Material Investigation and Long-term Sealing Behavior of Helicoflex® Metal Seals used in Containers for Spent Nuclear Fuel – 17135

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ABSTRACT

In Germany spent nuclear fuel is stored and transported in casks that possess bolted lid systems equipped with double jacket metal seals of Helicoflex® type in order to enable safe enclosure of the radioactive inventory. A dry interim storage period of 40 years was planned and the casks are licensed for that time span. However, due to political reasons and delayed disposal projects this time span is expected to increase significantly. It is therefore necessary to evaluate the long-term sealing behavior of the casks exceeding 40 years of operating time. In this paper, we discuss our approach to investigate the aging behavior of such metal seals, and to predict the long-term sealing behavior.

Accelerated aging component tests with seals are performed at temperatures up to 150°C to investigate the course of seal force, useable resilience and leakage rate. Even though these tests have already shown a significant decrease in seal force and useable resilience after aging times of less than 1 year, for most seals the leakage rate stayed better than the specified value. However, observation of the leakage rate has to be continued to determine the aging effect on the seal properties. As a main influence on the seal long-term behavior the increasing permanent deformation of the outer jacket has been recognized. It is made of aluminum or silver which means that the temperatures that are relevant for the aforementioned application in casks lead to creep deformation.

For further investigation of seal behavior a comprehensive research program concerning the material characteristics and aging behavior of the individual seal components, with an emphasis on the outer jacket material, has been launched. The aim is to get a better understanding of the time and temperature dependent creep mechanisms and deformation. The examinations are separated into tests on seal segments and tests on standardized specimen.

In order to be able to study the aging process, seal segments are compressed in flanges and stored at temperatures ranging from room temperature to 150°C. After defined time intervals the segments are analyzed by using standardized tests including structure investigation and creep tests. The results are compared with the behavior of the specimen basic raw material. This is done to ensure comparability of the thin jacket material and additional sheet material used for standardized tests. Thus, the prediction of component changes after different aging times at

different temperatures can be made. As a result forecasts on the long term seal behavior are intended.

INTRODUCTION

In Germany spent nuclear fuel is stored and transported in casks that are equipped with double lid systems. Originally, a dry interim storage period of 40 years was planned. Due to political reasons however, a significantly longer operating time is expected to be necessary. For this extended usage, the casks have to be licensed, which requires detailed knowledge about the influence of aging on the materials and components in order to predict the long-term leakage behavior. At the relevant seal position inside of the cask the operating temperature caused by the decay heat can reach 120 °C. The safe storage under normal conditions as well as accident conditions has to be ensured.

Bundesanstalt für Materialforschung und –prüfung (BAM) is involved in the qualification and evaluation procedures of the casks. For this purpose and to acquire and extend the knowledge about the component behavior investigation programs are run as well.

Metal Seals

The lids of CASTOR® casks, which are often used in Germany, are equipped with Helicoflex® metal seals to prevent the release of radioactive particles. These seals are made of three components as can be seen in Figure 1.

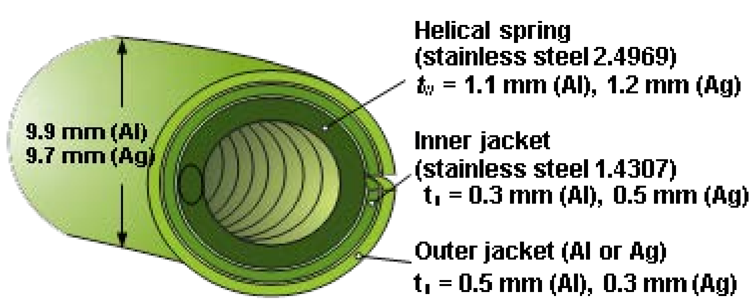

Fig. 1 Cross sectional view of a Helicoflex® metal seal [1]

The innermost part is a helical spring made of a nickel-chromium-cobalt base alloy (2.4969). This spring applies a restoring force to the layers and the lid when the seal is compressed in operation. The spring is surrounded by an inner lining made of stainless steel (1.4307). The purpose of the inner lining is to distribute the force of the spring while simultaneously preventing deformation damage caused by the spring wire shape to the outer lining. The outer lining is made of a soft metal, which in case of the relevant seal application, is aluminum or silver (Al respectively Ag – seals). The geometry of the seal in the compressed state is determined by the depth of the groove it is placed in when the lid is in contact with the cask. The lid is placed on the cask and the bolts are torqued. This leads to the compression of the seal to the operation point and in parallel a seal force is generated. The soft outer lining is plastically deformed, thus adapts to the sealing face and closes leakage paths caused by imperfections or general surface roughness.

The characteristic force deformation curve of a metal seal is shown in Figure 2. Additionally, the corresponding leakage rate is plotted. The curve visualizes the compression of a seal to its operating point, subsequent unloading and the simultaneously measured leakage rate. For the assembly in storage casks a leakage rate of ≤1.0E-8 Pa•m³/s is defined as the specified quality assurance criterion that has to be reached in order to ensure the intended functionality of the seal over the whole storage period.

In the beginning of the deformation the majority of the applied force is converted into elastic deformation. At the characteristic point Y_0 the leakage rate reaches the specified value for the first time. The increase of the applied force then causes primarily plastic deformation. Y_2 denotes the operating point. The following release leads to a nearly linear decompression until Y_1 is reached, the point at which the leakage rate exceeds the specified value. Another value of interest is the useable resilience r_u which corresponds to the decrease of deformation from point Y_2 to Y_1. That means, the value of r_u is the distance from Y_2 based on the deformation, a metal seal can be relieved from its operating point, before the specified leakage rate value is exceeded. This fact is relevant for the consideration of accident conditions with the possibility of axial lid movement.

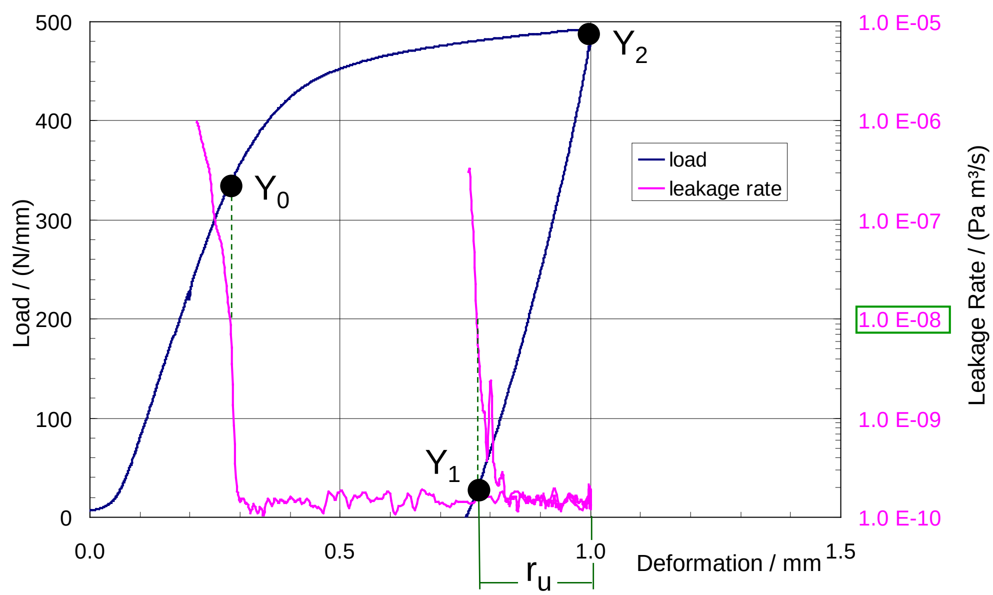

Fig. 2 Characteristic curve of a metal seal (exemplary Ag-Seal)

LONG-TERM INVESTIGATIONS

At BAM an investigation program is conducted, to study the time and temperature depending behavior of seal force, useable resilience and leakage rate. For this aging program, test flanges have been equipped with metal seals and stored at temperatures of 75 °C, 100 °C, 125 °C, 150 °C as well as room temperature. The seals that were tested possess a smaller inner diameter than the ones used in casks in order to enable the testing process with available testing machinery due to the necessary force that has to be applied to a metal seal to compress to the operating point. The cross sectional diameter is identical to the original seals, which is 9.9 mm for Al-seals and 9.7 mm for Ag-seals. The flanges are taken from the respective heating chambers at regular intervals and the compression of the seals is released in a universal testing machine while simultaneously measuring the compression force and the leakage rate. From the data the remaining seal force (F_R) and the useable resilience (r_u) can be obtained.

Exemplary results for Al-seals can be seen in Figure 3 and Figure 4.

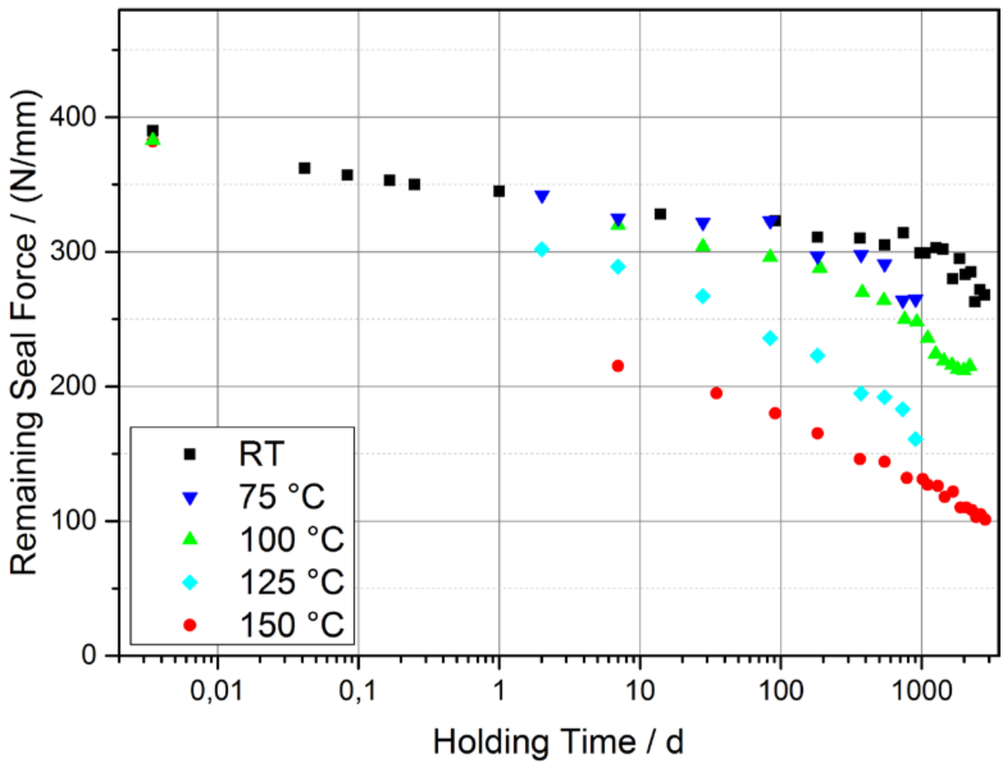

Fig. 3 Remaining seal force of Al-Seals at different temperatures

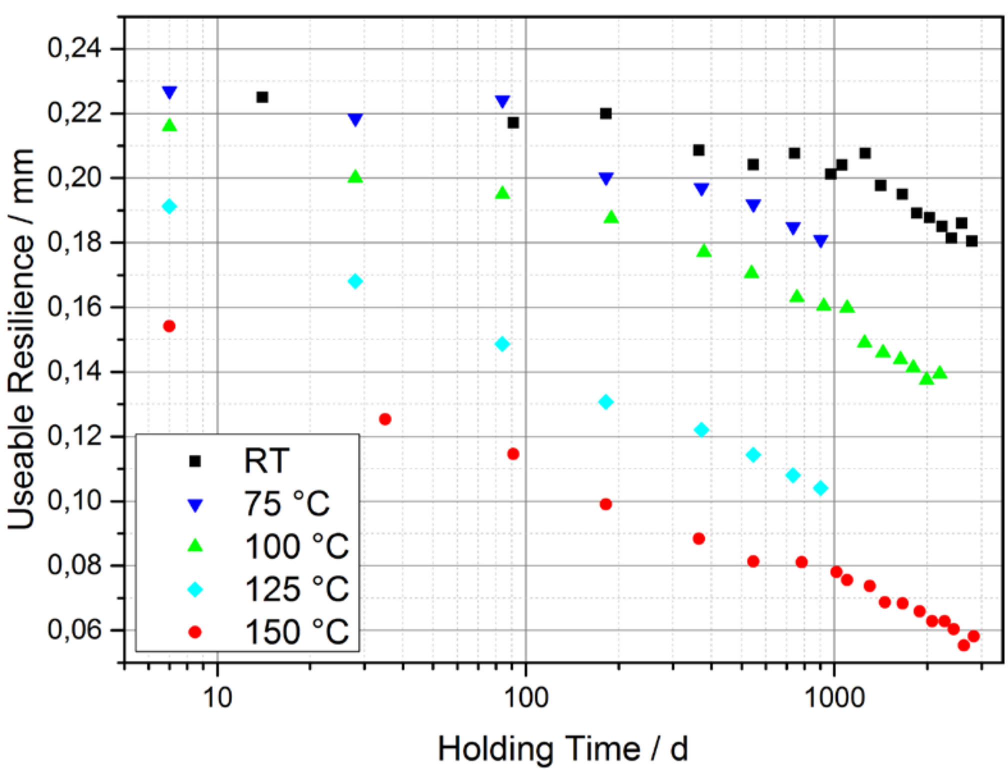

Fig. 4 Useable resilience of Al-Seals at different temperatures

The investigations show a significant decrease in F_R over time which results in decreased r_u as well. It was found that the main reason for that development is the creep of the outer lining. Due to temperature, time and compression the thickness of the outer liner decreases. Because of the simultaneously constant installation and groove geometry the spring compensates for the changed seal dimensions by expansion and thus relaxation. This results in a reduced F_R.

In spite of the decreasing seal force and useable resilience the leakage rate at the operation point as well as after decompression stayed within the required specified value. The only exception is the Al-seal that was aged at 150 °C. After 2 years of holding time the leakage rate at the operating point Y_2 exceeded 1.0E-8 Pa•m³/s and reached 6.0E-8 Pa•m³/s after 3 years [2]. In following tests however the leakage rate gradually decreased again indicating an improved sealing performance in spite of continuously decreasing seal force.
This development of leakage rate could be explained by two concurrent effects. On the one hand, creep leads to decrease of the outer liner thickness and therefore reduction of $F_{R,}$ with the result of a possible negative influence on the leakage behavior. On the other hand creep also leads to an improved contact between seal and lid or flange surface structure as well.

For further investigation seals with different torus diameters were assembled in flanges and aged at the same temperatures. The diameter of the whole seal rings are unchanged, the cross sectional diameter however is decreased to 5.6 mm for Al-seals and 5.4 mm for Ag-seals.

The trend of the general long-term behavior of the smaller seals is similar to the seals with larger diameter. The rate of decrease in seal force and useable resilience however is significantly higher. This can be explained by a higher strain on the seals contact area in case of the smaller seals. The diameter is roughly halved but the compression force is only about 20 % lowered. Therefore, the creeping of the outer lining is accelerated leading to a faster decrease of sealing force.

MATERIAL INVESTIGATION AND ANALYSIS

For the prediction of creep related occurrences it is common practice to employ empirical relationships such as the Larson-Miller-Parameter [3]. In the past this approach has been applied for the evaluation of the long-term behavior of metal seals as well [4, 5]. However, it has been shown that the calculation of a constant parameter C of the Larson-Miller-approach is not necessarily given because of its temperature dependency [6].

For further investigation of seal behavior, a comprehensive research program concerning the material characteristics and aging behavior of the individual seal components with an emphasis on the outer jacket material has been launched. The aim is to gain a deeper understanding of the aging and deformation processes and to derive appropriate analytical approaches for the prediction of the long-term behavior.

Thus, the following tests are planned:

Due to the heating and cooling times up to now it is not possible to perform measurements of seal parameters in short time intervals right after the initial compression test. Exceptions are the measurements with those seals which are stored at room temperature. However, the temperature dependent changes of the seal behavior especially during the first week of aging after initial compression are of interest as they are of importance for the understanding of mechanisms responsible for seal component performance. For the continuous measurement of the restoring seal force a new flange system is being designed. A compression load cell is assembled in the flux of force, which allows for the continuous measurement of the seal force. A simultaneous leakage rate measurement is intended as well.

Furthermore it is essential to gain more information about the property change of the individual seal components. For this purpose seals are cut into segments and compressed in flanges with groove dimensions equivalent to those of the test flanges. The seal segments are then aged at the same temperatures as the seals for the component tests. After aging times of 3 d, 10 d, 30 d, 100 d and 300 d the seal segments will be taken from the flanges. The components can then be investigated in terms of layer thickness, micro hardness, microstructural changes and creep process. With the additional data especially in the first weeks of aging an improved mathematical analysis of the process should be possible.

Besides the tests on seals or individual components detailed material properties are needed. The exact material designation and composition are known. Therefore substitute raw materials can be used to machine standardized specimens for creep tests in compression and tension mode. Thus, the involved creep processes can be investigated.

CONCLUSIONS

In order to evaluate the long-term safety of dry interim storage casks the aging behavior of the used metal seals is of high importance. The long-term investigations that have been done at BAM have shown the principal change of seal properties when aged at elevated temperatures. Even though several appropriate

phenomenological approaches have been proposed in the past, from the data it has not been possible to derive satisfying solutions for suitable analytical models to predict the seal behavior for storage periods exceeding the test periods.
In additional investigations the detailed material and component behavior is to be examined. With the obtained data and in connection with long-term component tests a contribution to the understanding of mechanisms responsible for the time and temperature dependent seal behavior is intended.

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