Understanding the Long-term Behavior of Sealing Systems and Neutron Shielding Material for Extended Dry Cask Storage

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Abstract: In Germany, the concept of dry interim storage in dual purpose metal casks before disposal is being pursued for spent nuclear fuel (SF) and high active waste (HAW) management. However, since there is no repository available today, the initially planned and established dry interim storage license duration of up to 40 years will be too short and its extension will become necessary. For such a storage license extension it is required to assess the long-term performance of SF and all safety related storage system components in order to confirm the viability of extended storage.

The main safety relevant components are the thick-walled dual purpose metal casks. These casks consist of a monolithic cask body with integrated neutron shielding components (polymers, e.g. polyethylene) and a monitored double lid barrier system with metal and elastomeric seals. The metal seals of this bolted closure system guarantee the required leak-tightness whereas the elastomeric seals allow for leakage rate measurement of the metal seals.

This paper presents an update on running long-term tests on metal seals at different temperatures under static conditions over longer periods of time. In addition, first results of our approach to understand the aging behavior of different elastomeric seals and neutron radiation shielding material polyethylene are discussed.

Keywords: Extended Storage, Dual Purpose Casks, Metal seals, Elastomeric Seals, Neutron Shielding Material

1. INTRODUCTION

In Germany, the decision was made to phase out of energy production by nuclear power stations. But since there is no repository available today this does not imply that questions of spent fuel storage are no longer relevant. Without an available repository it is expected that the initially planned and established dry interim storage license duration of 40 years in Germany will be too short and its extension will become necessary [1]. But since all data used for the safety case so far are also limited to this time span, for a storage license extension it is necessary to assess the long-term performance of spent fuel and all safety related storage system components in order to confirm the viability of extended storage. A topical overview on the German aging management approach for dry SF and HLW storage in dual purpose casks and the influence of aging mechanisms on transport safety and reliability of such casks is given in [1, 2].

The main safety relevant components for spent fuel storage are the thick-walled dual purpose metal casks which are approved for transportation and storage. These casks consist of a monolithic cask body with integrated neutron shielding components (polymers, e.g. polyethylene) and a monitored double lid barrier system with metal and elastomeric seals. The metal seals of this bolted closure system guarantee the required leak-tightness whereas the elastomeric seals allow leakage rate measurement of the metal seals. Irrespective of this application of elastomeric seals in SF and HAW storage casks only as auxiliary seals, they are of major interest for cask designs for low and intermediate level radioactive wastes.

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In order to ensure their full safety relevant functional capability during the whole extended storage period and for transportation after storage, the change of material properties of metal and elastomeric seals as well as neutron shielding polymers has to be investigated for long-term storage conditions.
For sealing systems, the sealing performance during long-term storage is crucial. This is mainly influenced by the reduction of the compression load which means also a reduction of the usable resilience. Therefore, for both types of seals the time-dependent sealing performance will be determined for different thermal and mechanical loads. For elastomeric materials also radiological aging has to be considered.
In case of neutron shielding polymers general changes of material parameters are investigated with special interest in changes induced by gamma radiation and aging, e.g. hydrogen release, crosslinking and mechanical behavior.

This paper presents an update on running long-term tests on metal seals at different temperatures under static conditions over longer periods of time. Due to creeping effects a reduction of the pressure force over deflection during loading and unloading does appear depending on prior holding times. Analytical approaches on basis of test data are developed and discussed to extrapolate seal performance (pressure force, elastic recovery, leak-tightness) to longer periods of cask operation.

In addition, first results of our approach to understand the aging behavior of different elastomer seals and neutron radiation shielding material polyethylene are discussed.

2. METAL AND ELASTOMERIC SEALS

Figure 1 shows a schematic representation of a German casks common lid and sealing system. The metal seals are responsible for the main sealing function and have to meet the high requirements of the specified leakage rate of 10^{-8} $Pa{\cdot}m^3{\cdot}s^{-1}$. 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.

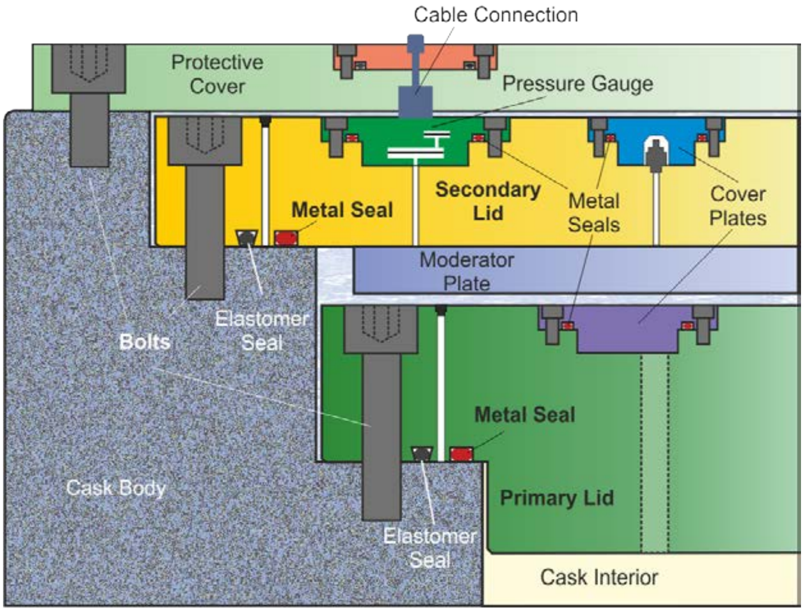

Figure 1: Lid area and sealing system of a transport and storage cask

The bolts fixing the lids onto the cask body have to be tightened 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 compression of metal and elastomeric seals 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.

2.1. Metal seal investigation

In Germany, metal seals used in dual purpose casks for SF dry interim storage are usually of the HELICOFLEX® HN200 type as illustrated in Figure 2. Such seals consist of an inner helical metal spring and two thin metal jackets. The outer metal jacket is made of aluminium (Al-seals) or silver (Ag-seals) to achieve and maintain tight contact between seal and lid or cask body surfaces.

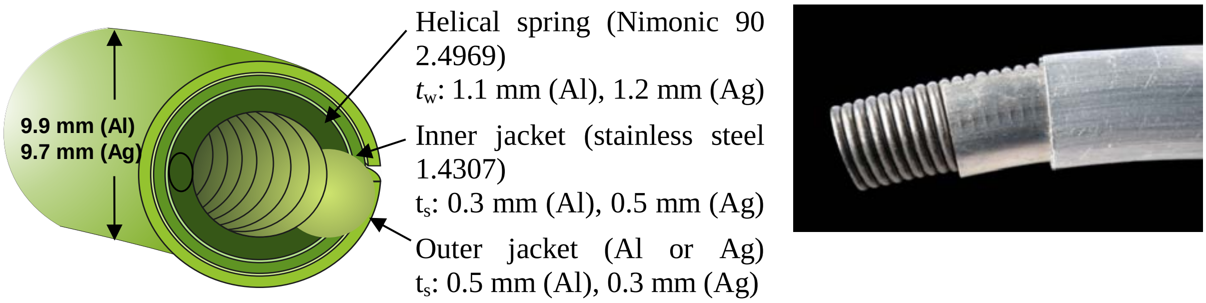

Figure 2: HELICOFLEX® HN200 seal type applied in test series and in dry storage casks

BAM is already performing investigation programs with such type of metal seals. To allow appropriable dimensions of our test setup, we use seals with a smaller overall diameter compared to seals that are installed in casks. However, the seal cross section diameter of about 10 mm as well as materials and dimensions of spring and jackets are identical to dual purpose cask seals to get representative test results. Depending on cask design and spent fuel decay heat, maximum temperatures at the seal area of cask lid systems reach about 110°C at the beginning of storage. Knowing that higher temperatures can accelerate aging mechanisms, in February 2009 and November 2010 BAM decided to start tests at three different temperatures: 20°C (ambient temperature), 100°C, and 150°C. The test flange systems and test conditions are described in detail in previous papers, e.g. in [3] and [6]. The test setup represents assembling conditions in casks and allows for simultaneous measurement of seal load-deformation relationship during compression and relieving procedures and standard helium leakage rate.

Major outcomes of these test series are decreasing pressure forces F_r and decreasing elastic recovery (useable resilience r_u until the specified leakage rate of 10^{-8} Pa·m^3·s^{-1} is exceeded) with holding times at the different temperature levels for both seal types (aluminum, silver). For example, the reduction of F_r and r_u depending on holding time and temperature is plotted over a logarithmic time scale for Al-seals in Figures 3 and 4, which illustrates a proper linear correlation and allows extrapolating very easily to longer time periods. This seal relaxation correlates to the reduction of the outer jacket thickness by plastic deformation improving the contact of the metal surfaces. As a result a proper seal function was observed even in case of nearly complete loss of pressure force. More detailed information can be found in [4, 5, 6]. In case of real casks, an exceeding of the specified leakage rate 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 bolts.

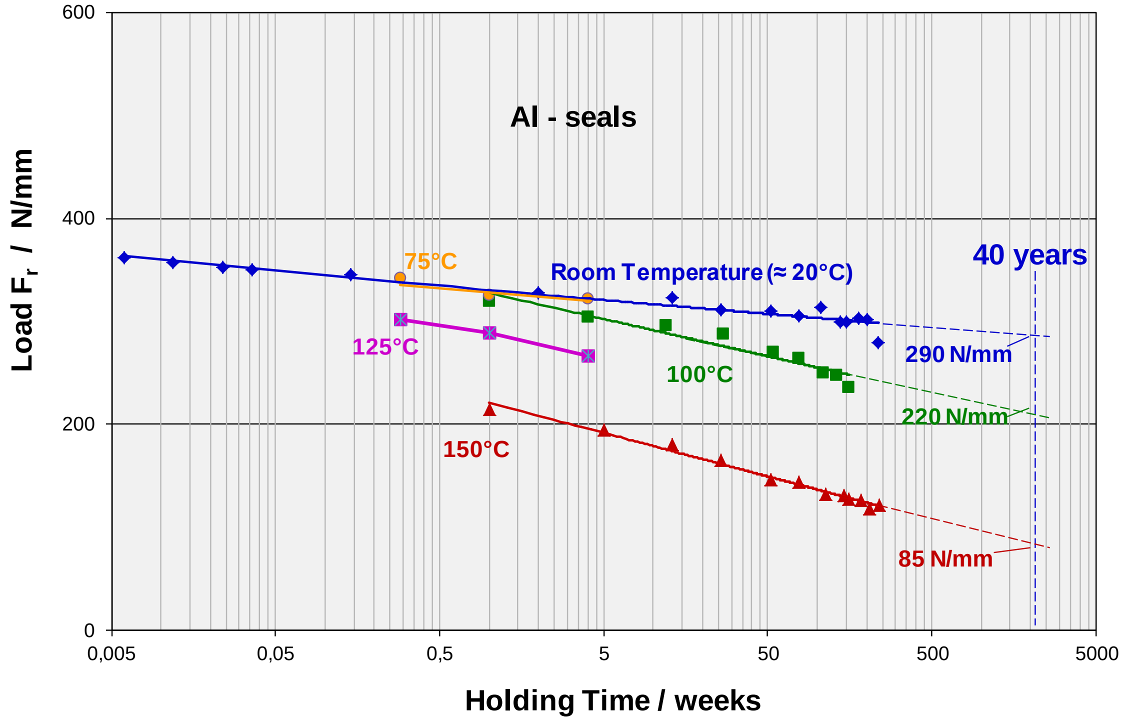

Figure 3: Reduction of restoring seal force F_r depending on holding time and temperature for current test periods and extrapolation up to 40 years (dashed lines)

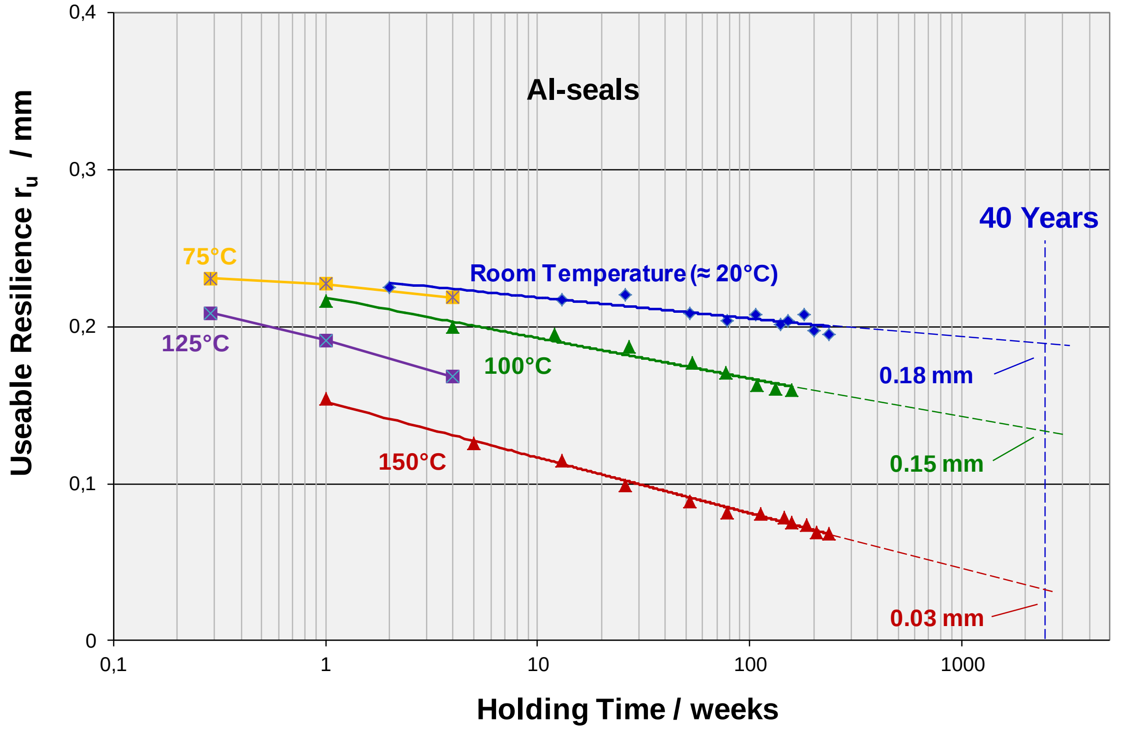

Figure 4: Reduction of useable resilience r_u depending on holding time and temperature for current test periods and extrapolation up to 40 years (dashed lines)

Recently performed investigations take account of the Larson-Miller relationship [7], 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 [8, 9, 10].
By using our test results for 20°C, 100°C, and 150°C, we also have discussed different approaches to derive an analytical time-temperature relationship between short- and long-term tests at these different temperatures to get predictable information on the long-term behavior of the restoring seal force and useable resilience in a shorter time. But the results of our investigations have shown that, e.g., the application of the Larson-Miller approach is not adequate so far [11]. Therefore it was decided that some efforts are necessary to modify an appropriate time-temperature relationship and that additional investigations are required, in particular for additional temperatures to gain more test data. Such tests at additional temperatures have been started this year for Al- and Ag-seals at 75°C and 125°C. First results for Al-seals at these additional temperatures are presented in Figures 3 and 4, showing that (i) there is almost no difference between 20°C and 75°C and (ii) that values for 125°C are well between those for 100°C and 150°C. As soon as we will have more data for the two additional temperatures, we will extend our analytical approach taking also into account different time-temperature relationships like Manson-Haferd formulation or Mendelson-Roberts-Manson parameterization [12].

2.2. Elastomeric seal investigation

It is known, that typical aging effects of elastomers can be caused by oxidation, irradiation and high temperature [13]. The consequence can be additional crosslinking and/or chain scission. Material properties of elastomers at low temperatures are determined by the rubber-glass transition (abbr. glass transition). During continuous cooling, the material changes from rubber-like entropy-elastic to stiff energy-elastic behavior, that allows nearly no strain or retraction due to the glass transition. Hence elastomers are normally used above their glass transition but the minimum working temperature limit is not defined precisely. Moreover, the influence of the above mentioned aging effects on changes of the minimum working temperature has to be discussed carefully.

Challenging requirements for reliable operation of elastomeric seals in radioactive waste containers are e.g. long-term use up to several decades, radiation effects resulting from the inventory, operation at elevated temperatures and at possible low temperatures during transport or within the storage facility, operation under static conditions and potentially under dynamic conditions in case of accidents. Although there are several common applications where some of these requirements apply as well, e.g. water pipes (static and long-term) or automotive applications (dynamic parts, temperature variations), the complete set of requirements is not often encountered as replacement is much easier and common practice for such applications. Therefore, the behavior of elastomeric seals at low temperatures as well as changes in material properties due to aging effects over long periods of time have to be investigated to understand the influence on seal performance. In addition, it has to be discussed, which changes occur due to aging at elevated temperatures and under irradiation influence.

To address these topics we started to investigate the behavior of elastomeric seals at low temperatures with regard to potential leak-tightness changes [14, 15, 16]. For the investigations, fluorocarbon (FKM) and ethylene-propylene-diene (EPDM) rubbers were selected as they are often used in radioactive waste containers. Some materials were purchased from a commercial seal producer and some materials were compounded and cured at BAM. The elastomers where studied by several thermo-analytical methods and compression set to characterize the material behavior at low temperatures. By performing compression set measurements, the degree of deformation can be determined that is not recovered after a certain time span after sample release. In the course of this investigation also a new method for characterization of elastomeric seal materials has been developed [17, 18, 19] that emulates the compression set measurement by using a Dynamic Mechanical Analysis device. By this technique time-dependent and temperature-dependent measurements can be performed semiautomatic and much faster than by standardized procedures. For instance, a clear temperature dependency of the compression set values was observed as the compression set increases with decreasing temperature. For example, this behavior is shown in Figure 5 for an FKM material. As the glass transition of this sample is in the range of -10°C to -23°C [19], the compression set increase on

cooling is connected with the material change from rubber-like entropy-elastic to stiff energy-elastic behavior, allowing nearly no retraction at even lower temperatures.
In addition to the described investigations, component tests were performed to determine the breakdown temperature of the sealing function of complete elastomeric O-rings and to correlate the temperature dependence of compression set with leakage rate, see e.g. [14, 15, 19, 20].

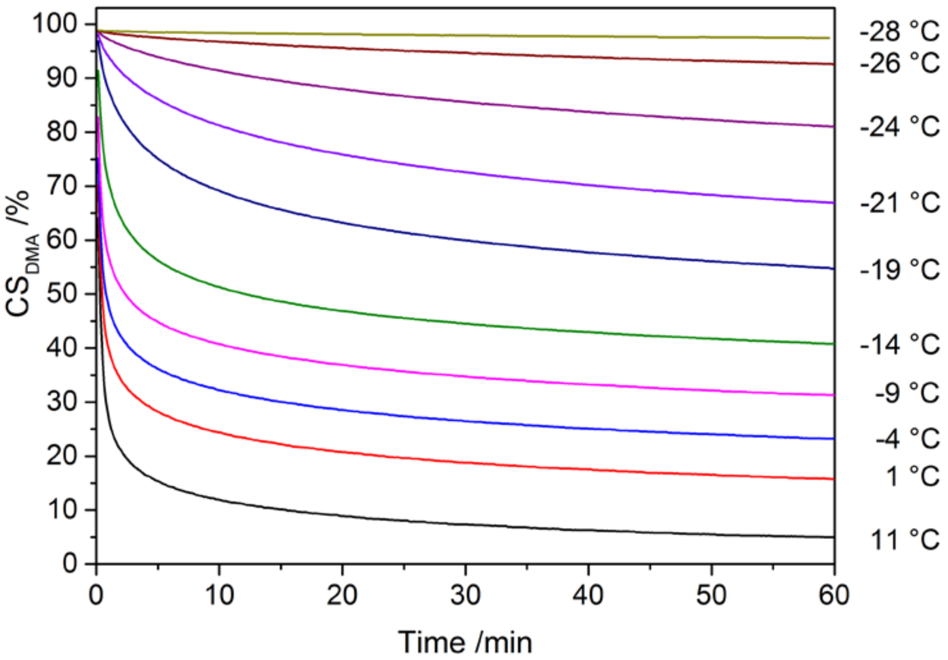

Figure 5: Temperature dependency of compression set values for an FKM rubber over time

To understand long-term performance of elastomeric seals accelerated experiments have to be performed. There are several standards that describe how to determine aging effects and how to interpret the results (see e.g. [21, 22]). These standards generally use a lifetime criterion and assume an Arrhenius-like behavior of aging processes. The question whether these assumptions are correct especially for the extrapolation over long periods of time is under discussion and several studies imply non-Arrhenius behavior, e.g. [23]. This is the reason for us to actually start an extensive experimental aging program with several materials to investigate the occurring changes of material properties of elastomeric seals by aging performed at different temperatures for up to two years. One goal is to determine the influence of compression during aging periods and therefore, compressed samples and O-rings under assembly conditions will be stored and later on analysed simultaneously. Aging temperatures are selected in the range from 75°C up to 150°C (i) to ensure significant acceleration of aging processes, (ii) to obtain sufficient amount of data, (iii) to prove the applicability of typical time-temperature-superposition approaches as e.g. Arrhenius, and (iv) to verify that no additional sample degradation occurs as a result of high temperatures selected for appropriate accelerated aging. The seal dimensions were chosen with a rather large torus diameter of 10 mm and an inner diameter of 190 mm to have sufficient amount of sample material for different analytical methods to be performed.
To allow a correlation of measured changes in material properties with seal function, a setup with seals mounted in flanges allows for leakage rate measurement by pressure rise method at chosen aging times.

As these tests will not consider irradiated samples, in addition we investigate elastomeric seals that were subjected to gamma radiation of up to 600 kGy to simulate a rather high gamma dose compared to seal application in casks. First results obtained for FKM samples indicate an increase of the lower application temperature limit. For example, as can be seen from Figure 6, there is a significant dose dependent increase of compression set comparable to the material behavior on cooling as displayed in

Figure 5. Detailed investigations are on the way to understand this observation in more detail, including known chemical changes of FKM samples induced by gamma irradiation.

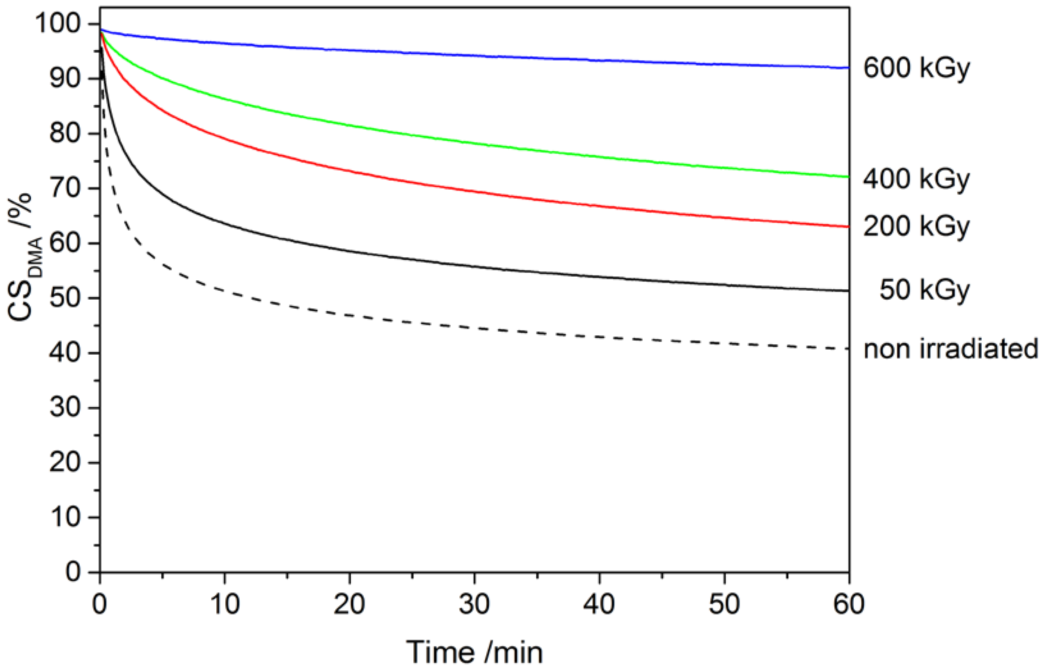

Figure 6: Gamma dose dependent increase of compression set values for an FKM rubber (measurements at -15°C)

3. INVESTIGATION OF (U)HMW-PE FOR NEUTRON RADIATION SHIELDING

Due to their extreme high hydrogen contents ultra-high molecular weight polyethylene UHMW-PE and high molecular weight polyethylene HMW-PE are a comprehensible choice as neutron radiation shielding material in casks for storage and transport of radioactive materials. As HMW-PE has a higher density compared to UHMW-PE, it is more efficient for neutron radiation shielding. However, in the broad operational temperature range from below room temperature up to, e.g., 160°C, which has to be considered depending on the heat generation of the cask radioactive inventory, HMW-PE shows substantial flow at higher temperatures above its melting region. Therefore, rods of HMW-PE are used only in cask positions at which the maximal operational temperature does not reach the main melting region; it is below 120°C. In contrast to this, UHMW-PE shows a better dimensional stability due to its higher molecular weight, even above the melting temperature of the crystalline phase, i.e., the polymer remains in a solid-like condition as the viscosity is still very high.

As a consequence of the radioactive inventory, for this application (U)HMW-PE have to withstand any type of gamma radiation induced degradation affecting safety relevant aspects. Several effects of ionizing radiation affecting structure and properties of the PE materials may be expected, such as chain scission, crosslinking, oxidation, provided that oxygen is present, and release of hydrogen. Especially the balance of chain scission and crosslinking determines the resulting changes in mechanical properties as it may affect the thermal expansion directly, and indirectly the recrystallization after (partial) melting of the semi-crystalline material. With regard to the application as neutron radiation shielding material, several safety relevant aspects have to be considered:

(i) neutron radiation shielding efficiency, which is influenced by thermal expansion and density changes,

(ii) influence on overall cask mechanical stability, which can be influenced by pressure build-up resulting from thermal expansion or hydrogen released as a result of decomposition of polyethylene chain molecules,

(iii) behavior under accident conditions (e.g. fire scenario, mechanical impact).

In order to determine (U)HMW-PE material changes we applied a set of characterization methods in order to identify changes in the neutron radiation shielding material that was gamma irradiated with a dose of up to 600 kGy. Such a high dose represents a typical value of gamma irradiation that may occur within 40 years of interim storage (currently approved duration of interim storage license in Germany) leading to major possible changes of the material. Detailed results were published in [24, 25]. It was confirmed that with the applied methods it is possible to detect and characterize structural changes of (U)HMW-PE induced by gamma radiation. In order to assure a sufficient neutron radiation shielding caused by the radioactive inventory, the obtained variation in density has to be considered to determine the (U)HMW-PE rods diameter. Until now, the obtained results indicate that the observed degree of changes of the irradiated material does not affect safety relevant issues for long-term neutron radiation shielding purposes under normal operation conditions. As the thermal expansion coefficient decreases due to the formation of crosslinks this aspect is not important under normal operational conditions. But for unusual high temperatures (e.g. under accident conditions), it might be of interest especially in combination with reduced flow properties of the irradiated HMW-PE.

To understand the influence of combined gamma radiation and thermal impact on (U)HMW-PE material property changes, in the future we intend to subject irradiated and not irradiated samples to accelerated aging at elevated temperatures as described for elastomeric seals aging experiments.

4. CONCLUSION

Due to major delays of siting and establishment of a final repository in Germany extended interim storage periods are inevitable in the future. To be prepared for lifetime extensions of interim storage facilities, systematic collection and evaluation of data on long-term behavior of relevant cask material and components is inevitable. One main criterion for long-term performance is the leak tightness of bolted lid closure systems.
Even if today two decades remain until most storage licenses expire in Germany, investigation programs for reliable long-term predictions are time consuming that necessitates starting such programs early. In particular, analytic approaches describing time and temperature dependent material behavior are of interest to gain reliable predictions and to establish accelerating test configurations. BAM has already commenced such test series for metal and elastomeric seals as well as neutron shielding polymers to understand material aging mechanisms in more detail for extended storage usage.

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