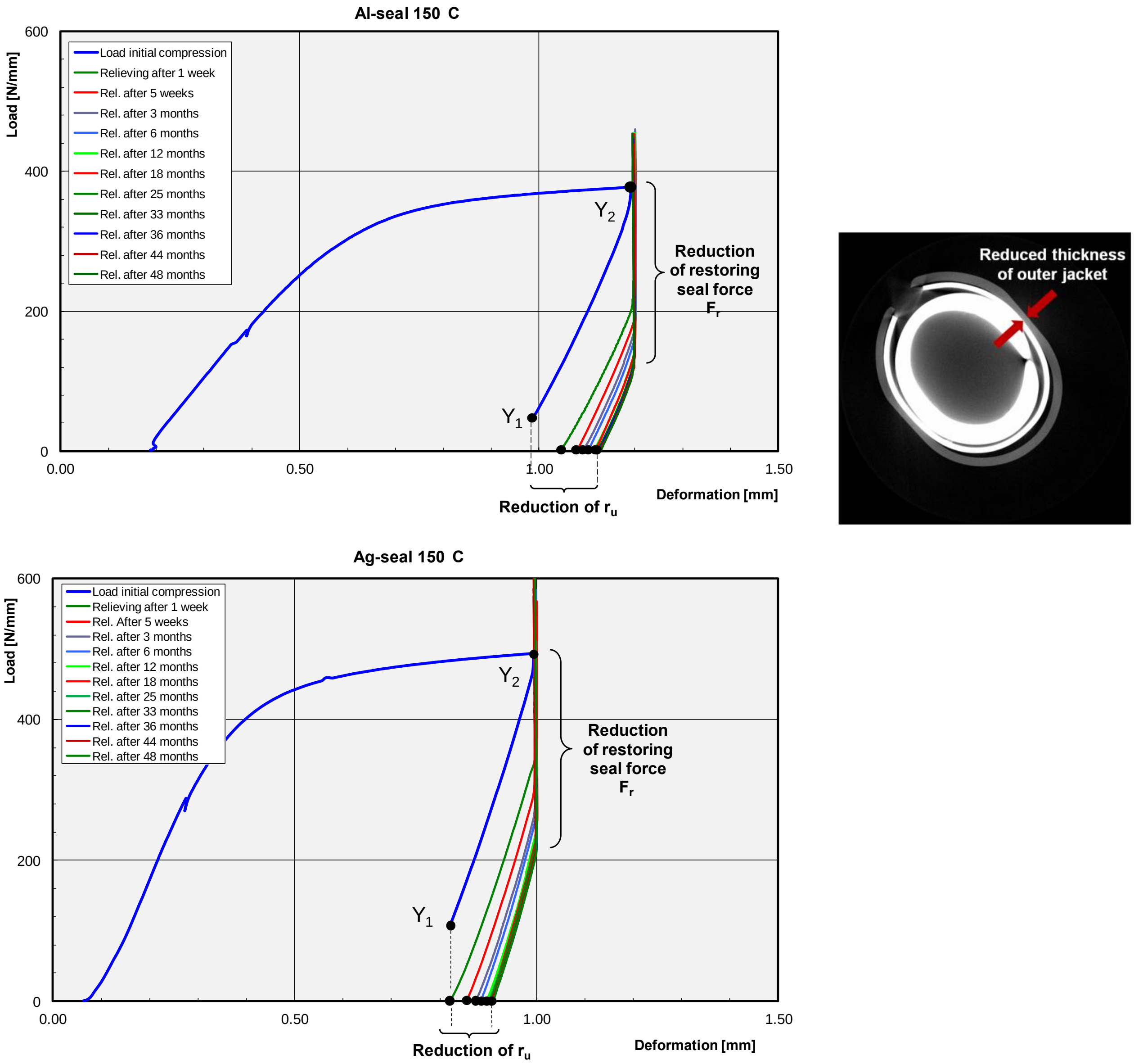


Figure 5. Reduction of restoring seal force (F_r = Load) and useable resilience r_u depending on time and temperature due to creeping
Right: Computer tomography scan of an Al-seal after 3 months at 150°C

TEST RESULTS CONCERNING SEAL FORCE

In Figure 6 the reduction of F_r depending on holding time and temperature is plotted over a logarithmic time scale, which illustrates a proper linear correlation and allows extrapolating very easily to longer time periods. Table 2 shows the percentage of restoring seal force left after certain periods of time depending on seal jacket material and temperature.

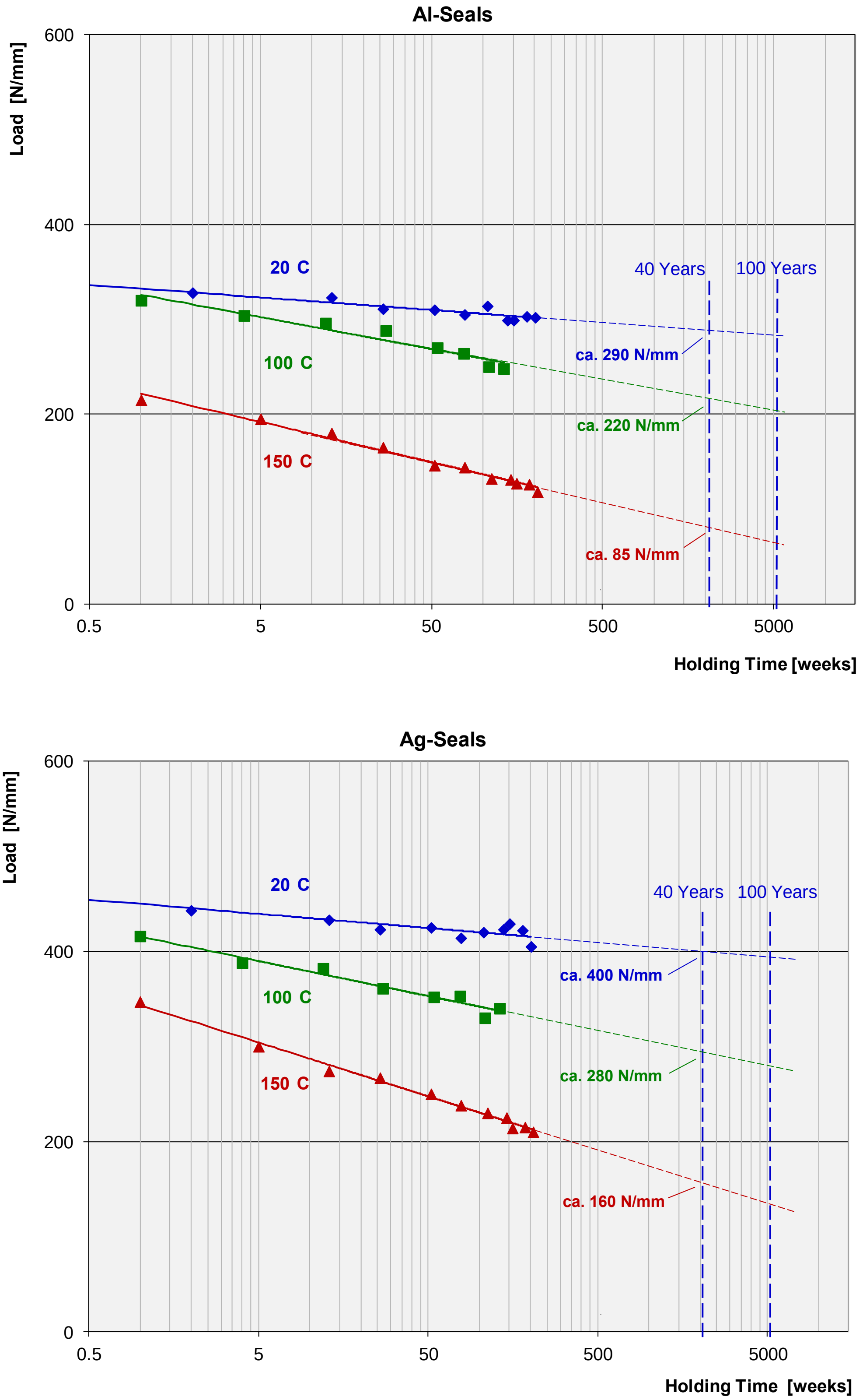


Figure 6. Reduction of restoring seal force (F_r = Load) depending on holding time and temperature for current test periods (up to 48 months) and extrapolation up to 40 and 100 years (dashed lines) in logarithmic scaling

Table 2. Percentage of restoring seal force left after certain periods of time depending on seal jacket material and temperature

Seal Type: Al Helicoflex® HN 200	Remaining restoring seal force F_r after time (percentage)		
	12 months	30 months	48 months
20°C (ambient temp.)	80%	77%	77%
100°C	71%	65%	---
150°C	38%	34%	31%

Seal Type: Ag Helicoflex® HN 200	Remaining restoring seal force F_r after time (percentage)		
	12 months	30 months	48 months
20°C (ambient temp.)	84%	83%	80%
100°C	72%	69%	---
150°C	51%	46%	42%

TEST RESULTS CONCERNING USEABLE RESILIENCE

The reduction of the useable resilience r_u is more significant for the evaluation of remaining leak tightness if an aged seal system is e. g. exposed to severe cask accident scenarios with significant mechanical effects on the mounted seal. Figure 7 shows test results at 20°C, 100°C and 150°C for the current test periods of up to 48 months. Table 3 contains the percentage of useable resilience left after certain periods of time depending on seal jacket material and temperature. Only in case of Ag-seals at 20°C no linear interpolation works because the Y_1 load (compare Figure 4) was not constant during the first two years. An applicable extrapolation of r_u should be only made, when the Y_1 load has approached constant values. In case of Ag-seals at 20°C this applies only for the last 4 measurements. In general it is essential to confirm the linear extrapolation approach by continuation of the running test series.

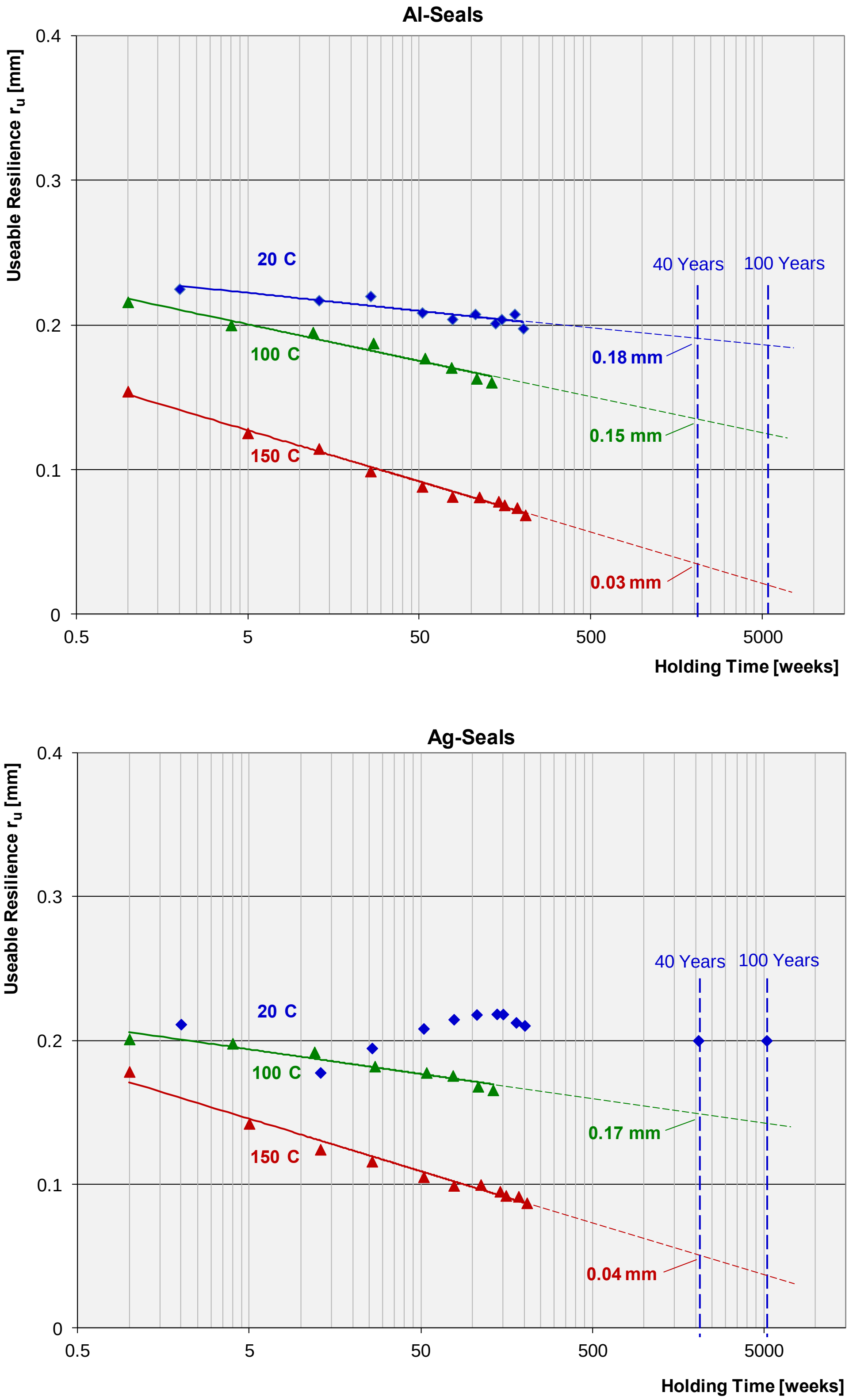


Figure 7. Reduction of useable resilience r_u depending on holding time and temperature for current test periods of up to 48 months and extrapolation up to 40 and 100 years (dashed lines) in logarithmic scaling

Table 3. Percentage of useable resilience left after certain periods of time depending on seal jacket material and temperature

Seal Type: Al Helicoflex® HN 200	Remaining useable resilience r_u after time (percentage)		
	12 months	30 months	48 months
20°C (ambient temp.)	95%	91%	90%
100°C	80%	73%	---
150°C	55%	49%	43%

Seal Type: Ag Helicoflex® HN 200	Remaining useable resilience r_u after time (percentage)		
	12 months	30 months	48 months
20°C (ambient temp.)	94%	99%	95%
100°C	89%	83%	---
150°C	58%	53%	48%

The remaining seal force values shown in Table 2 relate to the seal force at point Y_2, which has been determined during the initial seal compression. In contrast, for r_u (Table 3) the reference value is the one measured after one week holding time. The reason for choosing different reference values is because the remaining pressure force at point Y_1 to exceed the required leakage rate $Q_{He/St}$ measured during the initial relieving has not approached a constant level yet (for example see Figure 5). Only after one week the $\mathbf{Y_1}$ force measured was constantly nearly zero N/mm. As already mentioned the Y_1 values of the Ag-seal at 20°C are not located on a constant seal force level for almost 2 years. On the contrary, after a first decrease as result of the 3 months measurement an increase could be observed afterwards before a nearly constant Y_1 seal force value nearly equal zero could be observed after 2 years holding time. This fact explains the increasing percentage values for Ag-seals at 20°C in Table 3.

ANALYTICAL APPROACHES

Recently performed investigations take account of the Larson-Miller relationship [1], which was developed with regard to the long-term performance of metallic materials under consideration of time and temperature and also widely used for metal seals, see [3], [4] and [5]. Basically, Larson and Miller have used the time and temperature relationship to describe rupture and creep stresses by the basic correlation

$$LMP = f\{T \cdot (C + ln(t))\}$$

with time *t* in hours, temperature *T* in Kelvin and a material constant *C*.

Investigations described in [1] and [2] have shown that at a constant material-specific parameter C, the LMP can be generated combining different times (t) and temperatures (T). The parameter *C* can be derived from experimental data of several iso-static test series at variable temperatures with an equal constant load.

This time-temperature-equivalence parameter LMP as defined by Eq. (1) is considered to describe our so far measured test results of the restoring seal force F_r for Ag-seals. For the material parameter *C* a value of 11 has been used as described by [3]. However, in application with metal seals also other values for *C* have been reported, e. g. *C* = 14 in [5].

Figure 8 illustrates the relationship between our measured restoring seal force F_r and the LMP (except the initial load) concerning temperature and time.

$$LMP\ (T,\ t) = T \cdot (C + log_{10}\ (t)) \qquad (1)$$

with T in Kelvin, t in hours

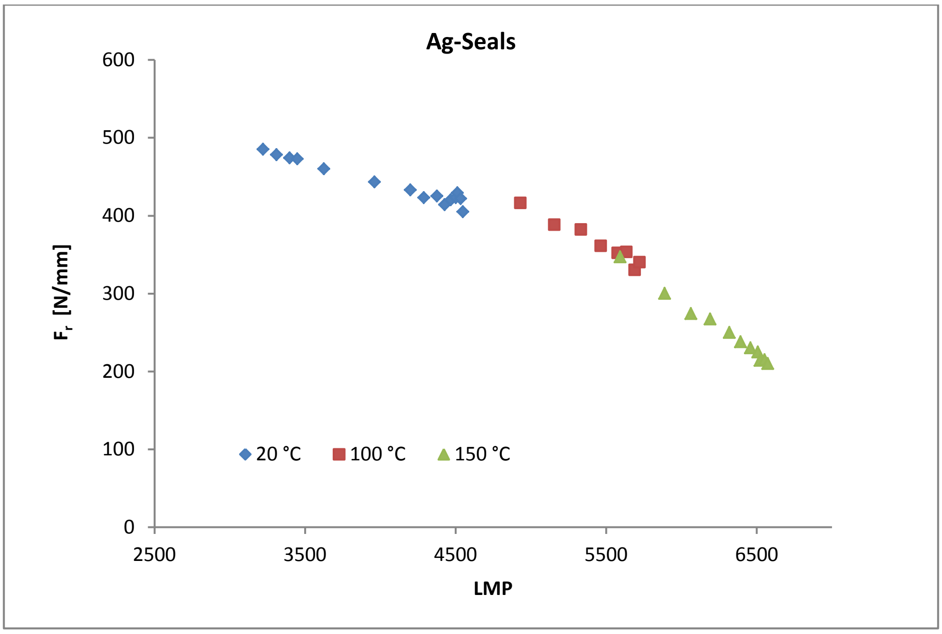


Figure 8. Correlation between measured restoring seal force F_r and LMP

Due to the fact that for a given material the Larson-Miller-Parameter should show a correlation between temperature and time, it is possible to calculate the corresponding time t_2 for a determined LMP value and a given temperature T_2 by using Eq. (2).

$$LMP = T_1 \cdot (C+log_{10}(t_1)) = T_2 \cdot (C+log_{10}(t_2)) \qquad (2)$$

Applying the material parameter *C* =11 t_2 values were calculated exemplarily for representative temperatures. The results in Tab. 4 show differences between the calculated time t_2 and the real testing period for approximately the same (measured) restoring seal force. In conclusion, it is to note that a first evaluation of our experimental data with the method of time-temperature equivalence (here LMP with *C*=11) does not provide useful results so far.

Table 4. Calculated time t_2 for T_2 compared with measured data

LMP	F_r [N/mm]	T_1 [°C]	t_1 [h]	T_2 [°C]	t_2 [h]
5724	340	100	22176	150	340
	347			150	168
4933	416	100	168	20	685986
	414			20	13104
4429	414	20	13104	100	7.5
	416			100	168
4290	423	20	4368	150	0.138
	495			150	0

calculated
measured

Further investigations are required, in particular for additional temperatures to gain more test data and to determine the characteristics of the material parameter *C*. Therefore, test data for the restoring seal force F_r were chosen to determine whether there is a linear relationship between time and temperature at several different iso-static linear loads. The corresponding material parameter *C* should be constant according to its approach.

Derived from Eq. (1) and with an interpolation function from the experimental F_r data (see Figure 6) time values in logarithmical scale and the temperature in inverse performance are shown in Fig. 9 and illustrate different constant values for F_r. These results indicate that the material parameter C depends on the restoring seal force F_r and the temperature T and seems not to be an independent material parameter for the investigated type of Ag-seals.

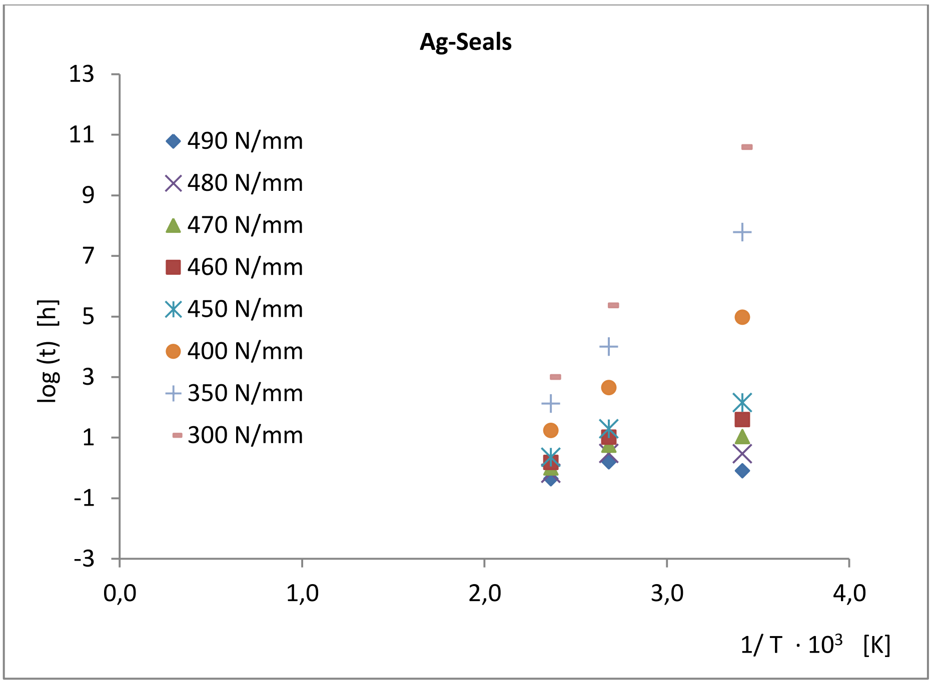


Figure 9. Time and temperature relationship for iso-static loads

CONCLUSIONS AND OUTLOOK

Since metal seals of the Helicoflex® type are an essential component of casks for long-term safe storage of spent fuel and high active radioactive waste and storage periods of more than 40 years are expected for the future, BAM has started additional investigations on the long-term behavior of such seals. Main criteria for long-term performance is the leak tightness of the screwed lid seal system under relevant stressors like creeping, ageing or dynamic forces and deformations in case of severe mechanical accident scenarios.

Investigations presented in this paper focus on time and temperature depending decrease of restoring seal force and useable resilience. BAM test results exist for time periods of up to 48 month at three different temperatures (20°C, 100°C and 150°C) so far and show good linear correlations over time in a logarithmic scaling. With that an extrapolation for longer periods of time, e. g. the relevant storage periods, is easily possible but has to be verified by continuation of the tests. Tests at additional temperatures (75°C and 125°C) are designated to be started this year.

Other aspects of the investigation program consider effects on the leak tightness of $Q_{He/St}$. Test results have demonstrated that the specified initial leakage rate of 10^{-8} $Pa \cdot m^3/s$ stays in force until nearly complete load relieve due to creeping and plasticization of the outer seal jacket material. Furthermore, analytical approaches, e. g. the Larson-Miller relationship, describing time and temperature dependent seal behavior are of special interest to gain reliable predictions and to establish accelerating test configurations. Their overall aim is to get reliable predictions of the long-term behavior of metal seals used in lid systems of dry interim storage casks. In order to get such results by using the Larson-Miller approach additional investigations are in progress.

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