Karlsruhe Institute of Technology

Interdisziplinäres Forschungssymposium für die Sicherheit der nuklearen Entsorgung

Interim storage - final disposal interdependences: Progressing fuel rod degradation processes constrain the disposal container optimization

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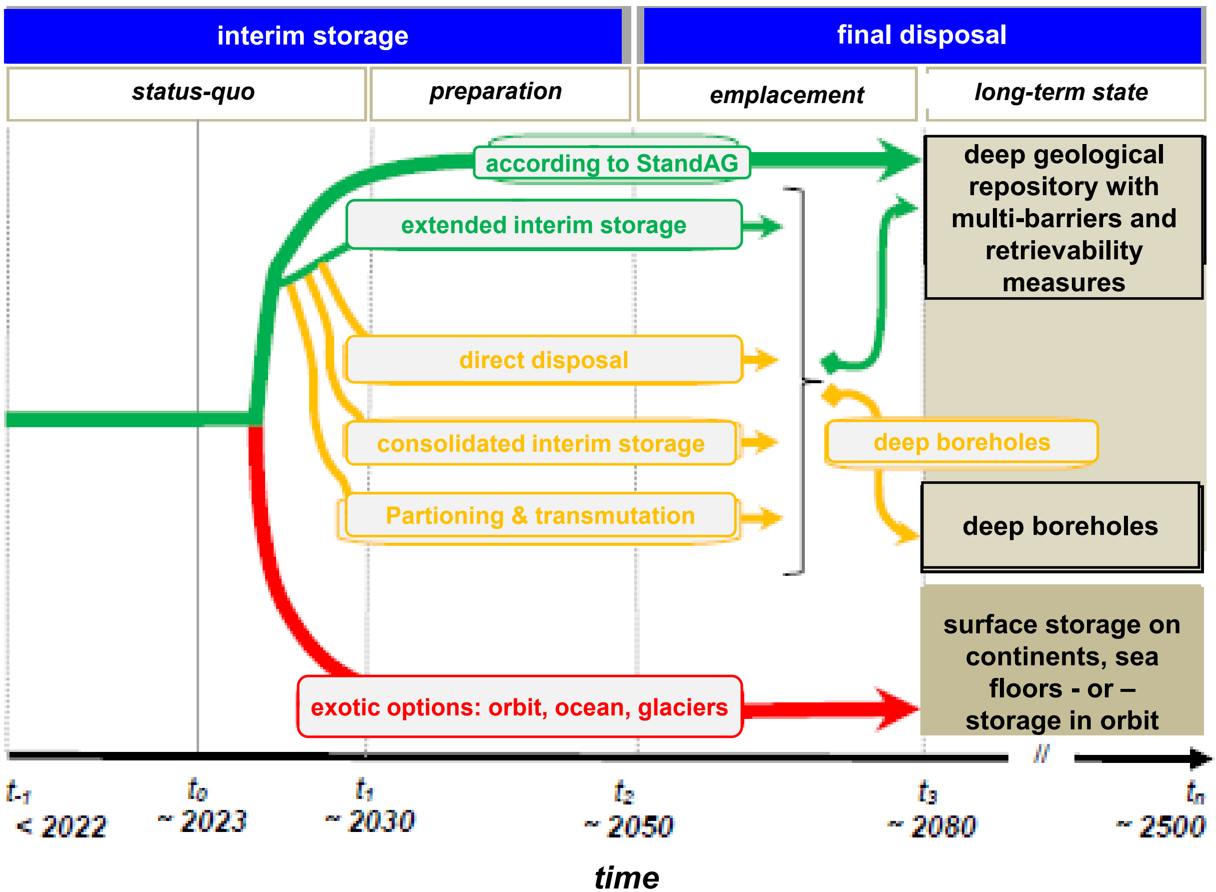
safeND Workshop 06 "Impact of extended interim storage of high-level radioactive waste on the final disposal", Berlin, 14th September 2023

INE

Institut für Nukleare Entsorgung

General perspective on long-term HLW management

“Pathways for waste management” (“Entsorgungspfade”):

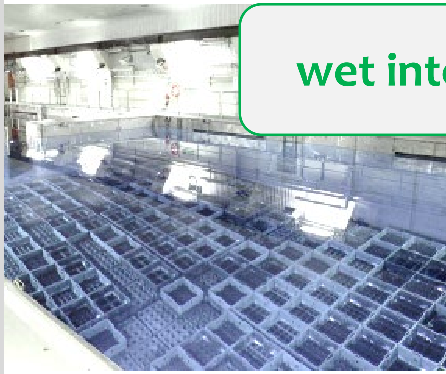


Focus on pathways from present state, i.e. **interim storage** in facilities at land surface → long-term state, i.e. **disposal in deep geological repository (DGR)** with retrievability measures

- disposal of spent nuclear fuel (SNF)
- reprocessing of SNF and afterwards disposal of HLW-glass

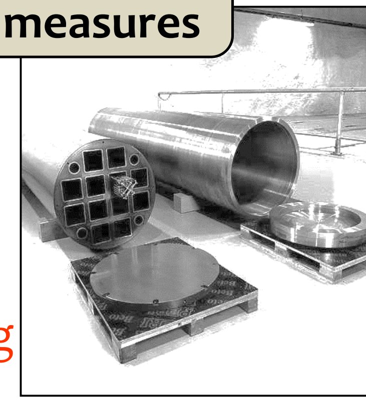
Figure adapted after Scheer, D., Becker, F., Hassel, T., Hocke, P., Leusmann, T., Metz, V. (2023) Trittsicherheit auf Zukunftspfaden? In: Eckhardt, A. et al., “Entscheidungen für die Zukunft: Ungewissheiten bei der Entsorgung hochradioaktiver Abfälle”, Springer.

Pathways from interim storage to final disposal



wet interim storage → **overpack for transfer** → **transfer** → **DGR with multi-barriers & retrievability measures**

final disposal container, design depends on host rock

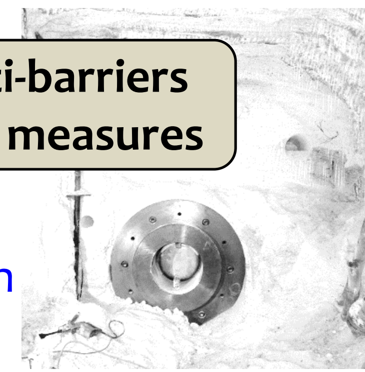


Safety of interim storage relies on permanent functioning of water cooling

Examples: Belgium, Finland, France, Japan, South Korea, Sweden (…)

dry interim storage → **transfer** → **DGR with multi-barriers & retrievability measures**

final disposal container, design depends on host rock



Passive safety provided by dual-purpose casks (DPC, e.g. CASTOR®)

Examples: Belgium, France, Germany, Japan, Switzerland, Ukraine, USA (…)

Image sources: H. Widestrand & P. Schillebeeckx (2016) SF Characterisation Program for Implementation of Repositories, POSIVA (2021) www.posiva.fi/en/index/news/pressreleasesstockexchangereleases/2021/excavationofworldsfirstfinaldepositiontunnels; V. Metz (KIT-INE)

Conditioning critical interface interim / final storage

wet interim storage

transfer

DGR with multi-barriers & retrievability measures

overpack for transfer

final disposal container

loading of transfer cask

conditioning in hot cell

repackaging into final disposal container

dry interim storage

transfer

DGR with multi-barriers & retrievability measures

final disposal container

conditioning in hot cell

repackaging into final disposal container

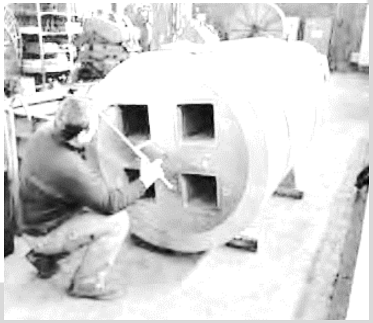
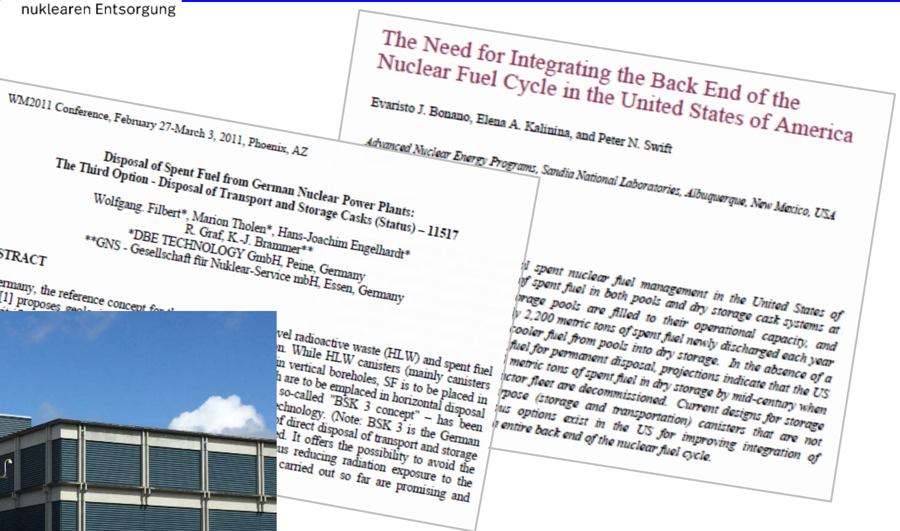


Image sources: H. Widestrand & P. Schillebeeckx (2016); NAGRA, courtesy of L. Johnson (2008); SKB, Technical Report, TR-01-03 (2000); GRS (2008) Endlagerung wärmeentwickelnder Abfälle in Deutschland, GRS-247 (2008); ANDRA (2006) 269 VA Phenomenological evolution of a geological repository; V. Metz (KIT-INE)

Uncertain approach: Use of DPC for final disposal


The Need for Integrating the Back End of the Nuclear Fuel Cycle in the United States of America

Evaristo J. Bonano, Elena A. Kalinina, and Peter N. Swift

Advanced Nuclear Energy Programs, Sandia National Laboratories, Albuquerque, New Mexico, USA

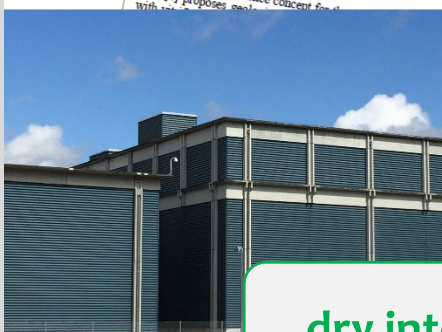
WM2011 Conference, February 27-March 3, 2011, Phoenix, AZ

Disposal of Spent Fuel from German Nuclear Power Plants:
The Third Option - Disposal of Transport and Storage Casks (Status) – 11517

Wolfgang Filbert*, Marion Tholen*, Hans-Joachim Engelhardt*
R. Graf, K.-J. Brammer**
*DBE TECHNOLOGY GmbH, Peine, Germany
**GNS - Gesellschaft für Nuklear-Service mbH, Essen, Germany

ABSTRACT

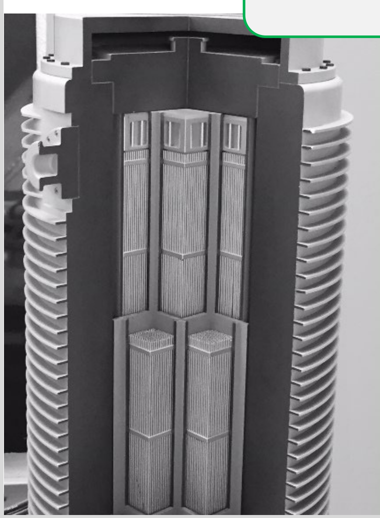
conditioning → e.g. filling of cask with Fe_3O_4 or other material to ensure sub-criticality



dry interim storage

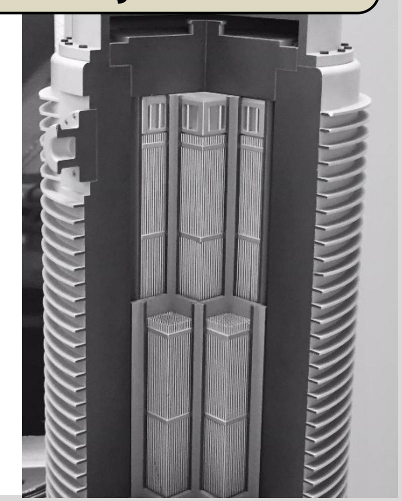
transfer

DGR with multi-barriers without retrievability measures



conditioning in hot cell

no repackaging, however conditioning of inventory



Sources: Bonano et al. (2018) MRS Advances. Filbert et al. (2011) WM2011 Conference. Graf et al. (2012) KTG Tagung. Image source: V. Metz (KIT-INE)

Duration of interim storage

- In general interim storage facilities are allowed to store spent nuclear fuel (SNF) and highly active vitrified waste (HLW) for few decades* years
- but, due to delays in site selection, licensing and construction of deep geological repositories, it will be needed to extend interim storage to longer time intervals, possibly around 100 or more years

- Presently, numerous states finalize their preparation of predisposal activities including detailed design of an interim storage system that should be able to safely preserve SNF and HLW for an extended time interval (more than 100 years), until a suitable final disposal facility is available

  ** In Germany, permits for individual DPC (Befristung der Aufbewahrungsgenehmigung) are limited to 40 years after closure according to AtG §6*

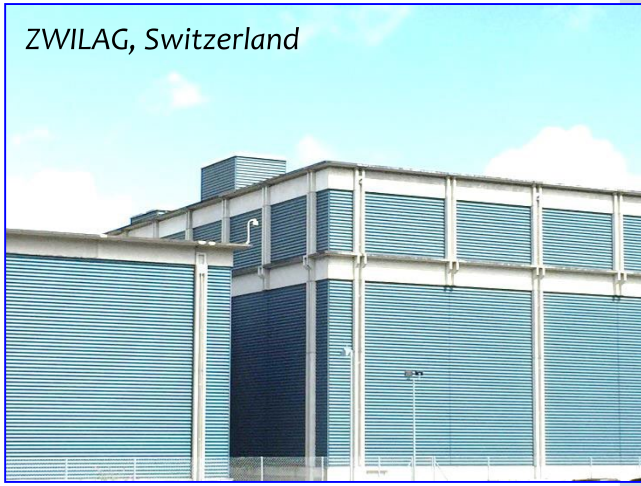
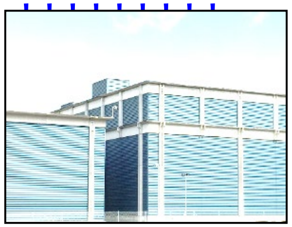


Image source: Metz, V., Schepperle, J. (KIT-INE)

One century of HLW interim storage in Germany



First loadings of CASTOR® THTR/AVR 1992; HLW 1997

expire of licenses for interim storage facilities (AtG §6): AVR Jülich 2013, other facilities 2034 - 2047

prolonged interim storage, successive transport from interim storage sites to conditioning and / or disposal site

1992

2013

2047

1990 | 2010 | 2030 | 2050 | 2070 | 2090

2017

~2046-2068

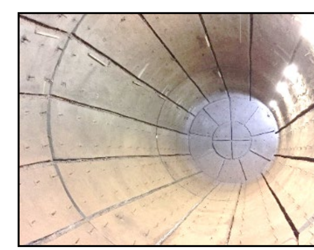
>2090

site selection process for deep geological repository

application / licensing process, building of deep geol. repository

emplacement of SNF/HLW-glass in repository, DGR

closure of DGR (2100?)



Sources: NaPro (2015); BGE (2022) press release 11. Nov. 2022; BASE (2022) press release 14. Nov. 2022; ESK (2023) Verlängerte Zwischenlagerung bestrahlter Brennelemente und sonstiger hochradioaktiver Abfälle in Abhängigkeit von der Auswahl des Endlagerstandorts, 23. March 2023

How long interim storage in surface facilities is secured?

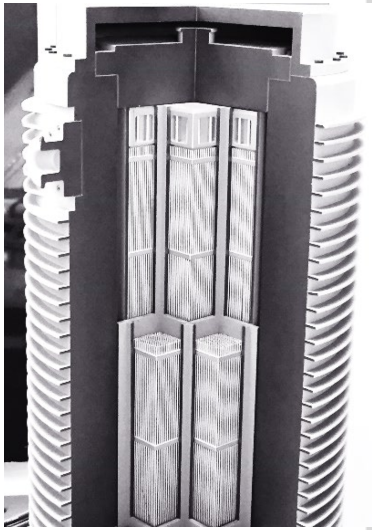
Security issues →

„The dry spent fuel storage facility project for Zaporizhzhya NPP is based on VSC-24 cask design … The storage facility project includes 380 VSCWWER-1000 casks.” (Steinberg et al., 2003)

One artillery shell (or more) destroyed a training building close to the reactors. Two other shells were found in the interim dry storage facility for spent nuclear fuel. *(Speicher, 2022)*

Sources: Speicher, C., 2022 (Neue Zürcher Zeitung, 26. April 2022; Steinberg, N., 2003 (IAEA-CN-102/49); Image sources: Guardian, 2014; IAEA, 2022; Energoatom, 2014

Increasing risks with ageing of cask and waste inventory

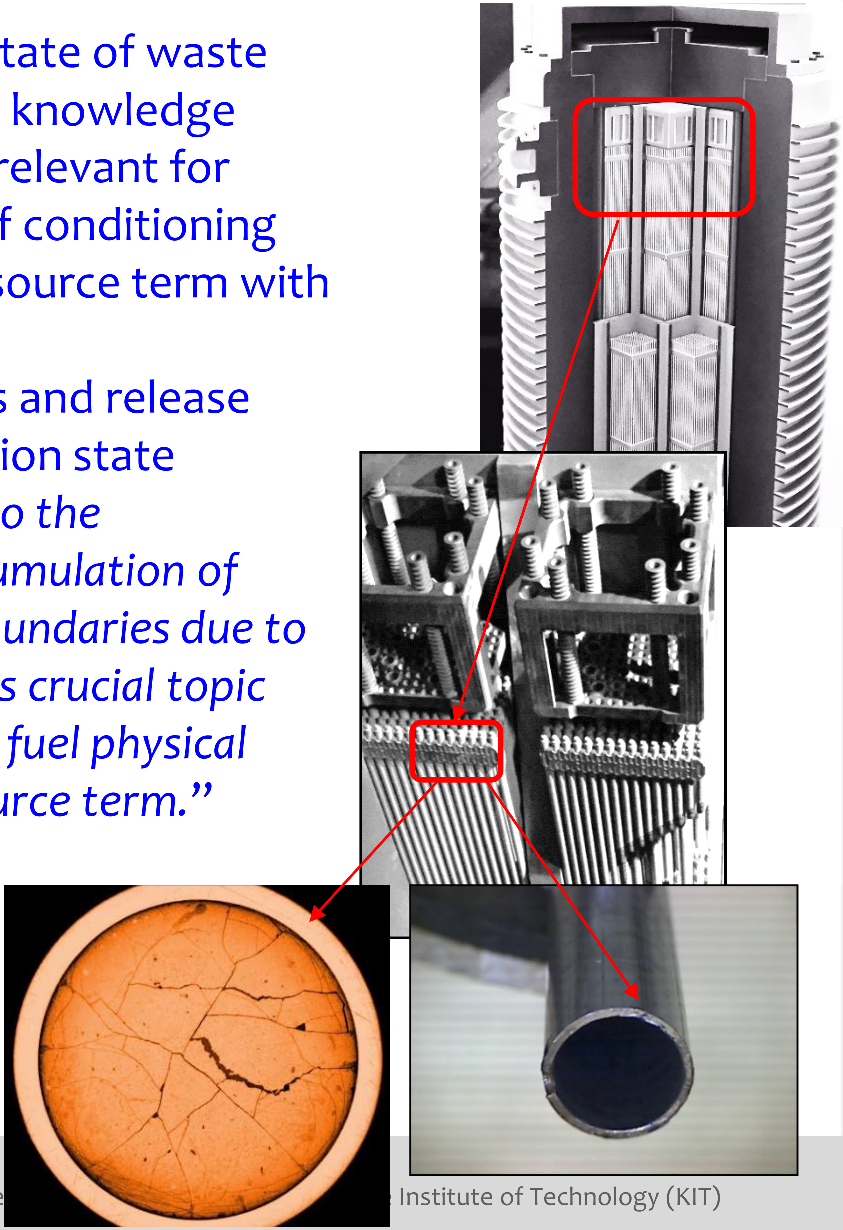


- With time, a continuous degradation of components of DPC (e.g. CASTOR®) and of the waste inventories (SNF and HLW glass) is progressing
- After interim storage, conditioning and repackaging of waste inventories from DPC to final disposal containers is needed
- Integrity of fuel rod cladding and stability of geometrical state of inventory are decisive for any conditioning and repackaging of SNF , in order to prevent any release of volatile radionuclides in the conditioning / repackaging facilities
- In case, the cladding integrity will not be ensured, reloading of fuel assemblies from DPC to final disposal containers will be considerably aggravated
- Degradation state of DPC and of inventories define the requirements for the design of final disposal containers as well as for design of conditioning facility (hot cell)

Sources: ESK (2015): Diskussionspapier zur verlängerten Zwischenlagerung bestrahlter Brennelemente und sonstiger Wärme entwickelnder radioaktiver Abfälle. Entsorgungskommission; ESK (2023) Verlängerte Zwischenlagerung bestrahlter Brennelemente und sonstiger hochradioaktiver Abfälle in Abhängigkeit von der Auswahl des Endlagerstandorts.

Waste degradation state relevant for long-term safety

- For time spans >> 40 years, uncertainty about state of waste inventories is considerably increasing → lack of knowledge
- Degradation state of waste inventory not only relevant for requirements of final disposal containers and of conditioning facilities but also for definition of radionuclide source term with respect to deep geological repository
- Stability of UO_2 matrix, cladding corrosion rates and release rates of radionuclides depend on SNF degradation state
- *"Major degradation of the physical state linked to the accumulation of irradiation damages, to the accumulation of microbubbles, to the de-cohesion of the grain boundaries due to ageing (…) The current lack of knowledge on this crucial topic leads us to conservatively assume that the spent fuel physical state is fully degraded when assessing the RN source term."* Poinssot et al. (2014)



Sources: Poinssot, C. et al. (2014) Journal of Nuclear Science and Technology 39. ESK (2023) Verlängerte Zwischenlagerung bestrahlter Brennelemente und sonstiger hochradioaktiver Abfälle in Abhängigkeit von der Auswahl des Endlagerstandorts.

Degradation of fuel rod cladding

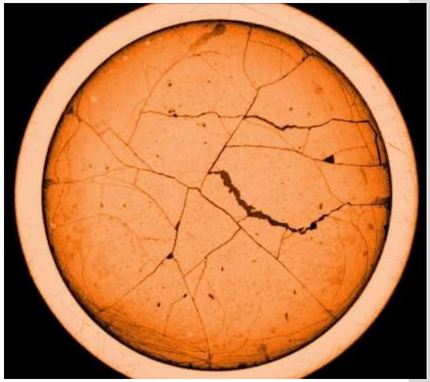
Long-term integrity of the cladding depends on

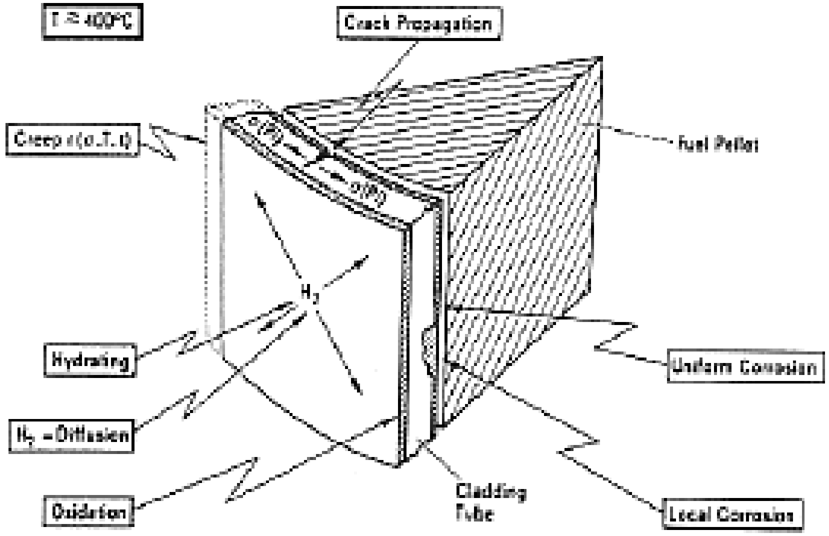
- resistance to ageing effects of the cladding itself
- Zr-hydride reorientation and delayed hydride cracking
- resistance of the cladding to internal pressure
- resistance of the cladding to pellet cladding chemical interactions

→ corrosion with volatile fission products at cladding / fuel interface

- continued swelling of fuel due to He accumulation
- fuel swelling due to oxidation of UO_2 matrix

→ in particular relevant for high burn-up UO2 fuels (HBU-SNF) and spent MOX fuels



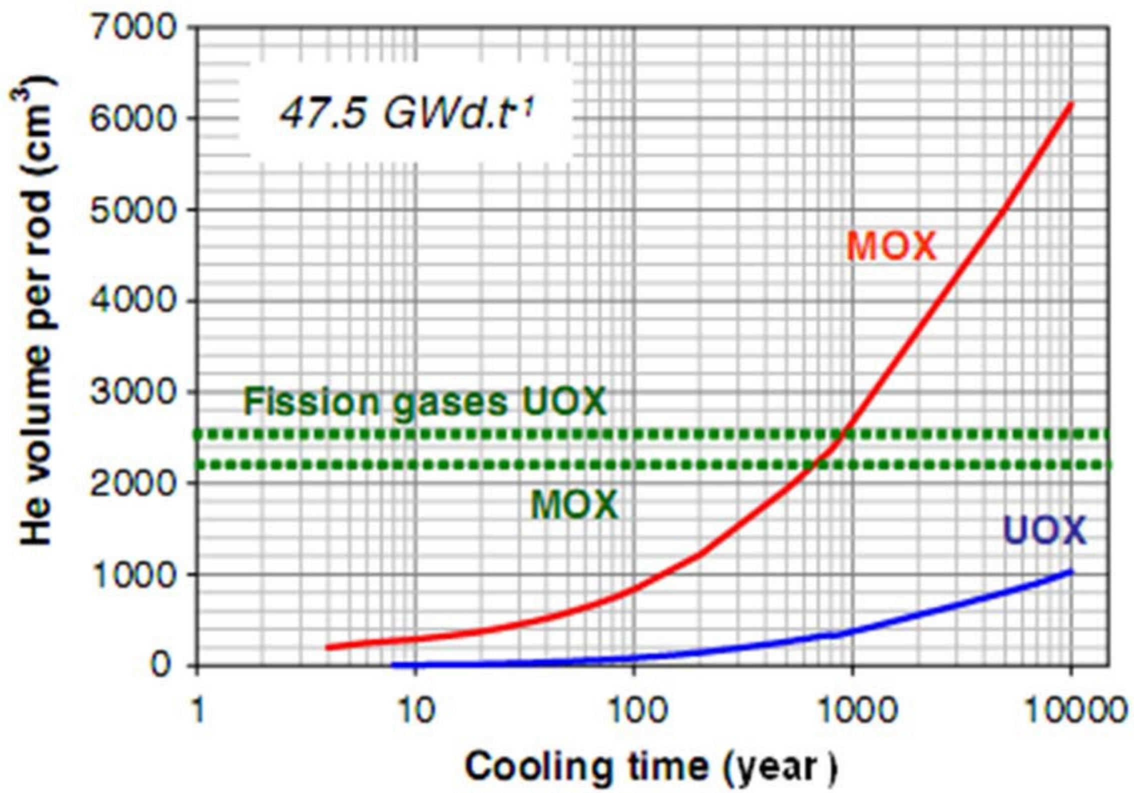


Source: IAEA (2003) Spent fuel performance assessment and research (SPAR-I project), Tecdoc-1343; Spykman (2018) Nucl. Eng. Technol. 50, 313-317

Evolution of gas inventory within fuel rods

Within fuel rods, gas inventory dominated by fission gases (Kr-83, Kr-84, Kr-85, Kr-86, Xe-131, Xe-132, Xe-134, Xe-136) in first centuries

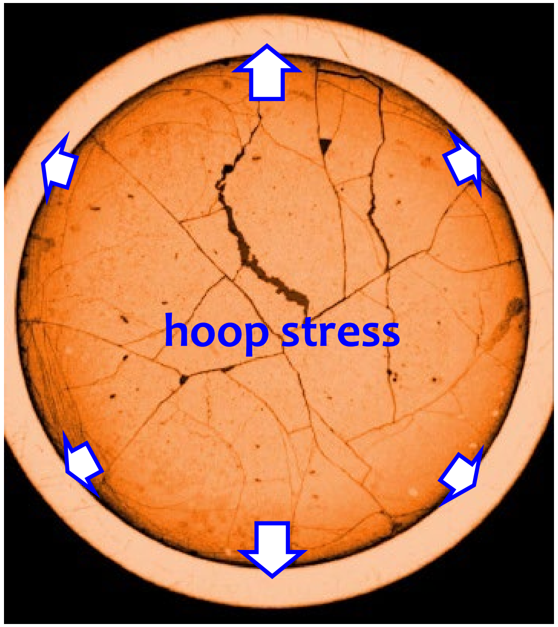
After about ~700 years, gas inventory will be dominated by helium (due to α-decay) – contribution of helium to pressure built-up to be negligible for a period of decades to few centuries

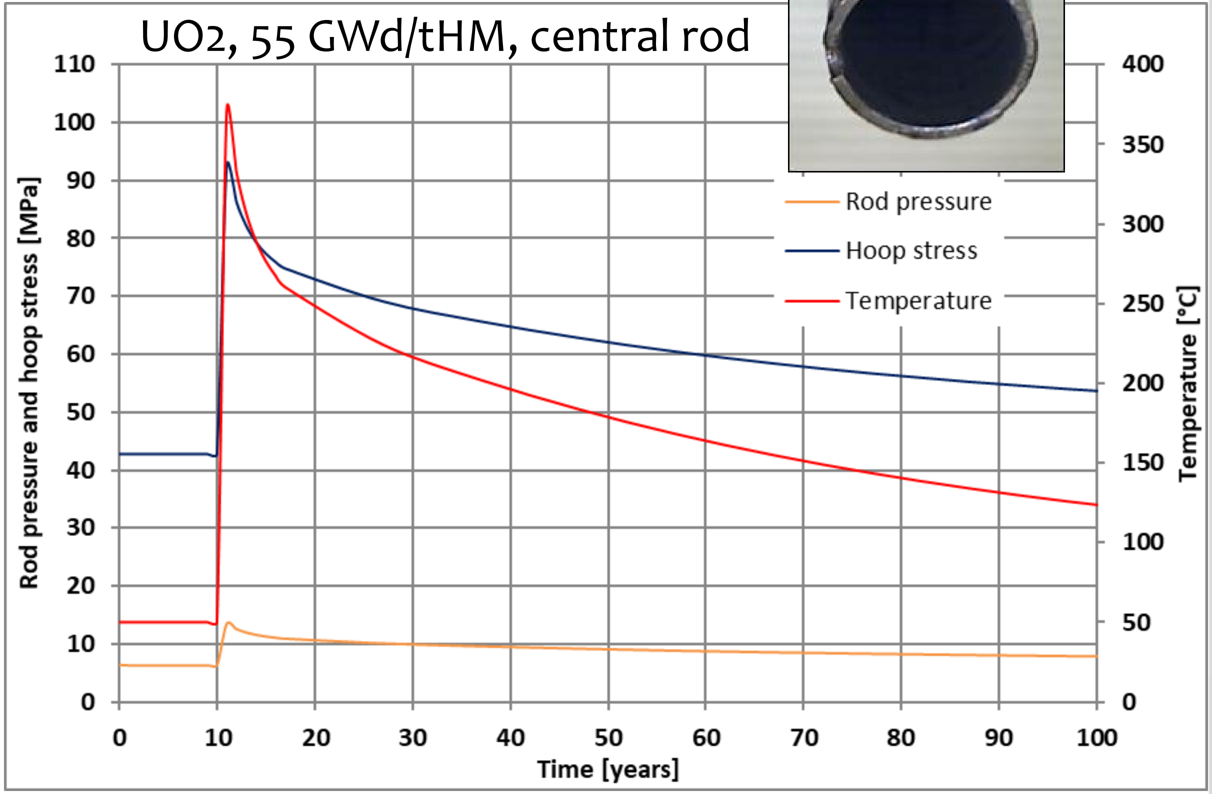


Sources: Poinssot, C. et al. (2014) Journal of Nuclear Science and Technology 39; Gonzalez-Robles, R. et al. (2016), Journal of Nuclear Materials 479.

Variation of hoop stress of fuel rod cladding

Hoop stress of fuel rod cladding and rod internal pressure increase due to fission gas production during irradiation, swelling of UO_2 matrix, temperature effects and others

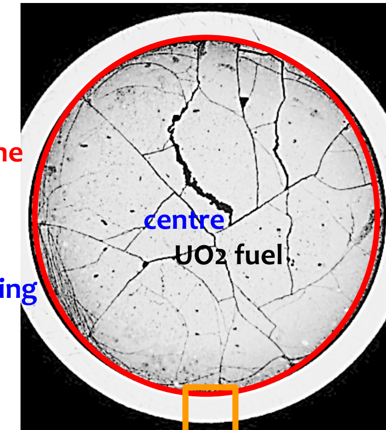




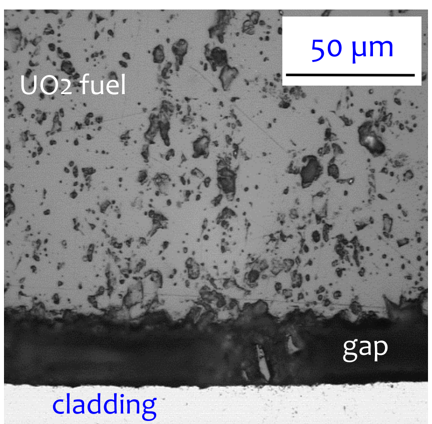
Sources: Rowold, F. (2018) GRS "Open questions on the road to reliable predictions“, GRS workshop “safety of extended dry storage of spent nuclear fuel, Garching; Institut für Nukleare Entsorgung, KIT-INE

Fuel-cladding chemical interaction

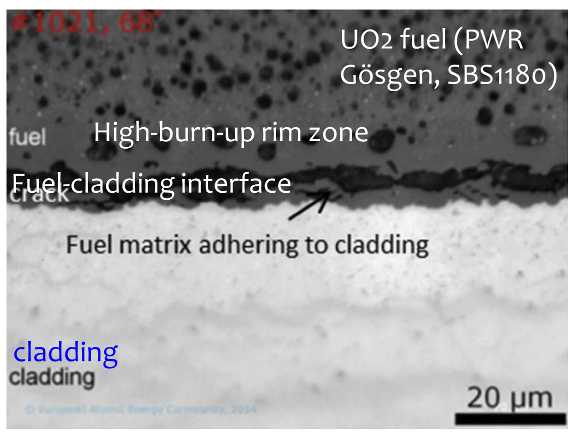
- Due to migration of Cl-36, I-129, Cs-137, Cs-135 from pellet centre to pellet periphery → enrichment in rim zone
- At low burnup, gap between fuel and cladding is still open
- At higher burnup, gap closed
- Cs–U–O–Zr–Cl bearing compounds determined by SEM-EDX, XPS at fuel-cladding interface (e.g. König et al., 2021) → fuel-cladding (chemical) interaction, FCCI



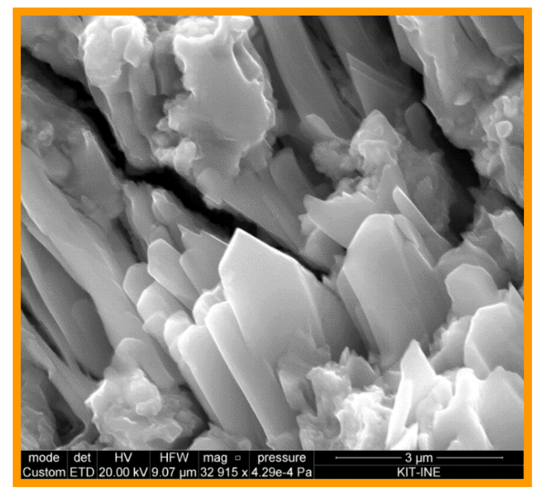
irradiated LWR UO_2 fuels



low BU: 20 GWd/tHM



medium BU: 50 GWd/tHM

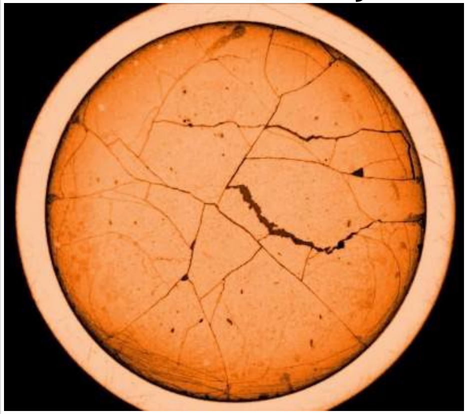


Sources: König, T. et al. (2021) SEDS; Lemmens, K., Gonzalez-Robles, E. et al. (2017) J. Nucl. Mat. 484, 307-323. Fanghänel, T. et al. (2013) Inorg. Chem.

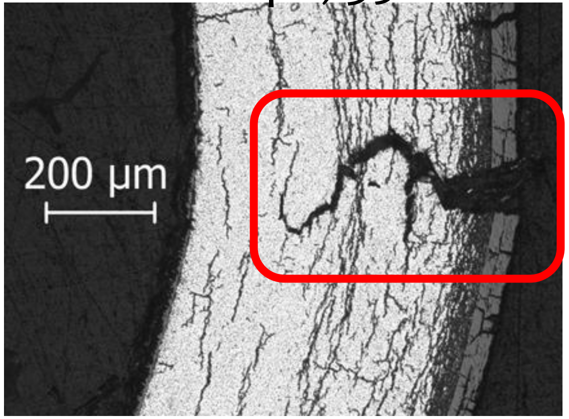
Cracking of Zircaloy due to hydride re-orientation

- At outer Zircaloy surface formation of zirconium oxide , ZrO2, and hydrogen due to contact of cladding with cooling water during reactor $Zr + 2\ H_2O \rightarrow ZrO_2 + 2\ H_2$
- Diffusion of hydrogen into Zircaloy and formation of zirconium hydrides, ZrH_{2-x}, which are oriented parallel to surface, initially
- During interim storage reorientation → radially oriented zirconium hydrides → cracking of cladding

UO2 50.4 GWd/t_{HM} with Zircaloy-4



ZIRLO cladding after H2 loading at 110 Mpa, 350°C



Zircaloy-4 of UO2 64 GWd/t_{HM}

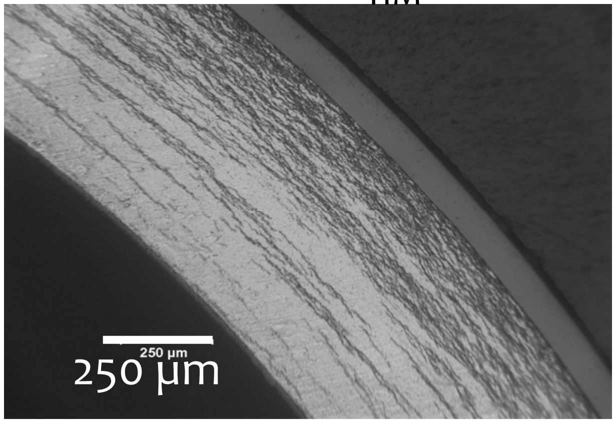


Image sources: M.C. Billone, T.A. Burtseva, J.P. Dobrzynski, D.P. Mc Gann, K. Byrne, Z.Han, Y.Y. Liu. FCRD-USED-2012-000039; M.C. Billone,T. Burtseva, M.A. Martin-Rengel, Effects of Lower Drying-Storage Temperatures on the DBTT of High-Burnup PWR cladding. FCRD-UFD-2015-00008. ANL-15121 ; Institut für Nukleare Entsorgung, KIT-INE

Disposal of Dual Purpose Casks as an alternative?

- DPC (e.g. CASTOR®) have been optimized for loading in reactors, transport and interim storage of HLW → but DPC are not "optimal" for final disposal (Bonano et al., 2018).
- Under repository conditions, long-term safety of final disposal of SNF and HLW-glass in DPC is rather doubtful
- How long does a DPC retain its integrity under final disposal conditions?

→ container integrity and corrosion resistance is questionable

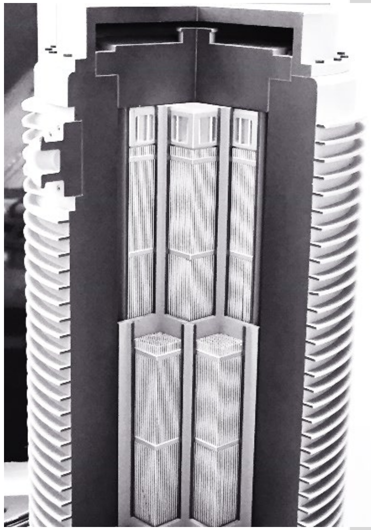
→ cooling fins would act as preferential surfaces for corrosion

- Existing DPC loaded with SNFare subcritical under interim storage conditions – subcriticality under repository conditions is uncertain

→ relatively high loading of DPC with fissile material

→ "*The studies so far do not conclusively demonstrate subcriticality…*" Filbert et al. (2011); assessment of subcriticality for disposal in granitic rock, argillaceous rock and rock salt not published;

→ for proof of subcriticality, scenarios with water ingress and chemical reactions for selective mobilization, transport and subsequent enrichment of fissile materials to be considered (Hardin et al., 2015)



$^{239}_{94}Pu$

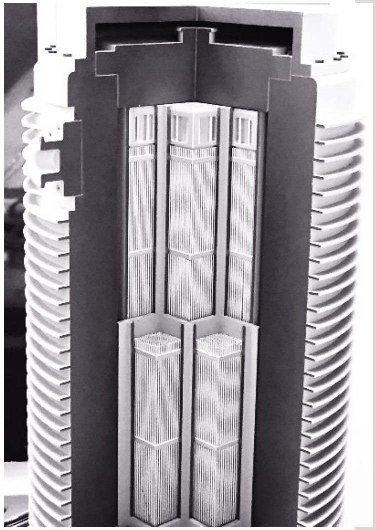
$t_{1/2}$ = 24110 a

σ_f=743b

σ_a=1019b

Sources: Filbert et al. (2011) WM2011 Conference.

Disposal of Dual Purpose Casks no general option



- Doubts about long-term corrosion integrity and sub-criticality
- corrosion of ~130 t Fe per DPC would produce enormous amount of H_2
- compatibility with geotechnical barriers not tested
- no retrievability concept

It's rather doubtful whether DPC can ensure long-term integrity without extensive conditioning, such as upgrading of corrosion protection or machining of cooling fins → disposal of small amounts of SNF in Dual Purpose Casks may be considered

"In Studien wurde auch eine direkte Endlagerung der Zwischenlager-behälter (vor allem CASTOR®-Behälter) diskutiert. Wegen der großen Unsicherheiten bezüglich einer Realisierung wird dieses Konzept im Rahmen dieses Papiers nicht weiter betrachtet […] Sollte eine direkte Endlagerung von TLB ohne weitere Behandlungsschritte wie das Verfüllen des TLB möglich sein, ist eine Konditionierungsanlage für die Umverpackung nicht erforderlich. Dies dürfte allerdings nur für eine Teilmenge der Fall sein" (ESK, 2023)

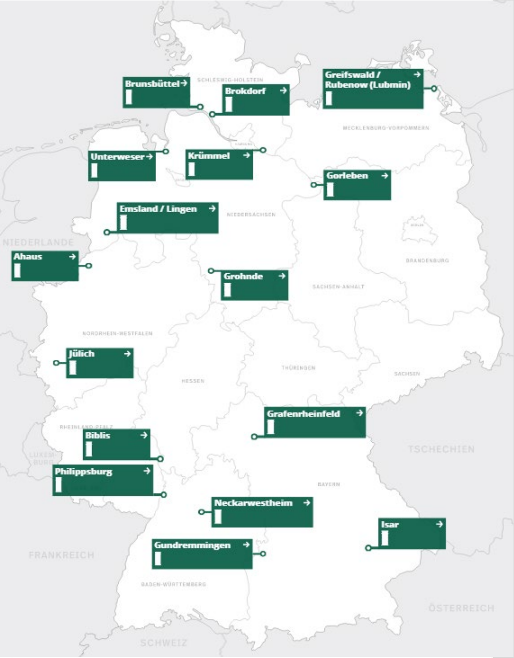
Source: ESK (2023) Verlängerte Zwischenlagerung bestrahlter Brennelemente und sonstiger hochradioaktiver Abfälle in Abhängigkeit von der Auswahl des Endlagerstandorts.

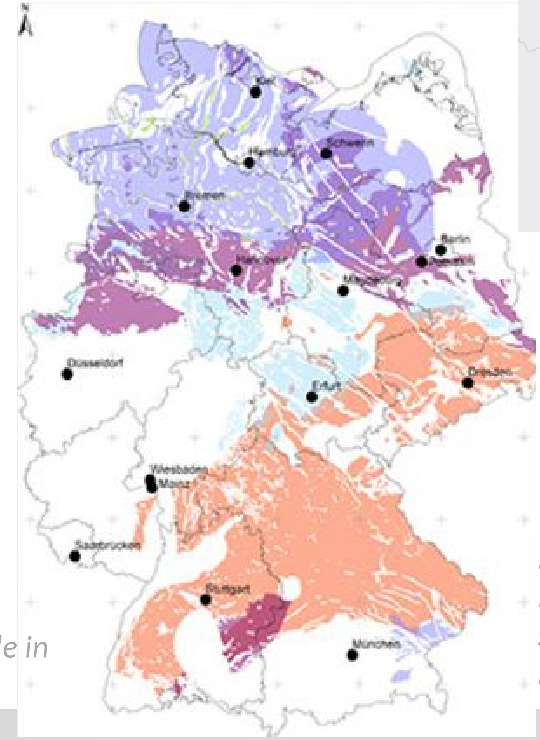
Interim storage in 16 facilities during one century?

- Considering a selection of the final disposal site in 2031, NaPro (2015) proposed continued interim storage in 16 facilities until transfer of high level waste to final disposal site

- Taking into account a significant delay in the site selection and thereby a prolonged interim storage, alternative options for the distribution [and design] of interim storage facilities may be considered (ESK, 2023)
  - one centralized storage facility
  - few "consolidated" / regional storage facilities

→ considerations about hot cells as part of an centralized facility or of consolidated facilities





Sources: NaPro (2015); ESK (2023) Verlängerte Zwischenlagerung bestrahlter Brennelemente und sonstiger hochradioaktiver Abfälle in Abhängigkeit von der Auswahl des Endlagerstandorts.

Image sources: BASE, https://www.base.bund.de/karte-zwischenlagerung/index.html#/ BGE, Teilgebietebericht

Final remarks and issues for discussion

- Degradation state of dual purpose casks and of inventories define the require-ments for the design of final disposal containers
- uncertainty about state of waste inventories is considerably increasing
- *„Die Auswirkungen von Alterung über lange Zwischenlagerzeiträume sind hinsichtlich der Zwischenlagerung selbst [… und] der sich anschließenden Entsorgungsschritte Konditionierung und Einlagerung von Bedeutung. [... Es] ist derzeit offen, ob sich hieraus Konsequenzen für die einzelnen Entsorgungsschritte und/oder die sicherheitstechnische Nachweisführung ergeben.“* (ESK, 2023)
- *“The current lack of knowledge on this crucial topic leads us to conservatively assume that the spent fuel physical state is fully degraded when assessing the RN source term”* Poinssot et al. (2014)
- Research needed to deal with relevant knowledge gaps
- Lab studies on Zr-hydride reorientation and pellet cladding chemical interaction, in particular with HBU-SNF, MOX -> last loading of DPC (CASTOR®) with SNF planned for 2027
- Real scale studies: Inspection of DPC and inventory of DPC for instance within EPRI program (USA), previously opening and inspection of DPC loaded with 45 MWd/kg SNF after 15 years of dry interim storage (ANL-03/17, NUREG/CR-6831)

Sources: Poinssot, C. et al. (2014) Journal of Nuclear Science and Technology 39. ESK (2023) Verlängerte Zwischenlagerung bestrahlter Brennelemente und sonstiger hochradioaktiver Abfälle in Abhängigkeit von der Auswahl des Endlagerstandorts.

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- Thomas Hassel (LUH, Hannover)
- Peter, Hocke, Dirk Scheer (KIT-ITAS, Karlsruhe)

Thank you for your attention!

TRANSENS
TRANSDISZIPLINÄRE FORSCHUNG ZUR ENTSORGUNG HOCHRADIOAKTIVER ABFÄLLE IN DEUTSCHLAND

Gefördert durch:

Bundesministerium für Wirtschaft und Energie

aufgrund eines Beschlusses des Deutschen Bundestages

Gefördert im Niedersächsischen Vorab der Volkswagenstiftung

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