The evolution of safety related parameters and their influence on long-term dry cask storage

Klemens Hummelsheim[a], Jörn Stewering[b], Sven Keßen[a] and Florian Rowold[a,*]
[a] Gesellschaft für Anlagen und Reaktorsicherheit (GRS) mbH, Garching, Germany
[b] Gesellschaft für Anlagen und Reaktorsicherheit (GRS) mbH, Cologne, Germany

Abstract: For spent nuclear fuel management in Germany, the concept of dry interim storage in dual purpose casks prior to direct disposal is applied. Due to current delay in site selection and exploration, the necessity of an extension of the storage period beyond the granted license time for 40 years seems inevitable. Compliance with safety requirements under consideration of aging effects, in particular safe confinement, radiation shielding, subcriticality and decay heat removal will be crucial for the extension of these operation licenses. Thermal loads, mechanical stresses, gamma and neutron radiation are considered to be the main contributors to aging and degradation effects of the fuel, its cladding and the cask over the period of long term storage including subsequent transport. In order to assess the long term safety of such a system, knowledge about the evolution of the influencing variables is required.

The paper at hand describes numerical investigations in the field of spent fuel long-term behavior, e.g. fuel clad temperature and hoop stress over a time period of 100 years. Analytical storage temperature and stress calculations for a generic cask with different burnups and loading patterns of UO_2 and MOX fuel will be presented. The gained results will be related to actual questions regarding long-term degradation effects. Furthermore, shielding analyses with regard to varying densities of the integrated neutron moderator of the cask will be discussed.

Keywords: Dry Cask Storage, PWR fuel, Temperature, Hoop Stress, Shielding.

1. INTRODUCTION

In Germany, the on-site and central facilities for dry storage of spent nuclear fuel have granted license time of 40 years. In the middle of 2013, the new Repository Site Selection Act [1] became effective, which regulates the course of actions in order to find a final disposal for heat generating waste in the Federal Republic of Germany. An underground disposal site is scheduled to be found by the end of 2031. Based on experience, the following application, licensing and legal actions by the public may take up several years until construction work may begin. Regarding a realistic construction time and the fact that the licenses of the first central storage facility Gorleben and the first on-site storage facility in Lingen expire in 2034 and 2042, an extension of the licensed storage period will be needed.

It is important to know that the temporary licenses of 40 years are based on administrative reasons and not on limiting physical or technical parameters. Related to this subject, the “Guidelines for dry cask storage of spent fuel and heat-generating waste” [2] stipulates that if the licensed storage period seems likely insufficient, further appropriate safety assessments concerning e.g. long-term behavior of fuel assemblies and cask components have to be provided by the licensee. Consequently, it has to be shown that the safety functions, in particular safe enclosure of the radioactive inventory, subcriticality, radiation shielding and decay heat removal will be fulfilled during the envisaged timeframe beyond 40 years. Further, important aspects for the strategy of long-term storage prior to final disposal are transportability of the casks and manageability of the fuel assemblies. Accordingly, additional knowledge and data about material and component performance in conjunction with predominant conditions are necessary for sufficient safety assessments generated by the applicants and safety evaluations conducted by the competent authority and its technical experts.

* Corresponding author email: *Florian.Rowold@grs.de*

Together with the Federal Institute for Materials Research and Testing (BAM) and Oeko-Institut e.V., GRS is currently working on a state of the art report on the safety aspects of long-term dry storage on behalf of the Federal Ministry for the Environment, Nature Conservation, Building and Nuclear Safety. The work of the GRS is focused on long-term safety aspects originated from the fuel and its enclosure. Investigations comprise on the one hand a variety of PWR fuels, which have been used in Germany from the past up to now, ranging from low to high burnup UO_2 and MOX. Important parameters are represented by time-dependent decay heat, released fission gases and subsequent temperature and hoop stress of the cladding. On the other hand, the state of the art in the field of possible fuel degradation mechanisms is a matter of particular interest for the GRS. Safe enclosure and manageability of fuel elements require that systematic failure of the fuel rods should not occur during the storage period and that the fuel assemblies have to keep their geometric arrangement. Evidence of conformity has been verified for the initial 40 years of storage by the licensees but by extending the storage periods, one has to account for new boundary conditions, such as the temperatures and stresses mentioned, and investigate its influence on the mechanical properties of the cladding. Further interest is devoted to the radiological behavior of the casks and the impact of the long-term application itself as well as postulated neutron moderator degradation due to radiation.

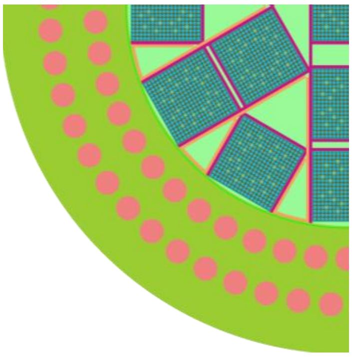
The paper at hand gives an overview of the work GRS performed in the field of safety of long-term storage until early 2014, consisting of burnup calculations, the development of generic models for heat transfer simulation, shielding analyses and cladding hoop stress prediction. First results of the numerical investigations for different cask loadings and fuel properties will be presented and discussed in the context of actual findings in the field of degradation mechanisms of dry storage systems.

2. MODELS AND METHODS

2.1. General Approach

The pursued dry storage of spent nuclear fuel in Germany in thick-walled dual purpose casks filled with helium and equipped with integrated neutron shielding components represents the underlying system of the described work and investigations. The generic model to simulate this system incorporates a typical cask designed to hold 19 PWR fuel assemblies of the 18x18-24 type (see **Fig. 1**). The maximum heat load of the cask is 39 kW. Further details will be given in the respective chapters.

Figure 1: Quarter section of the generic cask model



2.2. Burnup calculations

For the burnup and decay analyses, four types of fuel were chosen to cover the range of fuel used in German pressurized water reactors. An overview is provided in **Tab. 1**. Calculations were performed with the GRS in-house code OREST-V08. OREST-V08 is built upon the zero-dimensional burnup code ORIGEN, which is coupled with the one-dimensional HAMMER code for neutron flux spectrum and effective cross section determination. Beside burnup calculations, the code allows the determination of decay heat, activity and nuclide inventories for user-defined decay time steps. The fuel assembly is of the 18x18-24 type with an outside cladding diameter of 9,5 mm, pellet diameter of 8,05 mm and a pitch of 12,72 mm.

Table 1: Fuel data

Fuel	Enrichment [%]	Burnup [GWd/tHM]
UO_2	3,6	40,0
	4,4	55,0
HBU-UO_2	4,8	70,0
MOX	4,75 (Pu $_{fiss}$.)	55,0

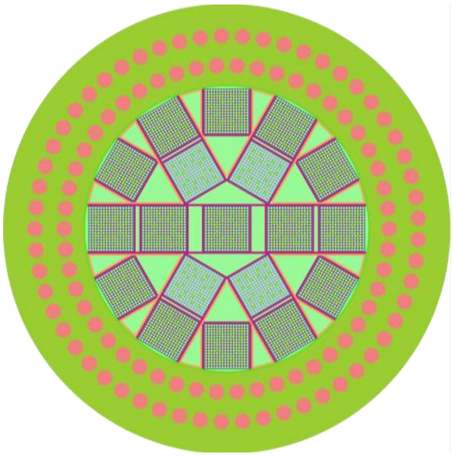
2.3. Cask model and heat transport

Thermal analyses were performed with the GRS Code COCOSYS V.2.4.0, whose intentional purpose is the simulation of severe accident propagation in containment systems of nuclear power plants. Its internal heat transfer module was used to create two-dimensional models of a generic cask and fuel assemblies. The basic principle is the radial subdivision of the structures into heat slabs and the definition of the heat transfer mechanisms between them. The input value is the average pin power derived from the burnup calculations. The code then determines the corresponding temperatures at the boundaries of the heat slabs. By repeated calculations for different time-dependent pin powers, the generation of functions of storage time vs. fuel clad temperature is being enabled.

To reduce complexity and resulting computing time, several assumptions and simplifications were necessary. Between the fuel rods, only heat radiation was considered whereas heat conduction through the helium was only applied between the outer fuel rods and the basket wall. Natural convection of helium inside the fuel element was neglected because of the two-dimensional approach and preliminary investigations showing low impact on heat transmission from fuel to cask environment.

Analyses were performed for three types of cask loading patterns. In the first type, the cask is homogeneously loaded, meaning that all 19 fuel element are of the same type with a maximum burnup of 55 GWd/tHM. In the second and third type, the cask is loaded heterogeneously with 15 UO_2 fuel elements (max. burnup 55 GWd/tHM) and 4 HBU-UO_2 or MOX fuel elements. The positions of the HBU or MOX fuel elements are represented by the lighter green in **Fig. 2**.

Figure 2: Scheme of heterogeneous cask loading



2.4 Fuel rod internal pressure and hoop stress

For the determination of the time-dependent fuel rod internal pressure, GRS developed a tool, which solves the Van der Waals equitation for the user-defined storage time steps:

$$p_k = \frac{n \cdot R \cdot T}{V - n \cdot b} - \left(\frac{n}{V}\right)^2 \cdot a \qquad (1)$$

The fuel rod internal pressure is determined by the amount of free gas, the free volume in the rod and the predominant temperature. As described, the temperatures derive from the thermal analyses, whereas the free gases are composed of the released fission gases Xenon and Krypton during operation and the fill gas Helium used for pressurization of the fuel rod in the manufacturing process. Fission gas amounts are given by the previously described burnup calculations in combination with a user-defined release rate. The amount of fill gas is calculated from the initial pressure level at room temperature in an iterative way. Free rod volume consists of plenum, gap and dishing volumes reduced by burnup dependent fuel swelling. Respective input data and quantities were based on publicly available data. The summation of the gas partial pressures leads to the internal gas pressure from which the cladding hoop stress σ_t can be calculated with Barlow's formula,

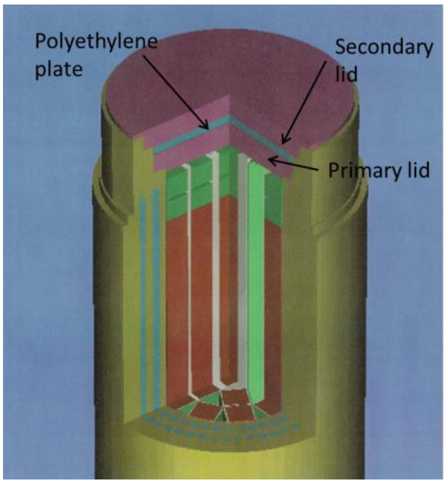
$$\sigma_t = \frac{p \cdot d}{2 \cdot s} \qquad (2)$$

where p stands for rod pressure, d for rod diameter and s for wall thickness.

2.5 Shielding analyses

A further matter of interest in our investigations is the evolution of the dose rate on top of the secondary lid during an extended storage period. Typically a storage cask is equipped with a polyethylene plate between the primary and secondary lid (see **Fig. 3**). The purpose of the plate is the moderation of neutrons. Thereby the absorption of the neutrons in secondary lid will be improved. Consequently the neutron shielding capability of the system improves as well.

Figure 3: Scheme of the double lid system



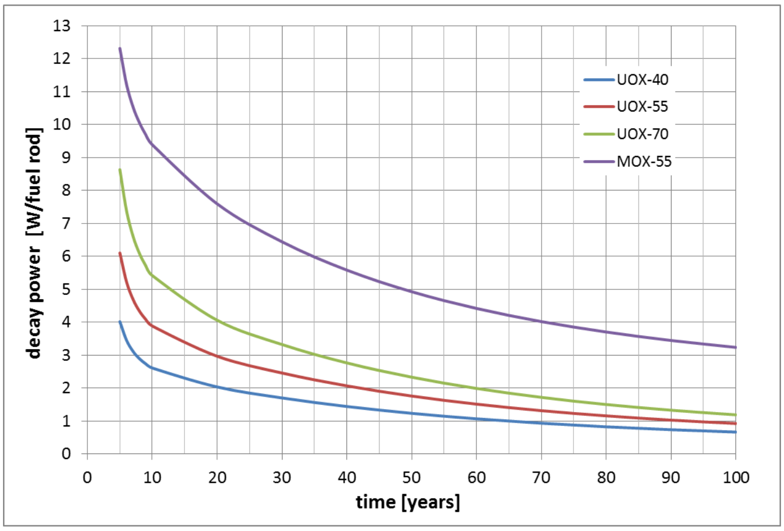
The equivalent dose rate outside the cask is made up by a source term that consists of neutrons and photons from fission and fission products and secondary gammas from capture processes. In addition, the activated parts of the fuel assemblies are considered. The source term resulting from the spent fuel was provided by OREST-V08 calculations. Activation was calculated with GRSAKTIV, an ORIGEN based code of GRS. The source is prepared in a 174 neutron and 39 gamma grouped spectrum. The shielding calculations were performed as follows: the fuel assemblies were segmented into homogenized fuel, plenum and head sections. The cask itself was modeled in detail. The particle transport calculations were done by MCNP5 with continuous energy cross section libraries based on ENDFB-VII data evaluation. To convert the calculated particle flux into dose equivalent rates, ICRP-74 conversion factors were used. The previously described homogeneous and heterogeneous cask loading patterns were investigated. The dose rate at the top of the secondary lid was calculated with a surface tally that gives an average value over the lid surface.

3. RESULTS

3.1. Evolution of temperatures, rod pressures and hoop stresses

Fig. 4 shows the decay power per fuel rod vs. time for the different fuel types, beginning five years after discharge. UO_2 fuel decay power depends on the final burnup. Between the HBU-UO_2 with 70 GWd/tHM and the moderate burnup UO_2 fuel with 40 GWd/tHM, decay power is about twice as high. For MOX fuel, the decay power level exceeds those of HBU-UO_2 and decreases to a lesser extent, e.g. the power level of the MOX fuel after 100 years is reached by the HBU fuel after 30 years.

Figure 4: decay power vs. time



By gaining the specific depletion characteristics of the different fuel types, we were able to use the results for different time steps as inputs for the thermal model of the cask and fuel assemblies. It is important to note that due to time and resource constraints, our investigations focused on the hottest fuel pin which are

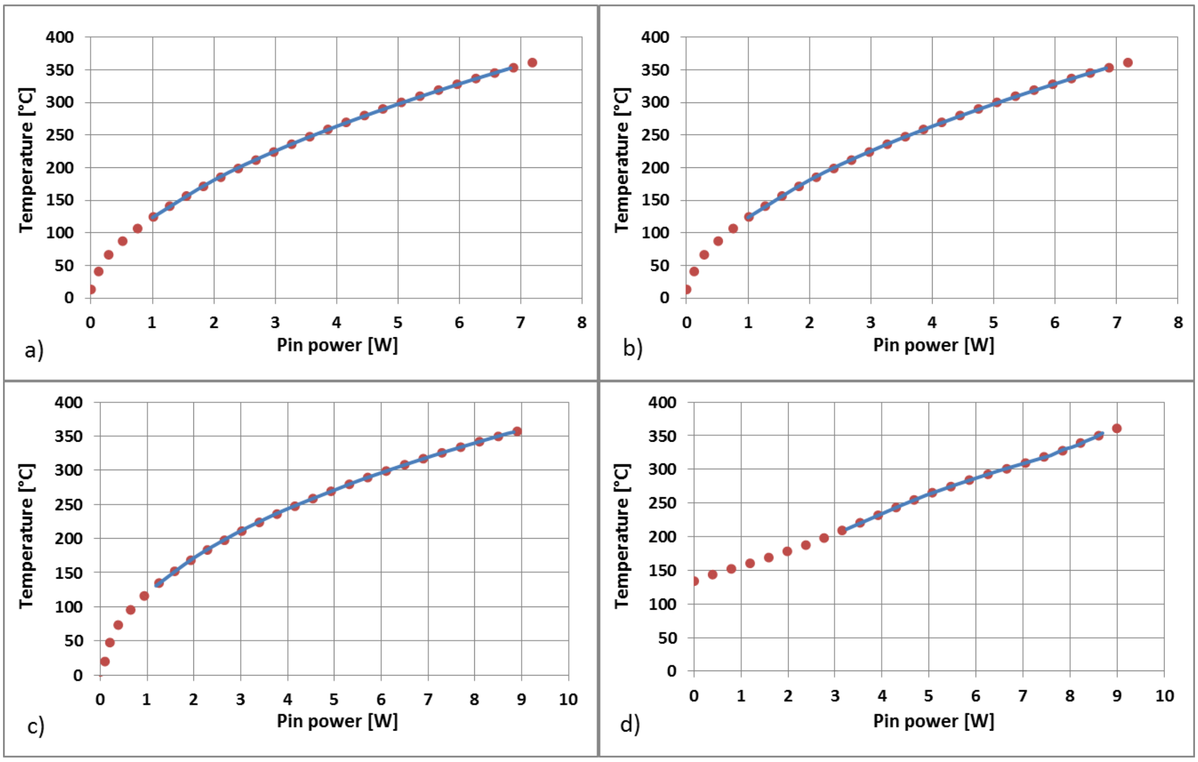
(1) the central fuel pin of the central fuel assembly in the homogenously loaded cask and
(2) the central fuel pin of the high burnup UO_2 and MOX fuel assembly in the heterogeneously loaded cask.

Nevertheless, this approach ensures conservatism by covering the most unfavorable conditions in terms of subsequent internal rod pressure and cladding stress determination.

Thermal analyses for the different cask configurations produced the results shown in **Fig. 5** with correspondent indexing:

a) 19 UO_2 fuel assemblies (FA) with 40 GWd/tHM burnup,
b) 19 UO_2 FA with 55 GWd/tHM burnup,
c) 15 UO_2 FA with 55 GWd/tHM and 4 HBU-FA with 70 GWd/tHM burnup,
d) 15 UO_2 FA with 55 GWd/tHM and 4 MOX-FA with 55 GWd/tHM burnup.

Figure 5: Cladding temperatures as a function of pin power



Fitting the curves in the pin power range for 100 years of storage (blue lines and **Fig. 3**) allows the setup of time dependent temperature functions. The difference in power ranges between the upper and lower diagrams is a result of the different loading patterns. With a maximum cask heat load of 39 kW, HBU and MOX FA are permitted to have higher heat loads at the expense of the remaining UO_2 FA in the cask, which reduces their required wet storage period. It is visible that the maximum temperature of the hottest pin is about the same for both configurations although the pin power is 30 % higher for HBU and MOX fuel. This effect is based on the positioning of the FA in the middle section of the basket (see **Fig. 2**), where additional heat transmission to central FA with lower power is provided.

As the temperature decreases very slowly, a quasi-static approach with a sufficient number of data points, e.g. every 10 years, was considered satisfactory for the following investigations. In order to calculate the time-dependent fuel rod internal pressure, knowledge about the initial free gas inventory in the rod is required. **Tab. 2** shows the total amount of gases generated till the end of reactor operation, resulting from the burnup calculations.

Table 2: Gas inventory at discharge

	Krypton	Xenon	Helium
Fuel	**EOL amount [mol/tHM]**		
UO_2 - 40 GWd/tHM	5,11	48,31	1,49
UO_2 - 55 GWd/tHM	6,87	66,85	2,11
UO_2 - 70 GWd/tHM	8,39	85,82	2,82
MOX-55 GWd/tHM	4,10	63,13	5,45

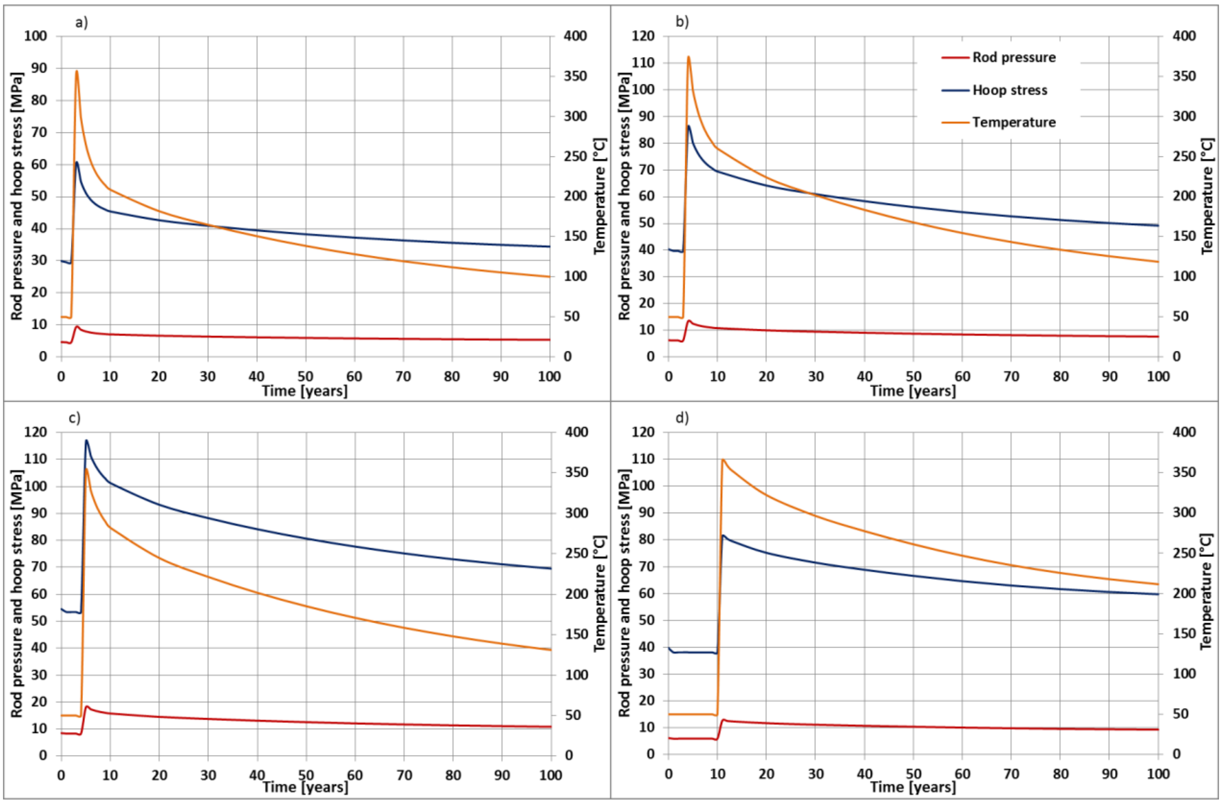
Fission gas release from the fuel increases with burnup but is mainly dependent on temperature and power history of the fuel. As a result, a broad range of fission gas release data exists. Taking into consideration the data range and recommendations given in [3], a pessimistic and at the same time conservative value of 15 % was chosen for the internal pressure calculations. Further input values comprise a fuel swelling rate of 1 % per 10 GWd/tHM burnup, a plenum volume of 16 cm³, a dishing volume fraction of 1,33 % and an initial pressurization level of 1,72 MPa. Most of the data originated from conversations with in-house experts of GRS. It is important to note that the data only represent one generic example of a PWR fuel rod. Fuel rods and fuel assemblies have been subject to steady research, development and optimizations. They are produced by a multitude of vendors which results in the existence of varying rod data.

In **Fig. 6** the results of the fuel rod pressure and cladding hoop stress calculations are presented. Additionally, the respective temperatures are provided for the

a) UO_2 fuel rod with 40 GWd/tHM,
b) UO_2 fuel rod with 55 GWd/tHM,
c) HBU-UO_2 fuel rod with 70 GWd/tHM,
d) MOX fuel rod with 55 GWd/tHM.

The diagrams show that wet storage periods had to be adjusted from 3 years for the low burnup UO_2 fuel to 10 years for the MOX fuel to reach maximum tolerable decay power levels. During that time, the cladding temperature was artificially held at 50 °C. At the beginning of dry storage the temperatures will increase rapidly and reach between 350 °C and 370 °C. Afterwards the temperatures decrease more slowly with higher burnup for UO_2 fuels. After 100 years, the UO_2 fuels have temperatures of 100 °C (a), 118 °C (b) and 131 °C (c). Due to the decay characteristic of MOX, the temperature decreases much more slowly and is at 211 °C (d) after 100 years. Governed by temperature, fuel rod pressure and hoop stress develop in the same manner but show a large variation across the different fuels. This is caused by the fission gas inventory and its release as main contributor to the rod pressure. Especially Xenon plays an important role with its fraction of 88 % of the fission gases. Since the generation of fission gases strongly depends on the burnup (see **Tab.2**), it is logical that HBU-UO_2 fuel rods will be affected the most. At the beginning of dry storage, the HBU-UO_2 (c) rod shows an internal pressure of 18,1 MPa and a clad hoop stress of 116 MPa. During a time span of 100 years, these values decrease to 10,8 MPa and 70 MPa. In comparison the low burnup UO_2 rod (a) starts at 9,3 MPa and 60 MPa and ends at 5,3 and 34 MPa. For the MOX (d) and UO_2 (b) rods with the same burnup of 55 GWD/tHM, the cladding hoop stresses at the beginning of dry storage are 81 MPa for MOX and 85 MPa for UO_2. Over the course of time, the hoop stress of the UO_2 rod decreases to 50 MPa whereas the hoop stress of the MOX rod decreases to only 60 MPa because of the higher temperature.

Figure 6: Evolution of temperatures, rod pressures and hoop stresses



3.2. Evolution of dose rates

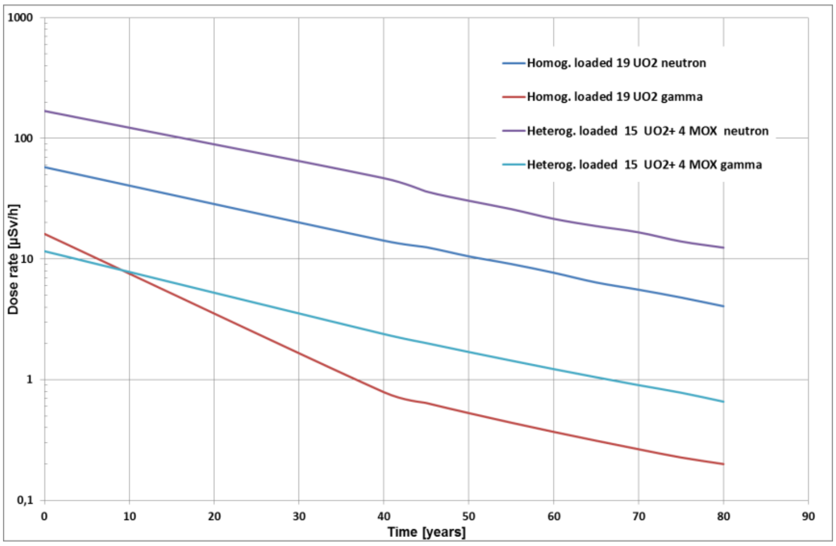
The shielding analyses were performed only for the homogenously loaded cask with spent fuel of 55 GWd/tHM burnup and for the heterogenic loading pattern with MOX fuel. It is important to note that for the spent UO_2 fuel a wet storage time of 5 years was used, the 4 MOX assemblies had a 10 years cooling time. The time axis in **Fig. 7** starts in the moment of cask loading. The presented results for the dose rates are divided in two parts: dose rates from neutrons and gammas. The neutron source data is dominated by spontaneous fission, the (α,n) part is nearly negligible. The gamma source originates from decay processes and from neutron induced reactions. Activation is a very small part and negligible after 50 years. The radiation transport calculation through about 35 cm of stainless steel in a very small angled direction cannot be done without variance reduction. Consequently, weight-windows were used to get statistically satisfying results. **Fig. 7** demonstrates that the highest dose rate on the lid will be expected for the heterogeneously loaded cask. Due to the higher neutron emission rate from MOX fuel (factor 8 to 9), the dose rate is about a factor of 3 higher than in the homogeneous case.

The gamma dose rate starts at a higher value for the uniform case because of a higher gamma source term. Then the dose rate decreases and both curves intersect. After 40 years of storage, the curves show similar characteristics and differ with constant ratio. In general, the gamma dose rate is more than one order of magnitude lower than the neutron dose rate.

The dose rate for neutrons will drop from the emplacement of the cask until a storage period of 80 years by a factor of 15. The gamma dose is a composition of fission product emission, activation, bremsstrahlung and secondary gammas. However, most contributors have no relevance to the final result. Fission products and secondary gammas solely contribute to the gamma dose rate. The gamma dose rates will decline with a factor of 80 for the homogeneously loaded cask and for the heterogeneously loaded cask with a factor of 18 respectively.

Additional calculations were done to show the impact of a reduced density of the moderator material in the previously described double lid system (see **Fig. 3**). The density was exemplarily reduced by 50 %. As a consequence, the neutron dose rate will rise by a factor of 3 to 4.

Figure 7: Dose rate evolution on the secondary lid of the cask



4. DISCUSSION

In this chapter, the results with respect to long-term safety of dry spent fuel storage will be discussed. Finding and erecting a final repository for spent nuclear fuel is a complex and time-consuming task. Many countries are adversely affected by significant delays, valid options, political issues or public resistance and consequently have to consider the option of long-term storage. Envisaged time frames extend up to 300 years in some countries. In this framework and under the aspect of safety, issues formerly considered less important such as aging management and long-term component behavior arose and are currently gaining more and more attention.

Regarding the spent fuel itself, the deterioration of cladding mechanical properties is an important aspect. One effect, which is often discussed in this regard, is hydride reorientation and embrittlement. During the reactor operation, the cladding absorbs hydrogen and after exceeding the solubility limit it precipitates in the form of circumferential hydrides. When the spent fuel is transferred into the dry storage cask and the cask is dried to remove residual water, the fuel heats up to around 400 °C where 200 ppm of hydrogen will dissolve again. During the storage period, the temperature will decrease and the hydrides will precipitate again. Under these conditions, a fraction of the hydrides could possibly precipitate as radial hydrides which are responsible for a reduction of ductility especially at low temperatures. This process is called hydride reorientation. The process is very complex because there is a multitude of parameters having an impact on the degree of reorientation e.g. the cooling rate, the thermal cycling, the temperature, stress levels and the cladding material itself. It has been reported that hydride reorientation was observed at stress levels above 70 MPa. For the ductile-to-brittle transition temperature it was observed that it strongly depends on the cladding material. Reported temperatures vary between 85 °C for M5, 150 °C for ZIRLO and 220 °C as the upper limit [4-8]. The results of the calculations show that the respective stress levels could prevail in fuel rods with a burnup higher than 55 GWd/tHM. By extending dry storage beyond 40 years, the possibility increases that the cladding temperatures fall below the ductile-to-brittle transition temperatures. With the concurrent decrease of the hoop stress and the mechanical load, adverse impacts should not emerge as long as the fuel is not exposed to additional loads. In the case of transport or accident scenarios, where additional loads are quite likely, this aspect should be considered.

Concerning radiation protection, an extension of the storage period will lead to further reduced dose rates on the surfaces of the cask. As a result, the working staff will be less exposed if work in the environment of the cask is necessary. Additionally, the postulated degradation of the neutron moderator after 40 years will not result in dose rates higher than those at the beginning of dry storage.

5. CONCLUSIONS

In order to assess the safety of long-term storage scenarios, the knowledge about the predominant conditions is required. These conditions originate from a multitude of variables which will vary for different dry storage systems. Our investigations aimed at the German concept of dry storage in thick-walled dual purpose casks. By using different computer codes and generic models, the GRS was able to cover most of the influencing variables. The analyses comprised different fuels, burnups, cladding temperatures and internal rod pressures. A set of results was produced with a conservative approach and allows the association with cladding degradation mechanisms. Since the long-term behavior of dry storage systems gains more and more attention on an international level, the presented work constitutes a contribution to the efforts.

Acknowledgements

This work was funded by the Federal Ministry for the Environment, Nature Conservation, Building and Nuclear Safety of Germany.

References

[1] *"Gesetz zur Suche und Auswahl eines Standortes für ein Endlager für Wärme entwickelnde Abfälle und zur Änderung anderer Gesetze (Standortauswahlgesetz - StandAG)"*, Bundesgesetzblatt 2013, Part 1 Nr. 41, (2013).

[2] Nuclear Waste Management Commission (ESK). *"Guidelines for dry cask storage of spent fuel and heat generating waste",* Revised version of 10.06.2013.

[3] M.A. McKinnon and M.E. Cunningham. *"Dry Storage Demonstration for High-Bunrup Spent Nuclear Fuel - Feasibility Study"*, Report No. PNNL-14390. August 2003.

[4] M.C. Billone et al. "*Used Fuel Disposition Campaign, Phase I Ring Compression Testing of High-Burnup Cladding*" Prepared for U.S. Department of Energy Used Fuel Disposition Campaign, Argonne National Laboratory, December 31, 2011

[5] M. Aomi et al. *"Evaluation of Hydride Reorientation Behavior and Mechanical Properties for High-Burnup Fuel-Cladding Tubes in Interim Dry Storage"*, Journal of ASTM International, Vol. 5, No. 9 (2008).

[6] R.S. Daum et al. *„Radial-hydride Embrittlement of High-burnup Zircaloy-4 Fuel Cladding"*, Journal of Nuclear Science and Technology, Vol. 43, No. 9 (2006), 1054-1067.

[7] K.S. Chan. *"A micromechanical model for predicting hydride embrittlement in nuclear fuel cladding material"*, Journal of Nuclear Materials 227 (1996) 220-236.

[8] P. Bouffioux, A. Ambard et al. *"Hydride Reorientation in M5® Cladding and its Impact on Mechanical Properties"*, Top Fuel 2013, Charlotte, North Carolina, September 15-19, (2013).