RECOOLING & OPENING OF SNF CASKS

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ABSTRACT

After fuel loading, drying of the interior and sealing of casks for spent nuclear fuel had been carried out, a certain error rate appeared at the Helium tightness test. In different cases, the tightness criterion of 10^{-8} Pa m^3/s was not reached. An interim storage over a long time has not been approved under these circumstances.

Many of changes were carried out to get a very high degree of cleanness. However, it was not possible to reach the above mentioned tightness criterion in each case.

The reasons had to be found to prevent time and money consuming reworks in the future and to seal the casks in the proper manner. A recooling technology had to be developed and tested to inspect and to seal the components appropriately.

The recooling procedure was uncomplicated, quickly manageable but nevertheless gentle. Effects on the tightness state of the spent fuel elements have not been noticed up to now.

The inspection procedures of the concerned casks have not been completed yet. During the investigations carried out till now, first reasons could be identified. The formation of white gray depositions on the sealing surface of the cask and on the seal itself as well as pitting at the aluminum-coated seal seem to be the reason for the not always reached tightness criterion. Additionally, faults during the production of the sheet aluminum for the aluminum coating of the seal could have caused this state. To avoid the detected depositions and the development of electrochemical elements in the sealing area of the cask, the procedure sometimes used to dry the cask interior together with the metal seal will not be applied anymore. Now, the seal is immediately and successfully pressed under water with the positioning of the primary lid. Only then, the neighbouring areas as the cask inner room and the seal space will be dried.

By the clearance procedure used now, time and cost intensive recooling is avoided. With this procedure, it is also possible to detect bigger tightness problems in advance.

In connection with the opening of the cask after recooling, the visible areas of the cask inner room and of the fuel elements could be inspected.

After a storage time of nearly four years, the good general state in the cask inner room has not changed as well as the state of the fuel elements after loading.

INTRODUCTION

Different clearance procedures and sealing technologies have been practiced at the SNF casks by EWN[a] till now with quite different success.

Three different variants of sealing procedures for casks were mainly used at the 44 casks processed to this day. One of these procedures proved to be quite problematic. The required tightness [1], [2] of the primary lid seal of < 1.0 x 10^{-8} Pa m^3/s could not be reached at some casks.

First, it was assumed that the tightness deviation of the primary lid from the desired value was mainly caused by pollution during the clearance procedure of the primary lid. It was particularly assumed that crud, other particles and rust from the tapped blind holes near the sealing surface could have been deposited in the sealing area before positioning the primary lid.

Due to the high requirements on dryness of the aluminium coated seals of the primary lid, the manufacturer introduced in the past the common drying of the cask interior and the non-pressed seals. It was assumed that remaining water droplets under the aluminium layer could cause harmful effects. To avoid this, the lid remained by spacers some millimeters above the sealing surface, in a kind of state of suspense. Another lid which was positioned over it sealed the complete system during the drying process. By a directed demineralized water jet, the seal surface should be cleaned. However, after several deviations from the tightness criteria, it was supposed that crud, other particles and rust were transported to the area of the seal surface.

By these reasons, the sealing procedure was changed once more. After a cleanness check of the seal surface underwater by TV-inspection, the primary lid is immediately positioned. Following, the seal is pressed by tightening the lid bolts.

All casks processed with this method reached far better results than the requested tightness criteria (Table I).

Table I Tightness criteria of SNF casks – licensing requirements

Storage Law [1] (Interim Storage North)	Transport Law [2]
10^{-8} Pa m^3/s for primary lid **<u>and</u>** secondary lid	10^{-8} Pa m^3/s for primary lid **<u>or</u>** secondary lid

It had to be decided how to proceed with the casks which did not reach the requested tightness criteria. The following possibilities were discussed:

- Recooling, unloading and investigation of possible reasons of all 7 casks
- Investigation of at least 2 casks after recooling
- Intermediate storage of the affected casks for at least 3 or 40 years

It was decided to intermediately recool all affected casks because of expected uncertainties with the approval authorities. Additionally, this decision helped to prevent further similar mistakes as well as high costs and delays by faulty processed casks. A programme was prepared to investigate the reasons and a recooling procedure for the casks with spent fuel elements was developed.

TIGHTNESS OF SNF CASKS

The used casks are equipped with a double lid system. The sealing system of the affected primary lid will be described briefly.

There are two grooves in the container lid of stainless steel. The metal seal which is fastened by clips is in the inner groove. An elastomeric sealing ring is positioned in the more outside groove. This elastomeric sealing ring is only needed for the tightness test of the primary lid. After positioning of the primary lid, both seals lie on the flat nickel-coated sealing surface of the cask body. The HELICOFLEX metal seal consists of three parts. The inner part is a ring of an elastic coil spring of Nimonic metal. This coil spring has a high-grade steel coat which is open to the outside of the cask, and the steel layer is coated with a plastically deformable aluminum layer.

By tightening the lid bolts, the seal is pressed together by approx. 1.2 mm, and thus, a sufficient tightness is reached.

It can be distinguished between the following three sealing procedures:

- wet pressed metallic seal
- dry pressed metallic seal
- dried and dry pressed metallic seal.

First method: The sealing ring is fastened at the primary lid. The cask is in the water. The lid is positioned on the SNF cask body. This process is remotely monitored. The sealing ring is already pre-pressed by the deadweight of the lid (approx. 7.5 Mg). Later, the seal is pressed once more by tightening the lid bolts. The cleanness of the sealing area is checked with underwater color cameras before the lid is positioned. Second method: In case of a dry-pressed seal, the dry-loaded or wet-loaded casks inventory is not under water during the positioning of the primary lid. The cask is emptied to a lower level than the cask sealing surface. The sealing surface is dried and cleaned and the seal is pressed dryly. However, this variant was only used in exceptional cases because of the radiation protection situation. Third method: The primary lid with the sealing ring and spacers is positioned under water on the cask. A distance of approx. 5 mm remains between seal and cask. Afterwards the cask is lifted out of the water and emptied to a level lower than the cask sealing surface. Another lid with auxiliary openings is mounted on the upper cask flange. Then, the complete interior is emptied through these auxiliary openings. After sealing of these openings the cask is vacuum-dried. With this procedure the seal can be dried too. Thus, it is prevented that water will remain between the aluminum layer and the high-grade steel coat of the seal, i.e. corrosion is not possible. Later, the upper serving lid will be removed. After removing the spacers, the lid bolts will be tightened and the seal is completely pressed.

The tightness test is carried out after drying the container interior and the room between the two sealing rings. This test is executed as an integral helium tightness test by vacuum procedure. For this, the cask interior is filled with helium with a pressure of 1000 hPa. Then, the helium leakage rate in the room between the two sealing rings is determined over a fixed time. The tightness test of the other cask lids is performed in the same way.

RECOOLING

In principle, it would have been possible to go back to recooling devices of other users of the cask family. However, an own equipment was designed and constructed for reasons of technology and costs.

To be able to assess if the recooling process affected the tightness of the fuel rods, special control steps had to be planned. Therefore, the determination of the activity concentration of krypton-85 in the cask is the first step before the beginning of recooling. The helium gas pressure in the cask will be increased some more than the ambient pressure so that the activity concentration of krypton-85 can be determined by a noble gas monitor. Gas samples will be collected to make later comparisons possible.

Another preparatory step is the removal of the installed molecular sieve candles [3]. These candles help to prevent corrosion on the aluminum seal during interim storage. The molecular sieve absorbs free water which could appear in the cask interior during this time. The collected water quantity is determined by the mass increase of the molecular sieve candles. The removal of the candles was performed very carefully and quickly. Therefore, the candles were immediately shrink-wrapped with steam impermeable foil.

Recooling Technology

After the evacuation of helium from the cask interior, the real recooling process starts by filling in demineralized water with a temperature of 25 °C. The demineralized water is supplied through a lance which goes down up to the bottom of the cask. First, the quantity of demineralized water supply is 1.5 to 2.0 m^3/h, later 0.6 m^3/h. Steam and gas will be evacuated from the upper inner cask room. Pressure relief valve, safety valve, control valve and non-return valve prevent inadmissible system states. In case of too high pressure, steam would be released into the water filled reloading pond. The escape of gas into the surroundings can be avoided by directed suction.

When the cask is filled with demineralized water up to approx. 95%, the activity concentration of krypton-85 (Table III) will be determined once more after a certain waiting period. This measurement is performed with a connected noble gas monitor.

By comparing the first and second measurement, the alteration of the fuel rod tightness is estimated.

In compliance with prescribed holding times, the cask will be filled up to a certain residual gas volume.

To avoid a too high hydrogen concentration from electrochemical processes , the gas room will be rinsed with nitrogen several times.

The cooled down spent fuel cask can be transported to the reloading pond to unload the fuel elements under water.

If higher deviations will be found by the comparison of the activity concentration of Kr-85 before and after recooling, the reason has to be determined. It becomes problematically, if a defined number of leaky fuel rods [3] is exceeded. This cask or another one with these fuel elements cannot be loaded anymore. A new loading variant has to be selected.

Evaluation of the Recooling Process

Recooling processes with spent fuel casks (Table II) executed till now have not caused technological problems.

Table II Cask details: Storage time and thermal energy

Cask No.	Store time till recooling [years]	Thermal energy at the time of recooling [kW]
035	2,0	7,1
104	3,7	6,1
032	2,2	8,9

During the filling procedure with demineralized water, the pressure in the cask increased insignificantly over the prescribed holding times. The highest achieved absolute pressure was 2020 hPa. The pressure increased from 2000 hPa to 2020 hPa during a holding time of 15 minutes at a residual gas volume in the cask of only just 5%. It is important to have exact knowledge of the free volume in the cask.

The volume of the cask interior determined arithmetically differs from the measured volume. The inaccuracy of the flow measurement of up to 5% has to be taken into account, too.

The duration of the real recooling process is only approx. 2.5 to 3 hours, i.e. the time between the beginning of filling demineralized water and the time the residual gas volume has reached about 5%. Holding times and residual filling approximately take another hour. Within these 3.5 to 4 hours of filling process, changes of the surface temperature are measurable at the thick-wall casks. At an environmental temperature of 24 or 25 °C, the temperature at the primary lid is increased by 3 to 4 K, and decreases by approx. 6 K at the bottom of the cask.

It could be observed that the tightness state of the fuel rods obviously does not deteriorate.

Table III. Cask Details: Fuel Rod Tightness

Cask	Change of krypton-85 activity concentration [Bq/m³]
035	0.52×10^6
104	0.10×10^6
032	3.00×10^6

The limit value[b] for the increase of the activity concentration of krypton-85 is a maximum of 190 x 10^6 Bq/m³. If this value is exceeded, the tightness state of the fuel elements must be taken into account for loading since the maximum number of leaky fuel elements in a cask is restricted.

INVESTIGATION OF THE SEAL

It was necessary to prepare a special test programme as early as possible to investigate the reasons for the non-fulfillment of the prescribed tightness criteria. Authorities and authorized experts required to take part already at an early stage in the selection of the investigation methods to be used. It was necessary to describe the methods to guarantee that the recooling and opening of the casks do not negatively influence the investigation results. Additionally all following control methods on site and in the laboratory had to be described. The positions of the sealing elements had to be secured and marked to allocate possible findings. Also the type of documentation and sampling for follow-up investigations in a special laboratory had to be defined.

On Site Investigation

Underwater colored cameras monitored the installation position of the metal and elastomer seals during the lifting process of the primary lid. Also, the presence of foreign particles and other pollution was checked. All image sequences were recorded.

The primary lid was inspected from below under water along the complete sealing rings by cameras. First unusual features could be found at the seals. The original mounting position had not changed. However, white gray depositions could be recognized here and there at different surface areas of the sealing rings.

The first control of the sealing area of the cask was carried out under water with the same method. In this connection, deposition areas of dried spots could be seen, especially due to their high-contrast borders. Also, brown, dot-like depositions were located here and there. This knowledge was not meaningful enough to explain the deviation from the required tightness criteria of the cask. In addition, the quality of the pictures was negatively influenced by light reflexes and sticky gas bubbles on the aluminium seal and the sealing area of the cask and made the evaluation difficult.

The next inspection of the sealing elements was carried out outside the water and thus under better conditions. The first suspicious circumstances (Fig. 1) were later confirmed in the laboratory. However, the visual inspection already showed that the seals were properly installed, at least defects and foreign bodies were not found at the first two investigated SNF casks. The deposition areas of the dried spots with their borders already detected with the underwater camera were now in the dry state more clearly seen on the sealing area of the cask. The print of the seal on the sealing area of the cask was here and there covered by dried spots. Such spots could also be seen on the aluminium coated seal at the same places. On the pressed area of the seal, defects were seen. Under microscope, defects became in some cases visible as stitch-like pitting scars. In some cases, brown depositions in the scars could be seen.

Fig. 1 Pitting scars on the aluminum coated seal of the Primary Lid

For further investigation in the laboratory, the obviously faulty sections of the seal were cut out.

Investigation by SEM/EDX[c]

The composition of the detected depositions (Table IV) and their thickness, and the composition of the depositions in the corrosion scars had to be investigated. Furthermore, foreign material from preservatives and other auxiliaries used for the clearance of the casks had to be found.
Dimension and depth of the existing corrosion scars in the Al-coat of the seal had to be determined as well. By transverse microsections (Fig. 2) in the sample material and assessment by SEM, it was intended to investigate the Al-coat damages.

At the areas of findings, the following elements were found by SEM/EDX investigation and roughly divided into three groups:

Table IV Elements from the SEM/EDX Analysis

mainly	secondary elements	trace elements
AL, O, Cl, Fe	Ag, C, Si, S, Ti, Ca	Na, Ni, P

Quantitative statements are not possible because of the composition of the surfaces.

Fig. 2 SEM picture at the place of pitting corrosion of the aluminum seal

The distinction between oxides and hydroxide was not possible. Boron as a component of earlier used boron acid was not detectable in the spectrum.

The main deposit of C, O, Ca, Ti, Si, P and S could be determined by comparison analyses. The elements can be assigned to the used CRC ceramic paste[d], the protective gloves and the elastomeric seal.

Comparison tests at undamaged spots of the aluminum seal mainly showed the presence of Al and O.

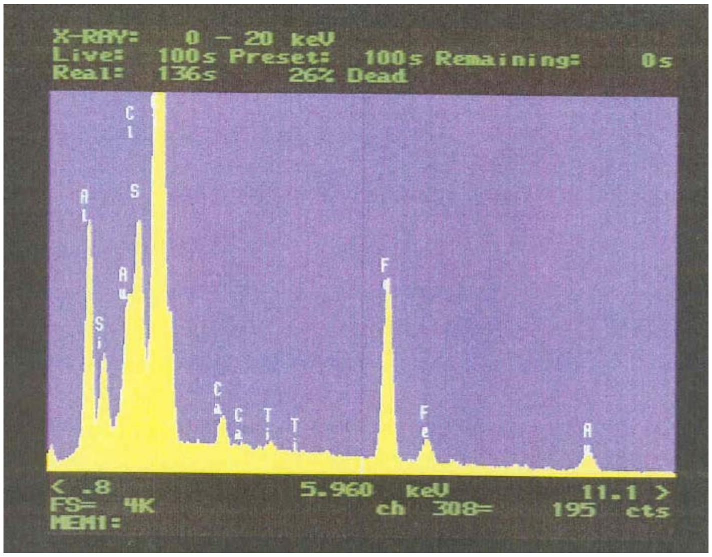

Fig. 3 EDX investigation of the aluminum seal at a Spot with Findings.

The arising suspicion at one sampled spot on intercrystalline corrosion in other places has not been confirmed later.

RESULTS

Several spots of deposition could be found on and beside the seal area. The thread blind holes are very often the starting point or the source of the deposits. In preparation of underwater use, these threads must be protected against corrosion because due to manufacturing there is no protecting nickel coat existing. The ceramic paste mentioned above is used to prevent corrosion in the threads.

Partly, the borders of the deposition areas can clearly be identified as result of drying process. On the spots of the aluminum coated seal damaged by pitting scars, the appearance of Cl and Fe is significant (Fig. 3). In the micrograph of the aluminum coat of the seal, pitting could be detected at the damaged spots. This was already seen before with a magnifying glass (Fig. 1).

Both at obviously damaged spots and undamaged spots, defects in the sheet aluminum in form of seams were detected.

Reasons for "Leakiness"

The measurement of pitting corrosion showed depths up to 0.025 mm. The deposition on the seal, mainly near the tapped blind holes, reached a thickness of nearly 0.020 mm.

The presence of Cl and Fe especially in the deposition borders leads to the termination of passivation of the aluminum and makes pitting possible.

Both the pitting areas and the depositions are sufficient – due to their geometry - to allow diffusion processes to cause insufficient tightness during the He-tightness test. Thus, also an Ag-seal would not have protected against leakiness resulting from the thickness of depositions during the applied drying procedure. If pitting and depositions appear together, the leak rate seems to be higher. If there are only depositions, it can be assumed that the leakage rate is some lower.

The formation of depositions can be explained by transport processes of the ceramic paste and by corrosion products from the tapped blind holes during evacuating and drying of the inner cask room under vacuum conditions/saturated steam state. The found Cl content could also come from the elastomeric seal and Fe with high probability from not Ni-coated tapped blind holes.

Seams in the sheet aluminum of the seal can be responsible for the insufficient He-tightness, too. However, this error source seems to be rather improbable at the moment since such defects have also been determined in visually intact seal areas by micrograph.

Finally, this can only be clarified after a comparison with an already used and tight seal.

Others: Fuel- and Cask State

The aim of the investigations of the SNF casks was to find the reasons for the insufficient tightness and a new successful sealing of the cask. Nevertheless, in this connection some other assessments of the general state of the open cask and an evaluation of the behavior of the cask components and fuel elements after a longer interim storage had been possible.

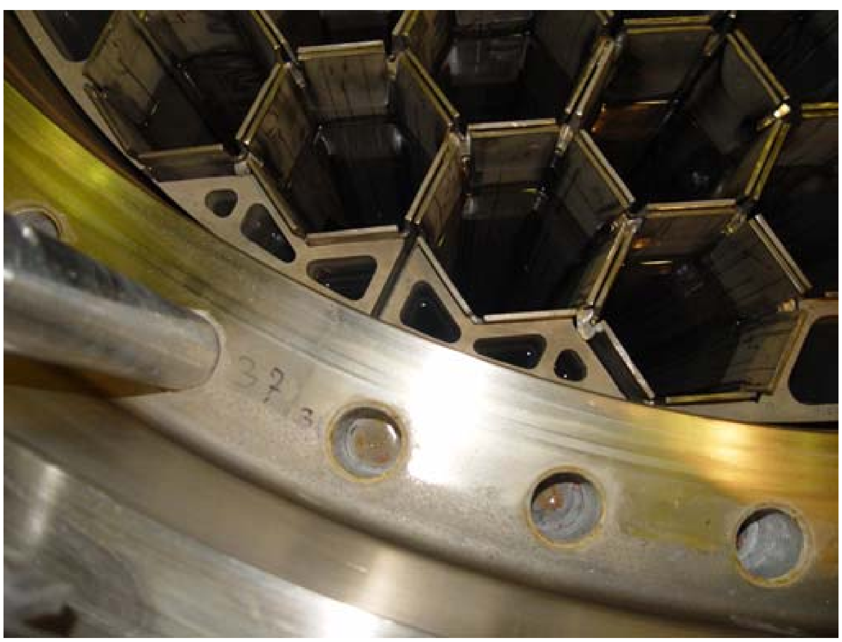

Fig. 4 View into the SNF cask – again unloaded after 3.7 years

The SNF casks which have been opened again till now, after different long storage times, (Table II) showed no recognizable negative changes of the inner state, i.e. the loaded fuel and the cask components. The storage time of 3.7 years maximum till the reopening is relatively short in comparison with the storage time of approx. 14 years [4] for the INEEL[e]-CASTOR V/21. However, the good visual state of the inventory and cask components was impressive.

The state of the basket for the spent fuel elements did not show indications of wear, corrosion or other changes (Fig. 4). The nickel-plated inside surface of the casks and the sealing area of the primary and secondary lid were bright blast except of the described depositions on the sealing area of the primary lid, and it looked like just prepared for first use. There were only indications of corrosion (rust) in the tapped blind holes which were not protected by a nickel coat and near them on the cask flange.

The visual state of the spent fuel elements showed no unusual signs and no perceptible changes compared with the first loading. Additional depositions, new pollutions, remarkable quantities of loose crud have not been detected.

The krypton-85 activity concentration was not determined during the first loading, so there are no reference values for the values before and after recooling (Table III).

However, from the measured values and their small changes, it can be concluded that the tightness state of the fuel elements has not or hardly changed.

There were no indications of damages after the inspection of the primary and secondary lid bolts. According to use requirements, all bolts were in a good state. Any damages by corrosion processes have not been noticed.

CONCLUSION

According to the collected knowledge, a tightness procedure had to be selected which definitively prevents depositions on the seal area of the sealing components. A damage of the passivation layer by higher concentration of Cl and Fe had to be avoided. Pitting formation by local electrochemical elements had to be stopped, too. All these problems were solved by modifying the already used but for a time not applicable procedure with an underwater-pressed metal seal. By immediate positioning of the primary lid under water on the proved and clean sealing area, the development of depositions is prevented. The development of local electrochemical elements between seal and sealing area is prevented, too.

A possible water inclusion between high-grade steel coat and outer aluminum layer of the metal seal is tolerated in this case.

With the now selected clearance procedure it is possible to detect bigger faults in advance by executing a pre-tightness test of the primary lid sealing before draining and drying the cask inner room. In case of bigger leakages, faults can be eliminated and a time and cost intensive recooling can be avoided.

The multitude of improvement measures for the cleanness in all areas of cask clearance will certainly contribute to good results in the He-tightness test. The high cleanness level should be maintained although the reasons for the failed tightness tests turned out to be other ones.

So far, the qualification of the loaded SNF casks for intermediate storage has been confirmed.

REFERENCES

1 Bundesamt für Strahlenschutz "Genehmigung … Zwischenlager Nord (ZLN) … Transportbehälterlager…", Salzgitter (1999).
2 Bundesamt für Strahlenschutz "Zulassungsschein D/4311/B(U)F-85 (Ref. 6), Salzgitter (2003)
3 G. BOEHME, J. GRIEP, S. SCHWEITZER "$157 \cdot 10^6$ MWh (electric) with 6217 Fuel Elements - Intact, Leaky, Damaged and Experimental. And now?" Energiewerke Nord GmbH, WM '00 Tucson (2000).
4 R. M. KENNEALLY (NRC), J. H. KESSLER (EPRI), "Behavior of Spent Fuel and Safety-Related Components in Dry Cask Storage Systems", Transactions, SmiRT 16, Washington DC, August 2001.

FOOTNOTES

[a] EWN: Energiewerke Nord GmbH – a German Company (former Nuclear Power Plant in Dismantling and Conversion to an Industrial Site)

[b] It is insinuated here that the leaving of approx. 1% of the krypton-85 activity inventory of one fuel rod represents a symptom for an inadmissible fuel rod leak.

[c] SEM/EDX: Scanning Electron Microscopy/Energy Dispersive X-Ray Spectrometer.

[d] A special ceramic paste.

[e] INEEL: Idaho National Environmental and Engineering Laboratory.