VERIFICATION OF ACTIVITY RELEASE COMPLIANCE WITH REGULATORY LIMITS WITHIN SPENT FUEL TRANSPORT CASKS

Annette Rolle **Bernhard Droste**

Sven Schubert **Ulrich Probst** **Frank Wille**

BAM Federal Institute for Materials Research and Testing, Berlin, Germany

ABSTRACT

Admissible limits for activity release from Type B packages for spent fuel transport specified in the IAEA regulations (10^{-6} A_2 per hour for normal and A_2 per week for accident conditions of transport) have to be kept by an appropriate function of the cask body and its sealing system.

Direct measurements of activity release from the transport casks are not feasible. Therefore the most common method for the specification of leak-tightness is to relate the admissible limits of activity release to equivalent standardized leakage rates. Applicable procedure and calculation methods are summarized in the International Standard ISO 12807 /2/. BAM as the German competent authority for mechanical, thermal and containment assessment of packages liable for approval verifies the activity release compliance with the regulatory limits.

Two fundamental aspects in the assessment are the specification of conservative design leakage rates for normal and accident conditions of transport and the determination of release fractions of radioactive gases, volatiles and particles from spent fuel rods.

Design leakage rates identify the efficiency limits of the sealing system under normal and accident conditions of transport and are deduced from tests with real casks, cask models or components.

The releasable radioactive content is primarily determined by the fraction of rods developing cladding breaches and the release fractions of radionuclides due to cladding breaches. The influence of higher burn-ups on the failure probability of the rods and on the release fractions are important questions.

This paper gives an overview about methodology of activity release calculation and correlated boundary conditions for assessment.

INTRODUCTION

The IAEA regulations /1/ specify the permitted release of radioactivity from Type B packages under normal and accident conditions of transport (NCT and ACT) in terms of activity per time. Applicants for an approval have to demonstrate the compliance with the required limits. BAM verifies on the basic of the International Standard ISO 12807 /2/ the correct release calculation within the applicant’s package design safety analysis report. It is important that all basic data used for calculation are reliable and conservative. When assessing spent fuel transport casks attention has to be applied on the specification of the design leakage rates and the fractions of radionuclides which are available for release from the containment.

SEALING SYSTEM

Several package designs have been developed for the transport of various types of spent fuel assemblies. The closure system of most of the casks is comparable: two flanges with inserted bolts and a metallic or elastomeric gasket in-between. An additional elastomeric seal is required to create the necessary volume for leak tests via test ports. The double jacket metal seals used for the cask designs in Germany consist of a circular spiral spring encased in two jackets; the inner layer is made of stainless steel, the outer layer mad of aluminium or silver. Material for elastomeric seals used usually is fluorocarbon- rubber or ethylen-propylen-dien-monomer-rubber.

LEAKAGE MECHANISM AND MODE OF CALCULATION

Direct measurements of radioactive release from Type B packages are not feasible. Therefore the common method for the specification of leak-tightness is to relate the admissible limits of activity release to equivalent standardized leakage rates.

Releasable radioactive material might be in form of gas, liquid, solid or a combination of these, and can be released through leaks or in case of elastomeric seals additional by permeation. Miscellaneous physical-mathematical models are available for different leak designs and kinds of fluid. In the field of package design testing the “one capillary leak model” has become accepted /3/. The maximum permissible activity release rate can be expressed in terms of a maximum permissible capillary leak diameter. The following equations describe the flow rates trough a capillary.

Gas flow

The modified Knudsen equation is valid for the whole range of molecular, transitional and viscous laminar gas flow.

$$Q = \frac{\pi}{128} \cdot \frac{D^4}{\mu \cdot a} \cdot \frac{(p_u^2 - p_d^2)}{2} + \frac{\sqrt{2\pi}}{6} \cdot \sqrt{\frac{R \cdot T}{M}} \cdot \frac{D^3}{a} \qquad [\mathrm{Pa\ m^3\ s^{-1}}]$$

A – capillary length [m]
D – capillary diameter [m]
M – relative molecular mass [kg mol^{-1}]
p_d – downstream pressure [Pa]
p_u – upstream pressure [Pa]
R – universal gas constant [$R = 8{,}31 \cdot \mathrm{mol}^{-1}\,\mathrm{K}^{-1}$]
T – temperature [K] (fluid)
μ – dynamic viscosity of the fluid [Pa s]

Together with the gas also particles could be carried, but below a standardized leakage rate of $Q_{SLR} < 1E\text{-}4$ Pa m^3 s^{-1} the release of fuel or crud particles is negligible due to a choking of the capillary /4/.

Liquid flow

Poiseuilles law is applied for the flow of liquids through a capillary.

$$L = \frac{\pi}{128} \cdot \frac{D^4}{\mu \cdot a} (p_u - p_d)$$

A cask design at conventional temperature and pressure conditions is considered to be liquid-tight below a standardized leakage rate of $Q_{SLR} = 10^{-5}$ Pa m³ s^{-1} /5/.

Permeation

Permeation of radioactive gases through metals is negligible for release calculation /6/. If elastomeric seals are used gas permeation is an additional release path way:

$$Q_P = P \cdot \frac{A}{l} \cdot \Delta p$$

P – coefficient of permeation [m² s^{-1}]
Δp – partial pressure difference [Pa]
L – thickness of the permeable material [m]

The applicant has to demonstrate, that the design leakage rates specified for the miscellaneous conditions of transport not exceed the maximum permissible standardized leakage rates calculated. The International Standard ISO 12807 /2/ gives the basic for the mode of calculation. There are seven substantial steps:

Step 1: determination of the total releasable activity
Step 2: determination of the equivalent A_2
Step 3: determination of the permissible activity release rate
Step 4: determination of the activity release due to permeation
Step 5: determination of the maximum permissible volumetric leakage rate
Step 6: determination of the maximum permissible equivalent capillary leak diameter
Step 7: determination of the permissible standardized leakage rate

The specified design leakage rates may not exceed the permissible standardized leakage rates.

DESIGN LEAKAGE RATES

Design leakage rates identify the efficiency limits of the sealing system under RCT, NCT and ACT and are deduced from tests with real casks, cask models or cask components, for example with flange assemblies. Component tests are important for the demonstration of the worst case conditions and for statistical validation.

The impacts resulting from the regulatory mechanical and thermal tests (see Tab. 1) can lead in deformations or displacements of cask components involving an unloading and/or a movement of the lids and/or seals; a rotation or a lateral sliding. As a consequence the leakage rate can increase.

Table 1. Impacts which can influence the tightness of the sealing system

IAEA Regulations, TS-R-1 /1/	loadings to be considered
Routine Conditions of Transport (RCT) § 612, 615	▪ acceleration (e.g. 2 g) in radial and axial directions ▪ operational temperature and pressure
Normal Conditions of Transport (NCT) § 722	▪ free drop from a height of 0.3 m (mass ≥ 15 t)
Accident Conditions of Transport (ACT) § 726 – 728	cumulation of: ▪ free drop from a height of 9 m ▪ 1m puncture test ▪ 800°C, 30 min thermal test separately: ▪ water immersion test

<u>Metallic seals</u>

RCT: If metallic seals of the Helicoflex-type /9/ are used, BAM accepts for RCT only a design leakage rate above 1E-8 Pa m^3 s^{-1}.This is the specified leakage rate that attests the sealing system the regular assembly status, and has to be demonstrated by a helium leak-test after assembling of the lid system, before delivery and before any transport by a Helium leak- test.

NCT and *ACT*: For NCT and ACT the design leakage rates can be higher depending on the test results.

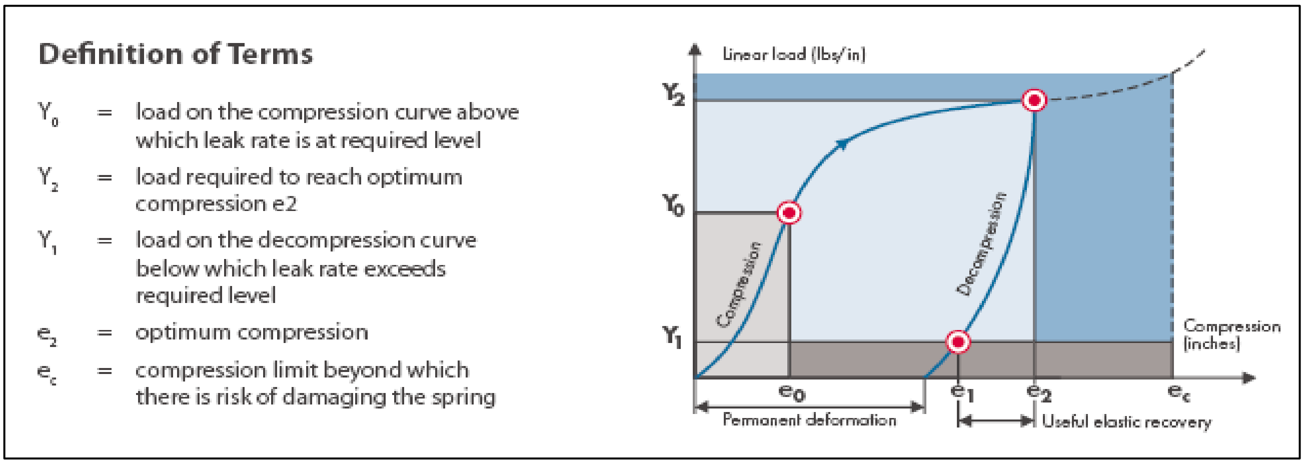
The vertical 0.3m drop (package mass ≥ 15 t) under NCT can cause short-term elastic deformation of the bolts that can lead to a short time relaxation of the lid flange. Component tests have shown that after a repeated compression provided that no seal dislocation occurs the specified leakage rate of $Q_{SLR} < 1E\text{-}8$ Pa m^3 s^{-1} is achievable again. Leakage rates measured after a repeated compression including a seal displacement (rotation) are considerably higher /7/. For implementing a value of 1E-8 Pa m^3 s^{-1} as design leakage rate for NCT, applicants have to demonstrate that there was a sufficient clamping load on the seal at any time during the drop event to prevent a movement of the seal. Otherwise the higher values have to be used.

The design leakage rates for ACT currently specified for spent fuel transport and storage cask designs used in Germany are deduced from measurements after drop tests with real casks, numerical analyses of influences of thermal impacts, and from component tests simulating a quasi-static lateral and a radial seal displacement. For verification of the specified values BAM requires additional tests to investigate the sealing behaviour after a dynamic lateral displacement of the seal.

When assessing the mechanical and thermal analyses BAM recalculate the maximum widening between the flange surfaces of the sealing system, which can be caused for example by deformation of bolts, bending of the lid and/or the cask body flange as much as by different thermal expansion of lid and cask body material due to different coefficients of thermal expansion or inhomogeneous heating under thermal impact. For RCT it is important that a possible widening may not exceed the useful elastic recovery of the seal, called $r_{u,}$ because above this range the efficiency of the seal is exhausted. Above a gap of r_u the standardized leakage rate exceeds the value of 1E-8 Pa m^3 s^{-1} and does not meet the specification anymore.

All specified design leakage rates for NCT and ACT can only be accepted if the possible widening after_impact loading is smaller than r_u (a short-term decompression above r_u is possible). For illustration of r_u see Fig. 1.

Results of BAM investigations show an influence of temperature and time on r_u /8/. Applicants are requested to provide a test program for justification conservative values for r_u. Until then BAM requires the use of a safety factor by permitting only a maximum flange distance widening of 0.5 r_u.



Definition of Terms

Y_0 = load on the compression curve above which leak rate is at required level

Y_2 = load required to reach optimum compression e2

Y_1 = load on the decompression curve below which leak rate exceeds required level

e_2 = optimum compression

e_c = compression limit beyond which there is risk of damaging the spring

Figure 1: Compression and decompression cycle of the Helicoflex - seal for illustration of the useful elastic recovery ($r_u = e_2 - e_1$) /9/

Elastomeric seals

Elastomeric seals show a very advantageous behaviour under mechanical stresses and they are able to compensate flange dislocations in a wide range. The design leakage rate for the use of elastomeric seals under RCT, NCT and ACT in packages used in Germany specified with $Q_{SLR} \leq$ 1E-5 Pa m^3 s^{-1} is mainly limited by permeation.

A critical point for these gasket materials is the behaviour at low temperatures. If new mixtures of elastomeric materials are used, BAM requires the determination of the critical low temperature for failure. BAM started an own research program about temperature and time depending behaviour of new elastomeric material mixtures /10/.

RELEASABLE RADIOACTIVE CONTENT

Spent fuel rods contain isotopes in form of gas, volatiles and fuel particles, which are only releasable through a breach in the cladding. Additional particles of Chalk River Unidentified Deposit, called CRUD, on the rod surface can contribute to the releasable content.

The activity of the radionuclides, which has to be considered for release calculation, can be calculated by using a computer code, like ORIGEN /11/.

The release of activity from the fuel rods into the cask internal cavity depends on the chemical and physical properties of the radionuclides, on the fuel characteristic, the conditions of the cladding tubes and on the amount and properties of CRUD adherences.
Based on experiments and examinations, conservative values for the prediction of the source term for leakage rate calculation were deduced in a NUREG report /12/ (Tab. 2).

Table 2. Release fractions used to predict the source term for release calculation from spent fuel transport casks /12/

		NCT	ACT
fraction of gases that are released due to a cladding breach	f_G	0.3	0.3
fraction of volatiles that are released due to a cladding breach	f_v	$2 \cdot 10^{-4}$	$2 \cdot 10^{-4}$
fraction of fuel fines that are released due to a cladding breach	f_F	$3 \cdot 10^{-5}$	$3 \cdot 10^{-5}$
fraction of CRUD that spalls off of rods	f_c	0.15	1.0
fraction of rods that developing cladding breaches	f_B	0.03	1.0

These values are determined for burn-ups of 33 to 38 GWd/tU and they have been applied also for the safety assessment of transport casks in Germany. In the last years fuel is being driven to higher burn-ups up to 65 GWd/tU and more. BAM requires applicants to investigate the possible influence of a higher burn-up on the release fractions.

Experience from actual approval procedures are discussed in the following:

<u>Release fraction of gases and volatiles:</u>
The generation of Kr-85 and H-3, that dominate the gas release, depends straight proportional on the burn-up. The release of fission gases from fuel occurs through a net of grain boundaries and depends on temperature and fuel microstructure. The release increases disproportional with a higher burn-up. Measurements have shown, that a fraction of 30% is still a conservative value up to a burn up of 100 GWd/tU. Up to a burn up of 80 GWd/tU even a release fraction of 15% is justifiable /13/.
For volatiles, dominated by Cs-137, Cs-134, Ru-106 and Sr-90, BAM accepts a release fraction of 0.02% as conservative estimate, also for higher burn-up considering the very low vapour pressures of the relevant compounds.

<u>Release fraction of particles:</u>
Changes in the fuel characteristic after a higher burn-up can affect the particle size distribution. Also the amount and the characteristic of CRUD fines are impacted by higher burn-ups. The release fraction of particles is at the moment of minor importance for BAM, because design leakage rates of spent fuel transport and storage casks equipped with metal gaskets are in most cases small enough to prevent any particle release.

Fraction of rods developing cladding breaches

The number of rods developing cladding breaches under NCT is a very important parameter for the release calculation. Such breaches enable not only the release of gas, volatiles and fuel particles into the cask interior, but also an increase of the internal cavity pressure. In this context cladding breaches imply only fine cracks, in contrast to the criticality assessment domain where more extensive damages like fuel rod breakage or ruptures are of main interest.

Higher burn-ups cause a higher embrittlement of the cladding material by up taken hydrogen, and a higher tendency to hydride reorientation as well as a growing oxide layer and coalescences of fuel and cladding material /14/.

The failure rate of the claddings under ACT for the containment assessment currently is also for less burn-ups assumed with 100%, due to a lack of experimental results. Up to now there was no need for a more detailed examination with reduction of the failure fraction for ACT in mind, because the release calculations for most of the cask designs that had been assessed show a sufficient distance to the required limit. For criticality safety assessment other considerations with respect to fuel rod failure mechanisms and rates are relevant /15/.

The influence of the degraded rod properties by higher burn-ups on the failure probability of the claddings under NCT, for example after a 0.3 m drop, is still an open question.

Applicants are requested to present additional evidences about a conservative value of the fraction of rods developing cladding breaches under NCT. In case of a lack of additional analysis BAM either limits the number of higher burn-up spent fuel assemblies permitted for one loading or checks if there is a sufficient safety margin in calculation (for certain designs it is possible to meet the release limits even if a failure rate of 100% under NCT is assumed). For very high burn-ups BAM currently requires an encapsulation of the fuel rods.

CONCLUSIONS

A standardized method for release calculation through a capillary leak is given by the International Standard ISO 12807 /2/. For cask designs with a high leak-tightness (standardized leakage rates 1E-5 Pa m^3 s^{-1}) release calculation can be reduced on the release of gases or volatiles, because the corresponding capillary diameters are small enough to prevent a release of liquids or particles.

The design leakage rates for NCT and ACT, which are determined from tests with casks or cask components, must not exceed the calculated permissible standardized leakage rates. When deducing conservative values for the design leakage rates it is important to consider all possible stresses and its impact on the sealing system. BAM currently requires additional tests to demonstrate the sealing behaviour after a dynamic lateral displacement of the seal.

If metallic seals are used a widening of the sealing system caused by deformation of cask components must not exceed the useful elastic recovery r_u of the seal. The influence of time and temperature on r_u has to be analysed.

There is a clear tendency to spent fuel with higher burn up, and the possible influence on the release fractions for the miscellaneous radionuclides has to be investigated. The release fractions for gases and volatiles determined for burn-ups about 33 to 38 GWd/tU are also applicable for higher burn-ups up to 80 GWd/tU. Higher burn-ups furthermore cause changes in the rod characteristic, which can influence the failure probability of the rod claddings under NCT.

REFERENCES

/1/ Regulations for the Safe Transport of Radioactive Material, 2009 Edition, International Atomic Energy Agency (IAEA), No.TS-R-1

/2/ International Standard ISO 12807:1996, Safe transport of radioactive materials - Leakage testing on packages

/3/ H. Kowalewsky: Modelling Contents and Leaks- Advantages and Limitations, PATRAM 1995, Las Vegas, USA

/4/ J. Higson, R. J. Moultan, R.A. Vaughan: Containment of radioactive powders by seals, 9th PATRAM 1989, Washington, USA

/5/ Takashi Miyazawa at all: Study on water leak-tightness of small leaks, PATRAM 2001, Chicago, USA

/6/ Kowalewsky, H., Becker, S., Heumos, K.: Prüfverfahren zur Bewertung der Dichtheit von Umschließungen radioaktiver Stoffe, Abschlußbericht zum Forschungsvorhaben des BMI, Nr.679, BAM, Berlin, 1983

/7/ S. Schubert, U. Probst, H.-P. Winkler: Behaviour of metallic seals in CASTOR-casks under normal and accidental conditions of transport: qualification requirements, Packaging Transport, Storage & Security of Radioactive Material, Vol. 20, 2009, No 4, pp. 179-182

/8/ U. Probst et al.: Investigation of seal effects according to axial compression variation of metal seals for transport and storage casks, Packaging Transport, Storage & Security of Radioactive Material, Vol. 19, 2008, No 1, pp. 47-52

/9/ Garlock Sealing Technologies, Helicoflex High performance sealing, Saint-Etienne

/10/ M. Jaunich, W. Stark, D. Wolff: A new method to evaluate the low- temperature function of rubber sealing materials, Polymer Testing 29 (2010) pp 815–823

/11/ ORIGEN-2.1;Isotope Generation and Depletion Code, RISC CODE PACKAGE CCC-371, Oak Ridge Nation Laboratory, Oak Ridge , Tennessee, USA, August 1991

/12/ NUREG/CR-648, UCRL-ID-124822, Containment Analysis for Type B Packages Used to Transport Various Contents, B.L. Anderson, R.W. Carlson, L.E. Fischer, Livermore, USA, 1996

/13/ E. Weber: Brennelemententwicklung im Zeichen der Nachhaltigkeit, atw, 50.Jg. (2005), Heft 11, S.645-651

/14/ R.E. Einziger, C. Beyer: Characteristics and behaviour of high-burnup fuel that may affect the source term for cask accidents, Nuclear Technology, Vol. 159, Aug. 2007, pp 1-13

/15/ V. Ballheimer, F. Wille, B. Droste: Mechanical safety analysis for high burn-up spent fuel assemblies under accident transport conditions, PATRAM 2010, London, UK