Test Results and Analyses in Terms of Aging Mechanisms of Metal Seals in Casks for Dry Storage of SNF – 14255

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ABSTRACT

In Germany the management of spent nuclear fuel is being pursued by the concept of dry interim storage in dual purpose metal casks prior to final disposal and corresponding storage licenses have been granted for a storage period of up to 40 years. Cause of significant delays in the national repository siting procedure, extended storage periods will become necessary in the future. Due to that fact interim storage of spent nuclear fuel in dual purpose casks must be safe for longer periods of time than the initially evaluated and licensed ones. A major safety goal for dual purpose casks is to ensure high quality of leak-tightness of lid closure sealing systems in view of the safe enclosure of the radioactive inventory. This can be achieved by using double jacket metal seals with a ductile outer liner and a close-wound helical spring in the core.

For the qualification and evaluation procedures of metal seals in Germany the Federal Institute for Materials Research and Testing (BAM) has been involved in from the early beginning. Regarding this subject BAM investigates the long-term behavior of metal seals under the influence of temperature and time using experimental data and analytical approaches. Experimental analyses indicate a time and temperature dependent decrease of the restoring seal force and the useful resilience, while the quality of leak-tightness is not affected. Hence, there is a fundamental interest of describing time and temperature dependency to gain predictable values and to achieve results within significant shorter periods of time and in addition, numerical analyses are intended for the future.

This paper gives a brief overview about the sealing principle, the test program and test results of metal seals of the HELICOFLEX® HN200 type. The effects are discussed regarding two different materials for the outer liner, aluminum and silver. Furthermore, analytical approaches of the evaluation of the test results under consideration of time-temperature relationships are discussed in detail with the main focus on the *Larson-Miller-Parameter* $LMP = T \cdot (C + \log(t))$, with *time t* in hours, *temperature T* in Kelvin and a constant material parameter *C*. The paper describes the derivation of parameter *C* from test results and the applicability of the *Larson-Miller-Parameter* for those metal seals in principle.

Overall a constant material parameter *C* based on the *Larson and Miller* method could not be found for the investigated type of seals. Therefore, a useful approach comparable to the *Larson-Miller-Parameter* is given.

INTRODUCTION

This paper presents recently performed investigations taking into account the time-temperature relationship based on the *Larson-Miller-Parameter* [1] relating to metal seals of the HELICOFLEX® HN200 type. Experimental test results reveal that the restoring seal force F_R decreases over time and with increasing temperature, so far without failing the specific leakage-rate. So it could be assumed that mainly time and temperature cause the decrease of F_R and both conditions can mutually interchange. On the basis of this interdependency there is a major interest to find a correlation between short- and long-term tests and to get predictable information about the long-term behavior based on short-term test data. Thus it has been analyzed how far the time-temperature relationship based on *Larson and Miller* [1] can be adapted to the remaining seal force; see also [2] and [3].

Metal seals are widely used for ambitious applications including like dual purpose casks for dry interim storage of spent nuclear fuel and they are usually of the HELICOFLEX® type as illustrated in Fig. 1.
Such seals consist of an inner helical metal spring and two metal jackets. The outer metal jacket is made of flexible aluminum (Al-seals) or silver (Ag-seals) to achieve and maintain tight contact between seal and lid or cask body surfaces. In the BAM test series seals with a smaller overall diameter compared to full-scale cask lid seal diameters are used to allow appropriable 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 dual purpose cask seals to yield representative test results.

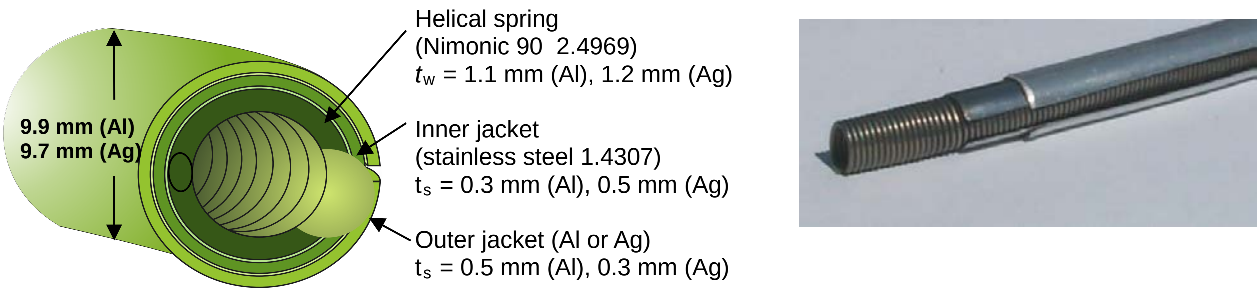

Fig.1. HELICOFLEX® seal type applied in test series typical for applications in dry storage casks.

BAM has developed special test flange systems for its specific test program concerning long-term test conditions (restoring seal force, useful resilience and leak-tightness) which is described in detail in previous papers e.g. in [5] and [6]. In the present paper the main focus is on the restoring seal force. 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 characteristic mechanical behavior of HELICOFLEX® seals can be illustrated by their load-deformation relationship during compression and decompression procedures (see Fig. 2). Depending on holding time and test temperature, the restoring seal force F_R decreases. This can be seen during the decompression of the flange system for both type of seals, however the percentage decline of the restoring seal force for Al-seals is higher than for Ag-seals. Furthermore, Fig. 2. shows exemplarily the course of the restoring seal force during decompression after a holding time of about 40.000 hours of a 9.9 mm HELICOFLEX® seal with outer aluminum jacket at a test temperature of 20°C. The remaining measured seal force F_R was down to about 280 N/mm from initially 390 N/mm. The particular elasticity of the test flanges were taken into account when determining the remaining seal force F_R values. Therefore the characteristic behavior for compression and decompression of each flange system were determined without seals, in particular in the range of flange contact.

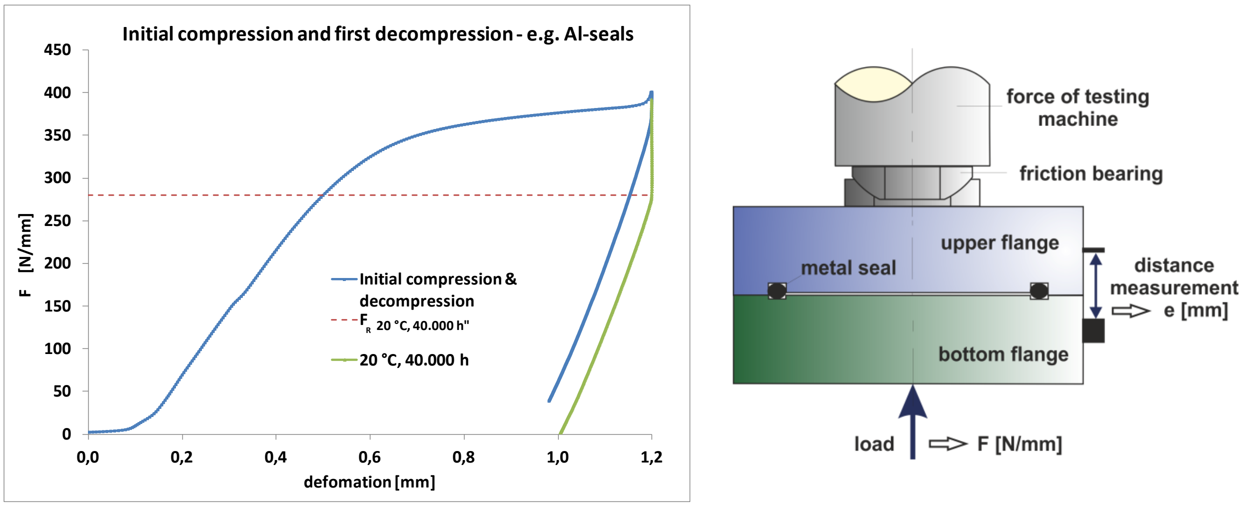

Fig. 2. Characteristic curve of initial compression and decompression of a HELICOFLEX® Al-seal and exemplarily restoring seal force at 20°C after 40.000h (left), BAM test flange system (right)

ANALYSIS OF TEST RESULTS

The following diagrams (Fig. 3. and Fig. 4.) illustrate the measured test data concerning the restoring seal force over time in logarithmic scaling for both seal types and depending on the test temperatures. These results reveal that the restoring seal force F_R decreases over time and with increasing temperature. All seals were compressed initially at room temperature and stored in flange systems during the testing time at room temperature (20°C) or in a heating chamber at higher temperatures (100°C and 150°C). The measurements of the restoring seal force were performed for all seals in a testing machine at room temperature. Since it takes several hours to cool the flanges from the heating chamber temperature to room temperature, the first measurements of the seals with higher testing temperatures were performed after one week. In contrast, seals which are stored at room temperature. Therefore they could be investigated already at much shorter times.

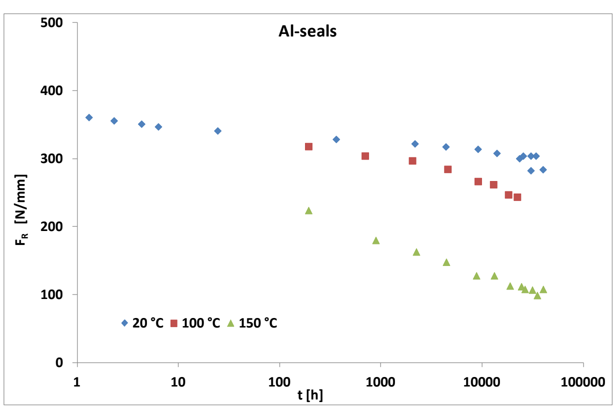

Fig. 3. Restoring seal force F_R over time log(t) for Al-seals

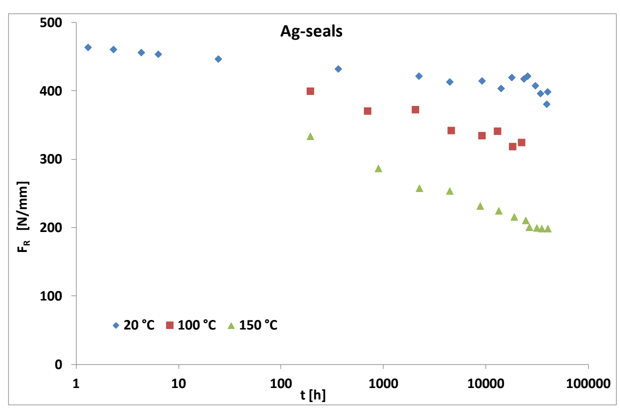

Fig. 4. Restoring seal force F_R over time log(t) for Ag-seals

Application of the Larson-Miller-Parameter

In 1952 *Larson and Miller* [1] applied the time-temperature relation expressed by the parameter *LMP = T · (C + log(t))*, with time *t* in hours, temperature *T* in Kelvin and a material parameter *C* for rupture and creep data of metallic materials. *Larson and Miller* purposed that creep rate could adequately be described by the *Arrhenius* equation. Investigations in [1] and [4] have shown that with a material parameter *C*, the *LMP* can be generated combining various time periods and temperatures for the use of short-term tests to determine long-term data under consideration of time and temperature. The parameter *C* can be derived from experimental data of several iso-stress tests at various temperatures.

In a first step the current test data (time- and temperature values) concerning the restoring seal forces for Al- and Ag-seals were used to calculate the time-temperature parameter *LMP* defined by Eq. 1. For the material parameter *C* exemplary values from literature ([2], [3]) 11 and 14 were used to check ability for our test data. Anyway, these *C*-values were not verified in detail. Other *C*-values in application with metals are used depending on the points of interest (e.g. *C* = 20 for creep and rupture processes [1]).

The relationships between restoring seal force F_R and *LMP* (except initial load) for the measured data depending on temperature and time and using *C*-values 11 and 14 are shown in Fig. 5. and Fig. 6., see below. Initial loads have not been considered because all seals were initially compressed at room temperature.

$$LMP\ (\mathrm{T,t}) = \mathrm{T} \cdot (C + \log_{10}(\mathrm{t}))\ \text{with T [K], t [h]} \qquad \text{(Eq. 1)}$$

The intention of the diagrams in Fig. 5. and Fig. 6. is to derive a master curve $F_R = F_R\ (LMP)$ from all investigated temperatures for both seal types based on the superposition principle. As the diagrams show, a master curve could not be found for the chosen constant material parameters (*C* = 11, *C* = 14).

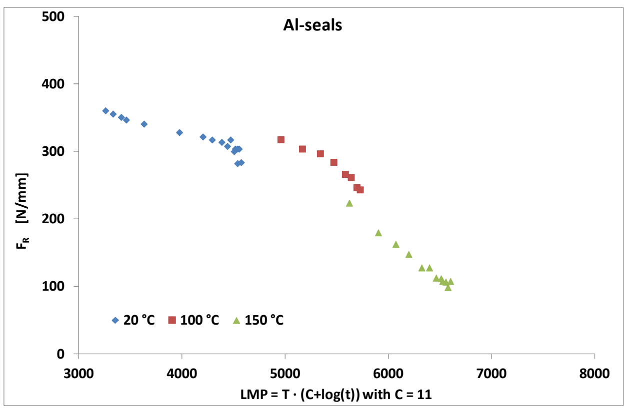

Fig. 5-1. Restoring seal force F_R and *LMP* (except initially load) for Al-seals with *C = 11*

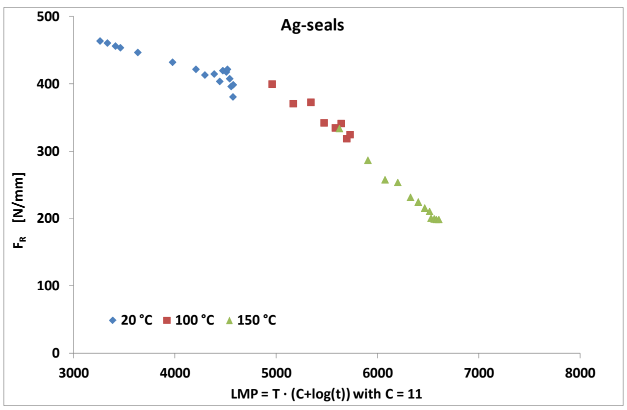

Fig. 6-1. Restoring seal force F_R and *LMP* (except initially load) for Ag-seals with *C = 11*

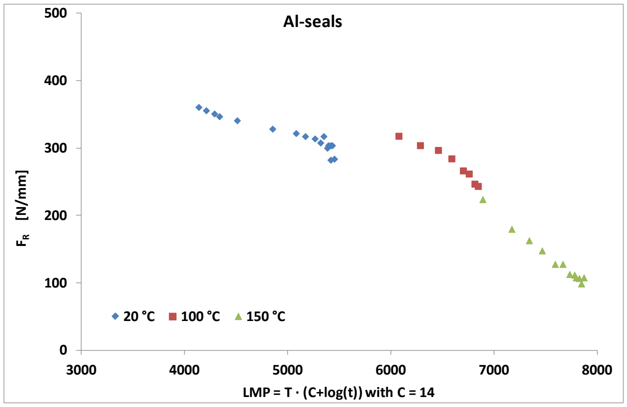

Fig. 5-2. Restoring seal force F_R and *LMP* (except initially load) for Al-seals with *C = 14*

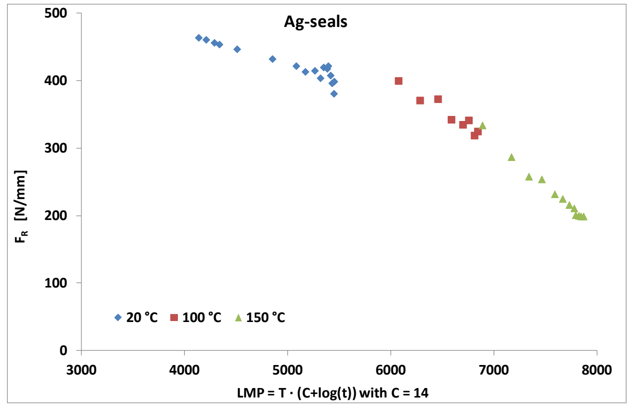

Fig. 6-2. Restoring seal force F_R and *LMP* (except initially load) for Ag-seals with *C = 14*

However in a second step calculations were carried out due to the correlative relationship between temperature and time with a constant material parameter *C* derived from Eq. 2. The outputs are shown in Tables I and II for several examples and demonstrate that there is no accordance with the experimental findings. Thus, the applied *C* - values are not useful to describe the test results appropriately.

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

TABLE I. Calculated time values t_1 and t_2 for Al-seals with *C = 11 and 14*

C	*F_R* [N/mm]	*T_1* [°C]	*t_1* [h]	*T_2* [°C]	*t_2* [h]
11	318	20	4.368	100	3
11	318	20	814.712	100	192
11	304	20	25.368	100	13
11	304	20	4.191.790	100	696
14	317	20	17.760	100	2
14	318	20	5.371.765	100	192
14	304	20	25.368	100	3
14	304	20	27.638.351	100	696

TABLE II. Calculated time values t_1 and t_2 for Ag-seals with *C = 11 and 14*

C	*F_R* [N/mm]	*T_1* [°C]	*t_1* [h]	*T_2* [°C]	*t_2* [h]
11	404	20	13.992	100	8
11	400	20	814.712	100	192
11	335	100	9.096	150	155
11	334	100	11.606	150	192
14	404	20	13.992	100	2
14	400	20	5.371.765	100	192
14	335	100	9.096	150	69
14	334	100	29.297	150	192

measured	*calculated*

Determining of the Material Parameter *C* by Larson-Miller

In a next step, we have investigated whether it is possible to determine a different value for the material parameter *C.* The *Larson-Miller* formulation assumes that every iso-stress line intercepts with the ordinate in a specified point $\{T^{-1} = 0; \log(t) = C\}$ see Fig. 7. by plotting time values log(*t*) versus inverse temperature (T^{-1}). Due to the boundary conditions of the specific test flange systems (with given depth of the seal groove and thus a constant compression) and the principle of force measurement iso-stress conditions cannot be measured for the seals. Therefore, iso-force states (F_R = const.) were considered for several testing temperatures and the corresponding time values ln(*t*) were calculated from Eq. 3. (see below). The range of iso-forces has been chosen in a way that the measured values of F_R for several testing temperatures are overlapped. Nevertheless, Fig. 8. and Fig. 9 show clearly that these ranges only exist for pairs of two selected temperatures (see red rectangles) and not generally.

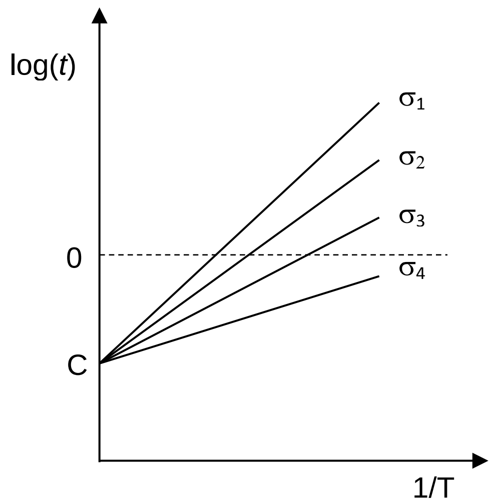

Fig. 7. Determining material parameter *C* by Larson-Miller

The measured test data of the decreasing seal force over time in a logarithmic scaling as shown in Fig. 8. and Fig.9. can easily be interpolated by a linear correlation like Eq. 3. Short-term measurements below 24 hours as marked by red dashed lines could only be performed at room temperature because test flanges at higher temperatures have to be taken from an heating chamber for restoring seal force measurements.

$$F_R\ (\ln(t)\,; T) = \mathrm{a(T)} \cdot \mathrm{ln(t)} + F_R(\mathrm{ln(t)} = 0; \mathrm{T}) \qquad \text{(Eq. 3)}$$

Based on these functions ln(*t*)-values for various iso-force states and depending on the test temperatures were calculated and are illustrated in Fig. 10. and Fig. 11.

For constant material parameter(s) *C* the linear extrapolation functions for each seal type should build one intersection point with the ordinate. But Fig. 10 and Fig. 11 show clearly that this is not the case; it cannot be found one single intersection point for each seal type concerning all test temperatures.

However, at least on pairs of temperatures ({20°C, 100°C}; {100°C, 150°C}) for each seal type intersection points can be determined (see *S* in Fig. 10 and Fig. 11). In this case a linear function depending on the restoring seal force F_R can also be found. Based on these findings it is possible to calculate the estimated time (ln*(t)*) for a given restoring seal force F_R.
For a chosen remaining seal force of 408 N/mm for Ag-seals the time value (ln*(t)*) is calculated exemplarily. In comparison with the appropriate measured value (of about 30,000 hours) the deviation of 1.6 hours between calculation and experimental results is negligible.

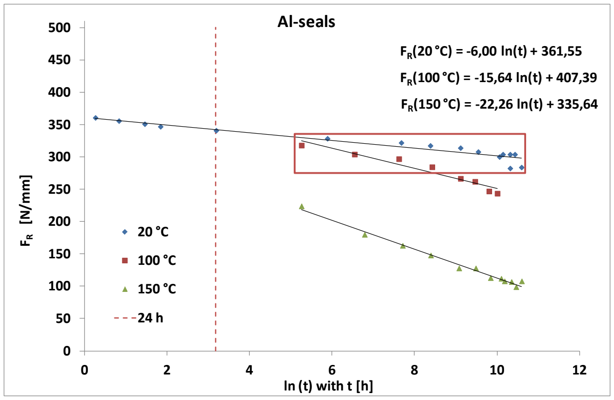

Fig. 8. Restoring seal force F_R over time ln(t) for Al-seals

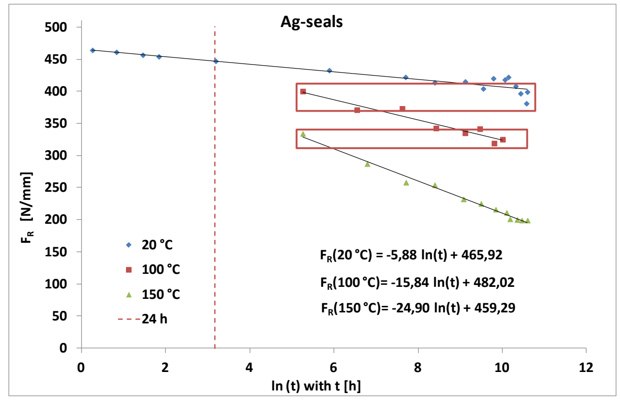

Fig. 9. Restoring seal force F_R over time ln(t) for Ag-seals

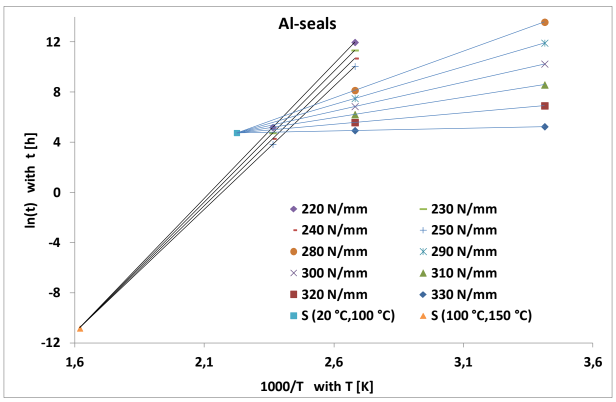

Fig. 10. Calculated ln(t) values over 10^3 times inverse temperatures for Al-seals including extrapolations

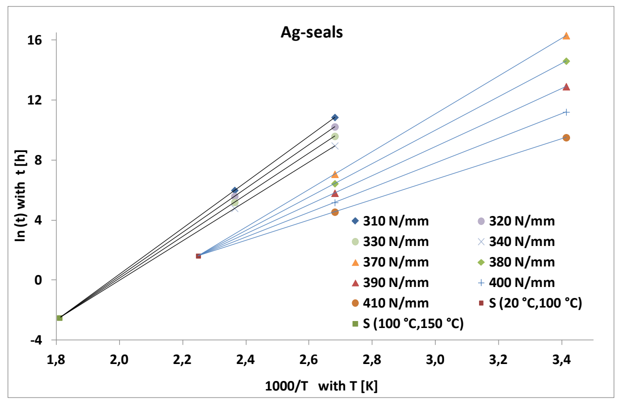

Fig. 11. ln(t) values over 10^3 times inverse temperatures for Ag-seals including extrapolations

With respect to the *Larson-Miller* approach (see Fig. 5 and Fig. 6.) and the foregoing described relations, master curves can be generated for pairs of temperatures for both seal types.
Fig. 12 and Fig. 13 show the derived linear dependency between restoring seal force F_R and a time-temperature parameter $b\,(T,\ t)$ which depends on the considered pair of temperatures exemplarily for Ag-seals. Examples of the application from this time-temperature relation and restoring seal force are given in Table III for Ag-seals. Due to the correlative approach between temperature and time for a seal type, for a determined value of $b\,(T,\ t)$ with $\{T_1,\ t_1\}$ it is possible to calculate the appropriate time t_2 for a given temperature T_2 by using Eq. 4. analog to Eq. 2.

$$b(T_1, t_1) = b(T_2, t_2) \qquad \text{(Eq. 4)}$$

In comparison to the deviation between calculated and measured time values (see Tables I and II) the deviation based on the modified time-temperature relation Eq. 4 is significantly lowered (see red marked results). As shown in Table III Ag-seals need to be carried out at a temperature of 150°C only for about 1350 hours to represent an equivalent of a 40-years operation time at 100°C.

Due to the fact that no consistent material parameter C for each seal type could be identified and the previously described time-temperature relationship $b\,(T,t)$ is valid only for two temperatures, already from the diagram in Fig. 8 and Fig. 9 and Eq. 3 it is obvious that a time-temperature equivalence exists according to the relation $F_R\ (\ln(t_1);T_1) = F_R\ (\ln(t_2);T_2)$.

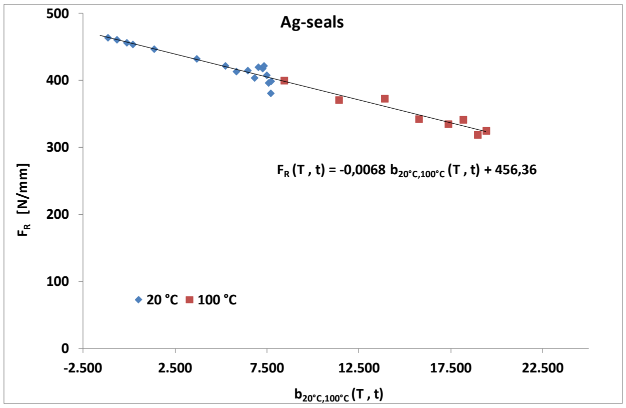

Fig. 12. Restoring seal force over a modified time-temperature relation for Ag-seals and pair of temperatures {20 °C, 100 °C}

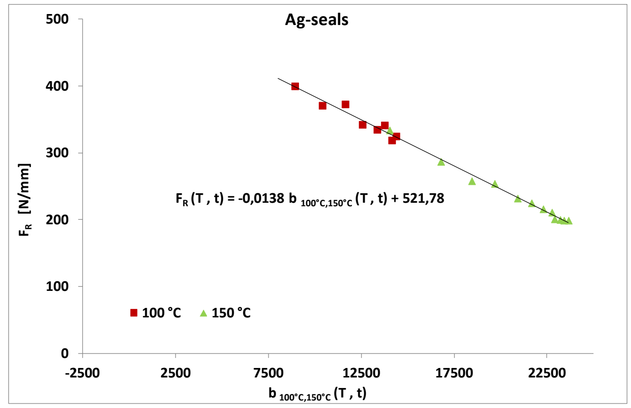

Fig. 13. Restoring seal force over a modified time-temperature relation for Ag-seals and pair of temperatures {100 °C, 150 °C}

TABLE III.
Examples of the application from time-temperature relations b (*T*,*t*) and restoring seal force correlation for Ag-seals

T_1 [°C]	t_1 [a]	t_1 [h]	$F_{R\text{-measured}}$ [N/mm]	T_2 [°C]	t_2 [h]	F_R [N/mm]
100	10	87.600	-	150	559	302
100	20	175.200	-	150	869	291
100	30	262.800	-	150	1125	284
100	40	350.400	-	150	1351	280
100		9.096	335	150	132	338
100		22.176	325	150	233	323

measured *calculated*

Discussion of Results

The results of our investigations have shown that the use of constant values for the material parameter *C* from literature ([2], [3]) or the application of the *Larson-Miller* approach [1] in general is not adequate. Some efforts are necessary to modify an appropriate time-temperature relationship being valid only for two selected temperatures. Further investigations concerning additional temperatures and taking into account different time-temperature relationships like *Manson-Haferd* formulation or *Mendelson-Roberts-Manson* parameterization [7] are necessary.
With a view to the granted license for the interim storage of spent nuclear fuel in Germany first results show that experiments for Ag-seals need to be carried out at a temperature of 150°C only for about 1350 hours to represent an equivalent of a 40-years operation time at 100°C and for Al-seals significantly less with only 313 hours.

Assuming that mainly time and temperature cause the decrease of F_R and both conditions can mutually interchange it is important to expect and demonstrate that no structural changes occur in the region of extrapolation. In regarding of Al-seals tested at 150°C in comparison to those which were tested at 100°C structural changes may occur because of the significant higher decrease of F_R shown for Al-seals tested at 150°C. Further investigations for additional temperatures should provide more insight. In addition, more information about initial decrease of the restoring seal force and an overlap of iso-force states for more than two temperatures may be obtained.

CONCLUSIONS

In the present paper different approaches have been discussed to derive an analytical relationship between short- and long-term tests to get predictable information about the long-term behavior of the restoring seal force of metal seals in a shorter time. Investigations for two different HELICOFLEX® metal seal types reveal that it cannot be found one material parameter *C* for each seal type based on the *Larson-Miller* approach [1]. However, a modified time-temperature relationship was developed. Based on this relationship it is possible to interchange time and temperature for pairs of testing temperatures and to quantify short-term tests at higher temperatures to simulate long-term effects of the restoring seal force at lower temperatures. Further investigations for additional temperatures are in progress.

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