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GEOLOGICAL DISPOSAL OF FUEL DEBRIS?
COMPARISON OF THE INTERACTION OF FUEL DEBRIS AND
OF SPENT NUCLEAR FUEL WITH WATER



B. Grambow Prof. Emérite IMT Atlantique/SUBATECH Nantes
Former director of SUBATECH and
group leader at the advanced science research center at JAEA

Spent fuel leaching studies in water since 1975 with the intention to study the option of direct geological disposal of spent nuclear fuel



Analyses of cooling water of Fukushima reactors to deduce from the evolution of actinide and fission product concentration on fuel debris behavior in water

Review paper:
Grambow, Nitta,
Shibata, Koma,
Utsunomiya,
Takami, Fueda,
Ohnuki, Jegou,
Lafolley, Journeau

2021

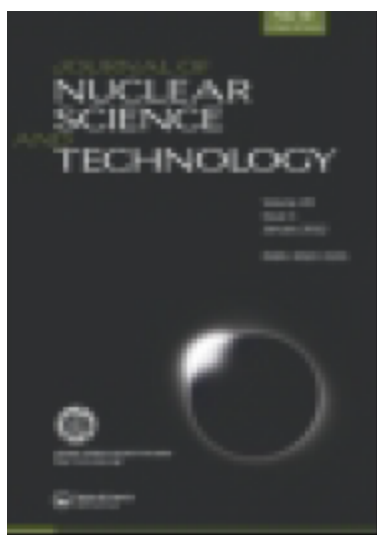


Fuel debris behavior under geological disposal conditions



Confinement options

This paper



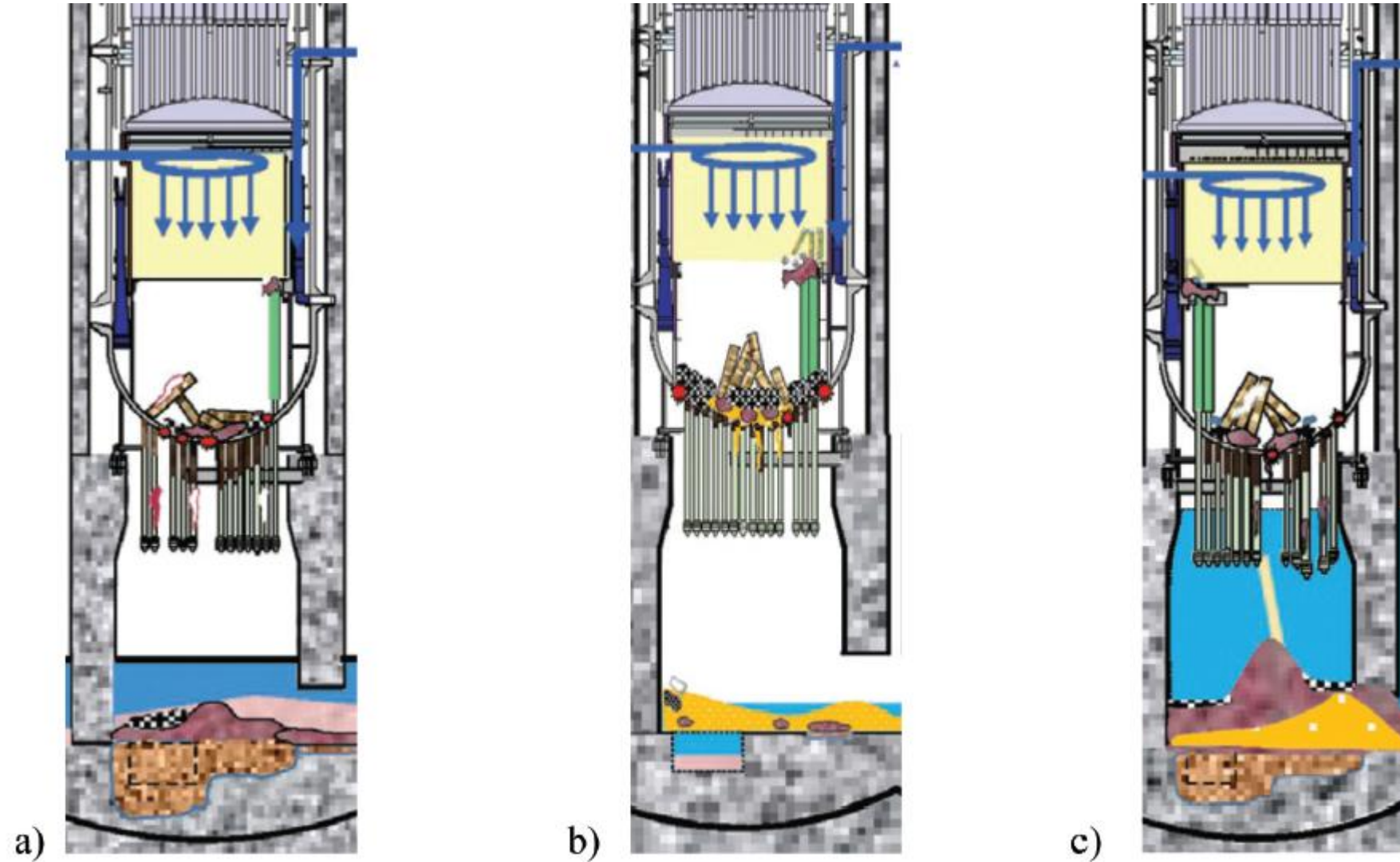
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Ten years after the NPP accident at Fukushima : review on fuel debris behavior in contact with water

Bernd Grambow, Ayako Nitta, Atsuhiko Shibata, Yoshikazu Koma, Satoshi Utsunomiya, Ryu Takami, Kazuki Fueda, Toshihiko Ohnuki, Christophe Jegou, Hugo Laffolley & Christophe Journeau

Fuel debris location in the 3 reactors



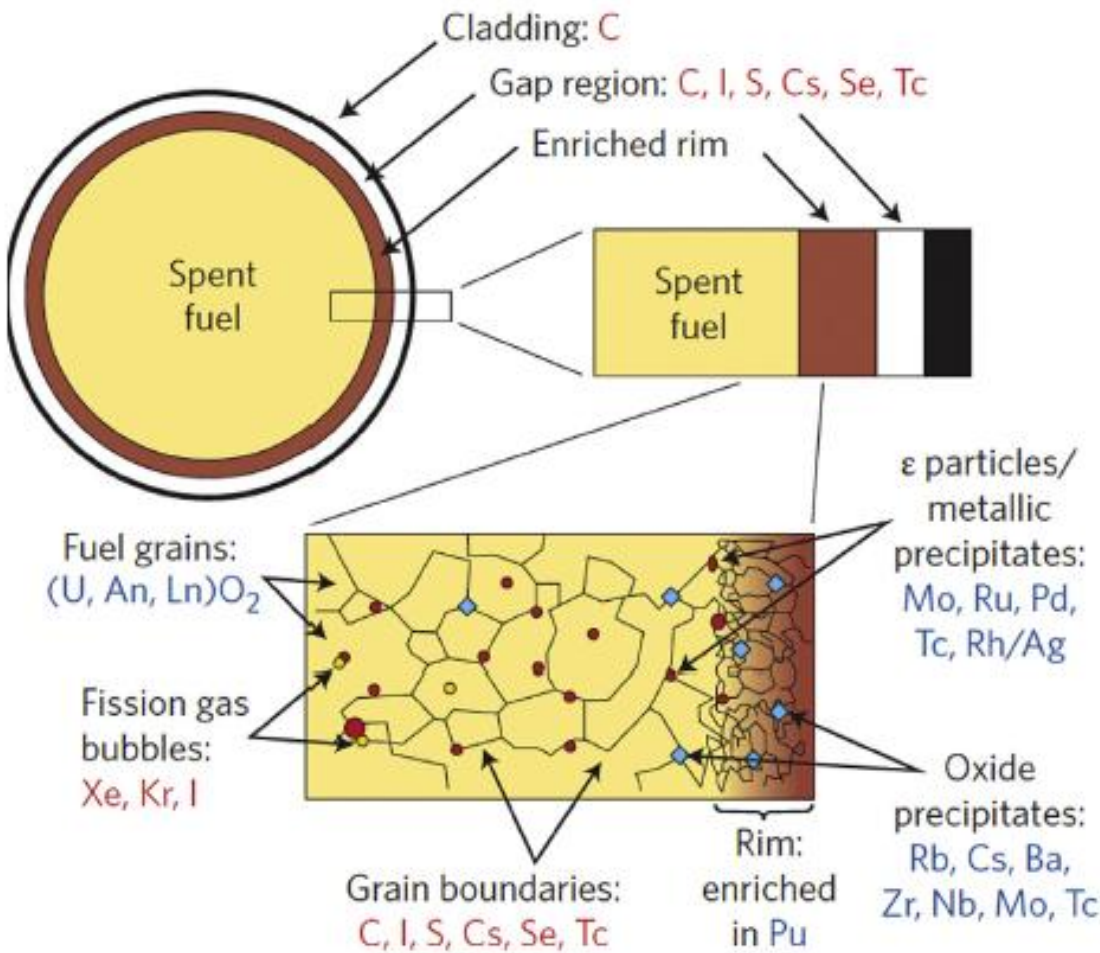
Interim endstate figures (as of December 2018) for a) unit 1, b) unit 2 and c) unit 3. Zooms on RPV and pedestal

Fuel debris composition

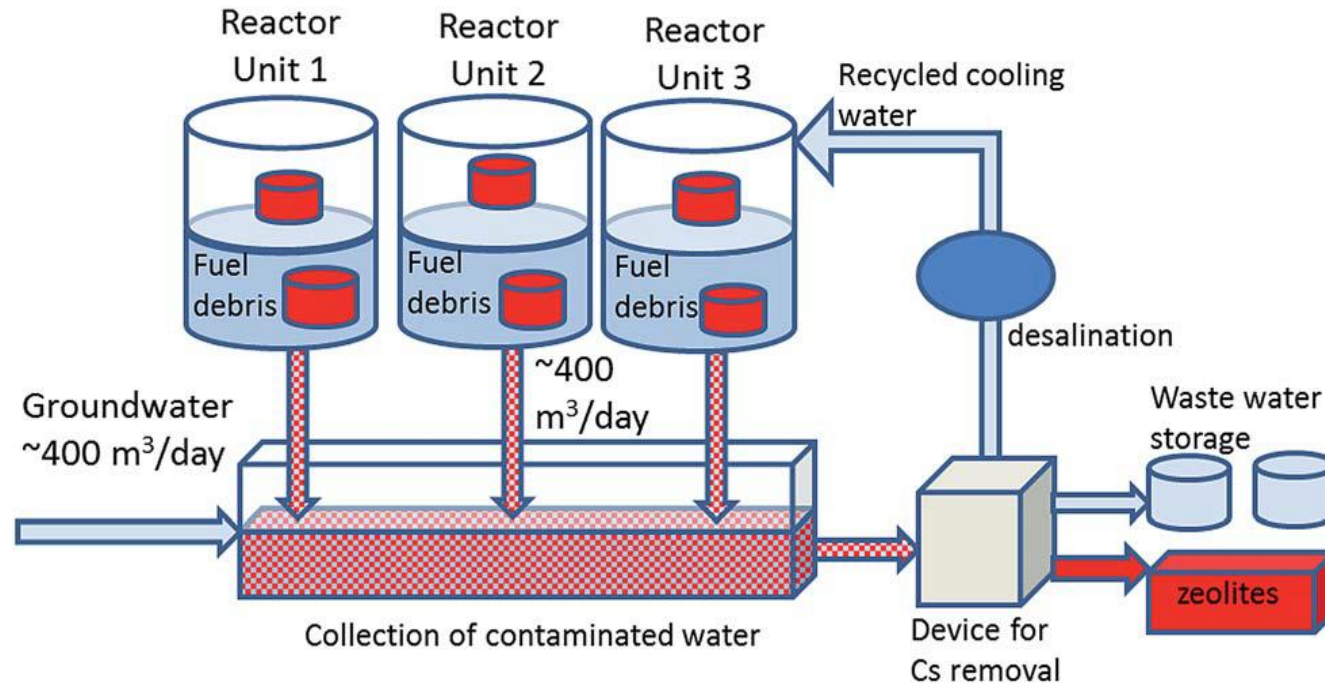
Table 1. Some typical compositions proposed for Fukushima Daiichi fuel debris (wt%).

Composition	UO ₂ + FPs	U	Zry	Oxi- dized Zry	Stain- less Steel	Oxi- dized steel	Boron
Steel-B ₄ C [10]					99.5		0.5
1F2 lower head (Average BSAF [139])	37		37	16	6	3	1
1F1 top metal layer (ASTEC [21])	2	14	9	1.4	73		0.5
1F1 middle oxide layer (ASTEC [21])	40	9	6	29	16	0.3	0.2
1F1 bottom metal layer (ASTEC [21])	28	12	8	21	30		0.3
1F2 liquid phase at lower head before failure [139]	9		5	8	78		
1 F2 particulate debris at lower head before failure [139]	52		19	17	10		1

Spent fuel



Permanent coupling of fuel by cleaned recycled water

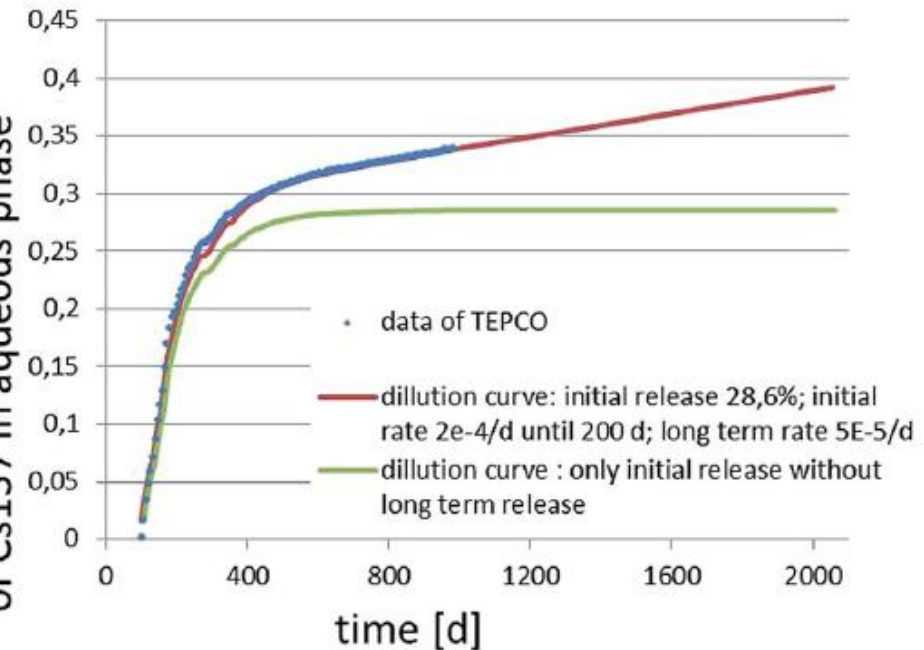


A giant leaching device

Analyses of collected water by Tepco



Cumulated fraction of inventory of Cs137 in aqueous phase



Atoms of $^{134/137}\text{Cs}$, ^{90}Sr or ^{129}I are released from the fuel debris in the reactor into the cooling water million to hundred million time faster than the atoms of actinides like Uranium or Plutonium

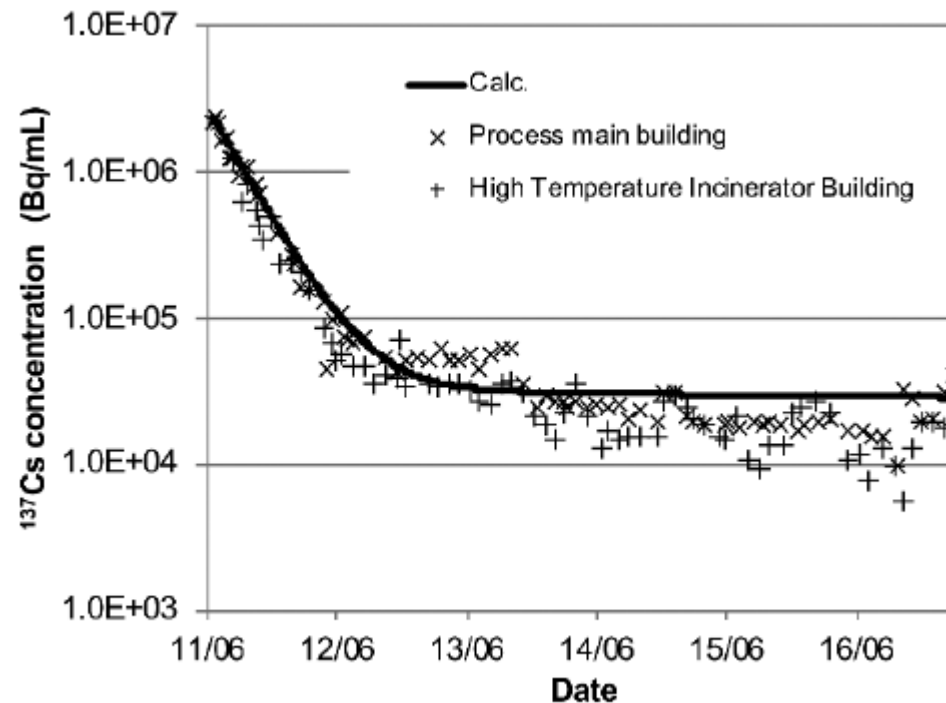
A similar but less extended effect has been observed in spent fuel disposal studies, when putting decladded spent nuclear fuel in experiments in water. This fraction of fast released radionuclide consists of similar elements, mainly of $^{134/137}\text{Cs}$, ^{129}I , etc. It is called IRF, “instant release fraction”

Permanent cooling of fuel debris consists in permanent leaching (removal) of the inventory of mobile elements from the debris.

The release rates of mobile elements decrease with time, as the most mobile fractions are washed out

Temporal evolution

- The release rate of Cs from the fuel debris in the reactors has been decreased by a factor of 100, indicating that the fuel debris becomes over time more stable (less release) in water, as **the instable fraction is washed out**

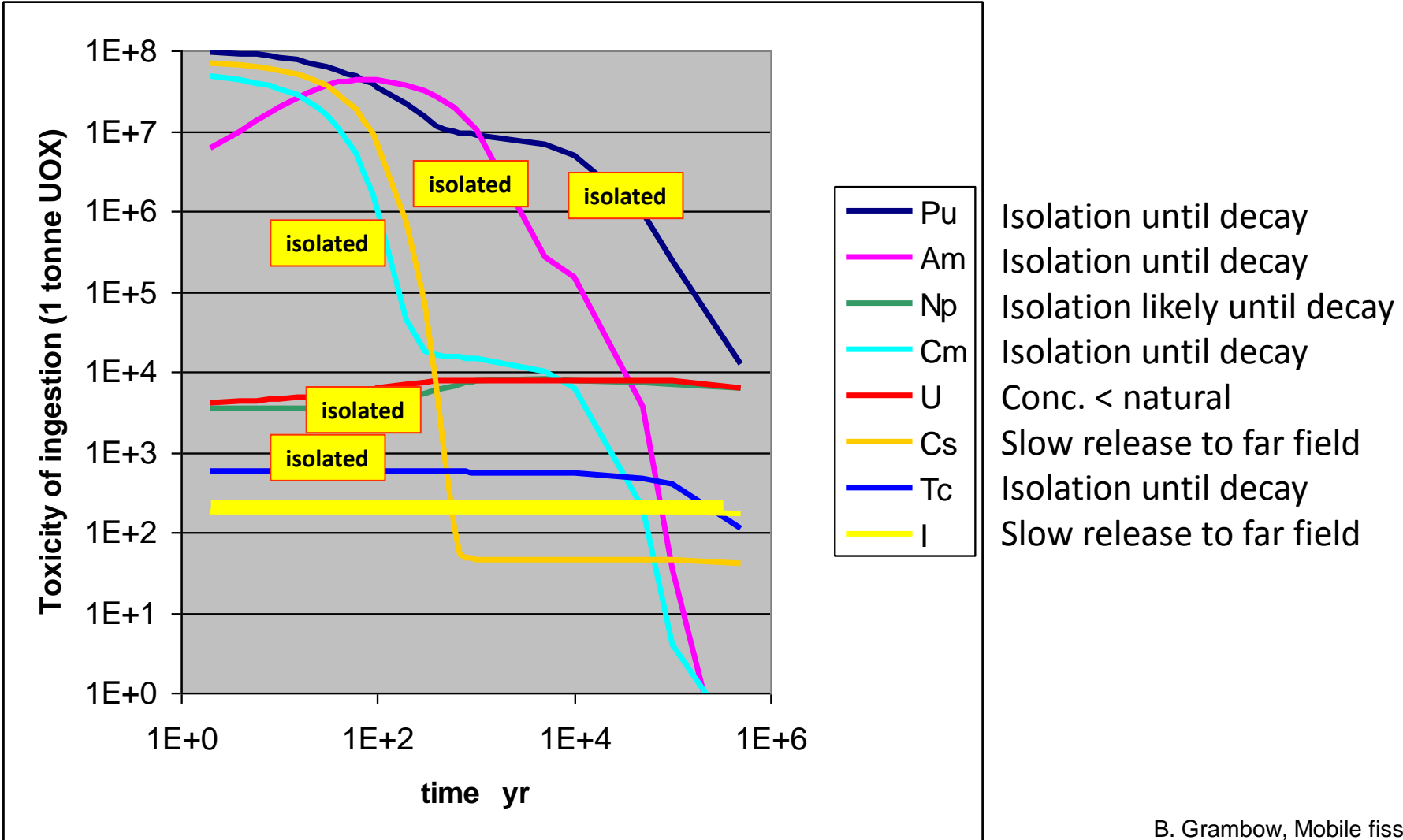


After leaching (removal by action of water) of the mobile fraction, the residual fraction shall be more stable than spent fuel

- The stability of spent fuel in water is assessed in Sweden, Switzerland and Finland to be larger than million years
- It stems from the stability (low solubility) of the UO_2 matrix, not from an additional barrier
- The Instant release fraction IRF of spent fuel
 - Leads to higher initial release, but has smaller long term effect in geological disposal
- Some fuel debris contain chemical elements, which are favorable to its stability in water over time,
 - such as Zr (low solubility and no oxidation),
 - rare earths, and
 - actinides,
 - Si, Ca or iron
- Fuel debris shall be at least as stable in water as spent fuel , if not more

Near field isolation of radiotoxicity in the geosphere during disposal

A key safety indicator: reducing conditions



B. Grambow, Mobile fission and activation products in nuclear waste disposal, Journal of Contaminant Hydrology, (2008) 102, 180-185

Actinides are very insoluble, limiting release from the debris, despite surface area is probably much larger than of spent fuel (in case of solubility control of release, surface area is not so important)

4  B. GRAMBOW ET AL.

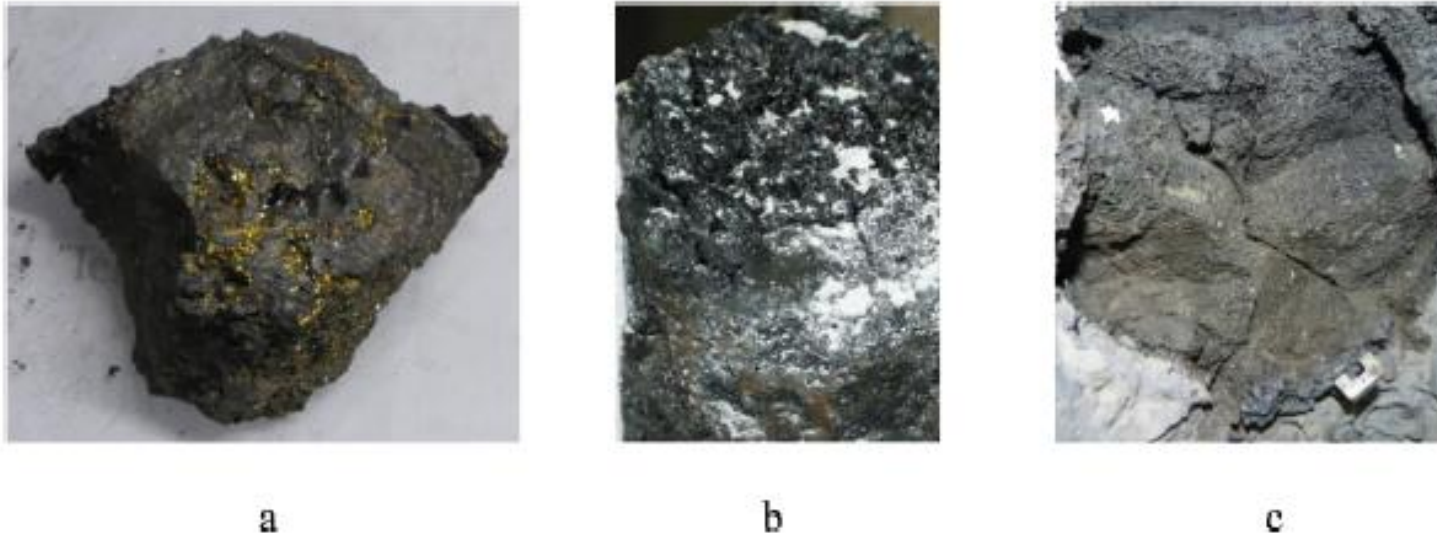
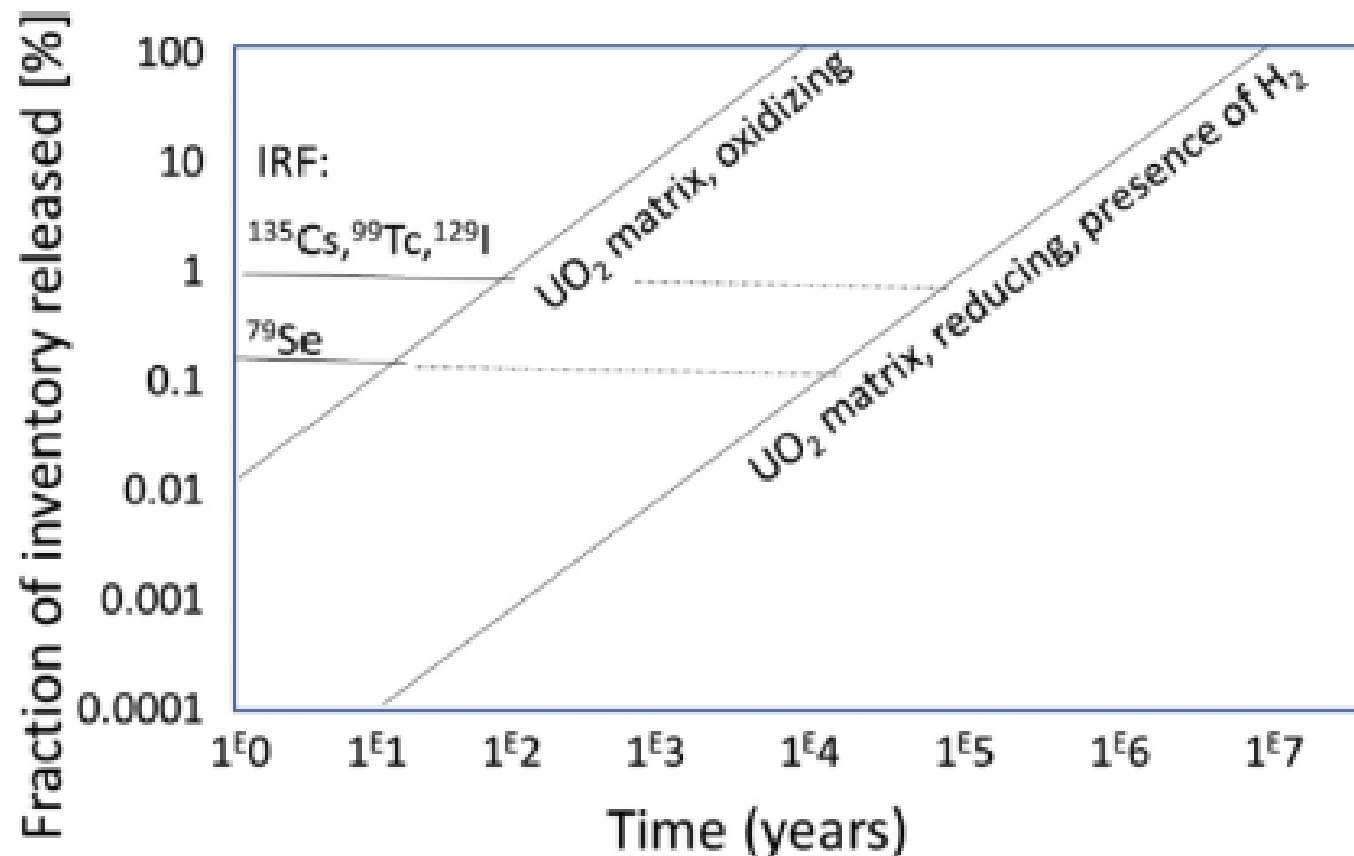


Figure 2. Some prototypic fuel debris fabricated during out-of-pile experiments a) VULCANO test (in nitrogen) with 1F2 average lower head composition from BSAF b) COLIMA CA-U3 composition with typical BWR MCCI composition c) VULCANO VF-U1 corium-concrete interaction test [26] (The magnet indicates the presence of a steel phase; undissolved gravels visible within darker oxidic

Reducing geochemical conditions further decrease solubility of U, Pu, Np, while solubilities of Am and Cm remain low

General simplified source term model for radionuclide release from spent fuel

Distinguishing labile nuclides (« Instant release fraction », IRF; few % ^{135}Cs , ^{79}Se , ^{129}I , ^{36}Cl ...) from UO_2 -matrix bound RN (the rest of Cs, I, Se, the total of actinides +Tc inventories...)



Important to assure reducing geochemical conditions (absence of oxygen) for disposal of fuel debris

IRF increases with burnup and linear power (fuel temperature)

Table 1 Estimates for IRF values in [%] for PWR fuel as a function of burnup in GWd/tU.^a

	<i>PWR</i>	<i>PWR</i>	<i>PWR</i>	<i>PWR</i>	<i>PWR</i>
Burnup	37	41	48	60	75
Fission gas	0.3	0.4	1	3.6	6.5
% FG in rim	0	0	2	4	8
¹⁴ C	10	10	10	10	10
³⁶ Cl	0.9	1.2	3	10.8	19.5
⁷⁹ Se	0.2	0.2	0.2	0.2	0.2
⁹⁹ Tc	1	1	1	1	1
¹⁰⁷ Pd	1	1	1	1	1
¹²⁶ Sn	0.1	0.1	0.1	0.1	0.1
¹²⁹ I	1	1	2.5	7.6	10.5
^{135,137} Cs	1	1	2.5	6.1	7.8

In fuel debris an analogue « IRF » corresponds to the fast washed out fraction in the reactors. It is for ^{134/137}Cs > 30%, for ⁹⁰Sr > 20% due to the much higher temperature than in normal reactor operation.

What about protecting future humans from the fuel debris in the long term?

The debris remain dangerous for thousands of generations

Final disposal!

The only way to avoid irradiation risks for future humans: to isolate the fuel debris forever within an inaccessible deep (>300 m) geological formation (clay rock, granite...)

Some ideas requiring further thoughts...

Encapsulation in highly stable containers?

- Cu containers in case of granite due to presence in granite of water accessible fractures
- SS or cast iron containers in case of clay

maybe filling of remaining void spaces in the debris containing containers by suitable materials

Further conditioning of fuel debris for disposal

BOROSILICATE GLASSES AS WASTE FORMS FOR ACTINIDES AND FISSION PRODUCTS

Classical waste glass melting



Cold crucible

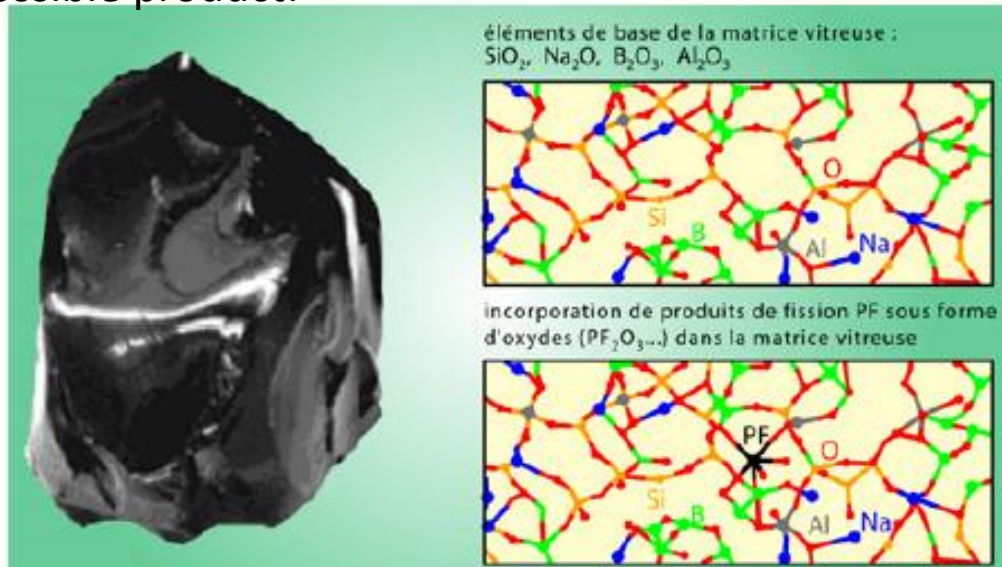


- Unique host phase for Pu, U, minor actinides, fission and activation products
- Vitrification by dissolving the debris to form a glass phase: Very high temperature melting required (maybe by cold crucible technique?).
- However: Vitrification provides no safety gain for geological disposal: actinide release is controlled by low actinide solubility in groundwater in a repository and not by glass dissolution rates.
- Solubility of actinides in a repository environment do not decrease by vitrification.
- The kinetics of mass transfer of remaining soluble radionuclides (I, Cs...) from vitrified debris to the environment may decrease

Different ways for vitrification

Classical vitrification of HAW like in reprocessing by (1) dissolving all the debris in highly concentrated nitric acid, (2) calcination of the fluid and adding the calcinate together with glass forming constituents (Si, Al, Na...) (3) to the melter at about 1200°C by induction furnace or by a joule heated melter or at even higher temperature in a cold crucible melter). Direct melting of fuel debris in glass melt may require too high temperatures?

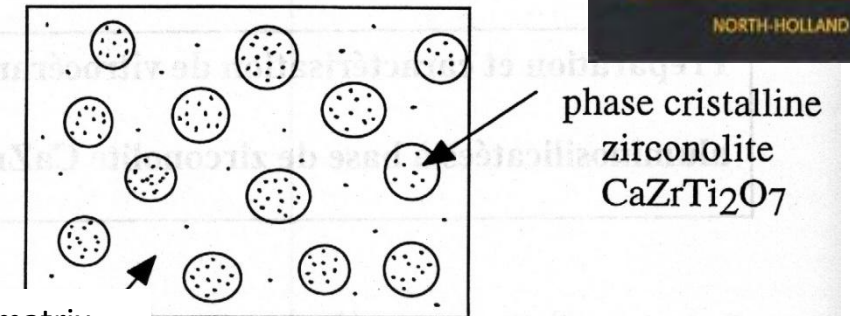
The possible product:



Photography of a piece of French R7T7 glass and a sketch for the confinement of radioactive chemical elements in the molecular glass structure

Maybe fabricate a waste glass by encapsulating small particulate fuel debris a glass matrix forming some kind of a glass ceramics. This would avoid the dissolution of the debris in nitric acid. These materials may also be produced by sintering. Work was done in the 80th, see Lutze and Ewing: Waste forms for the future (1988)

A glass ceramic matrix

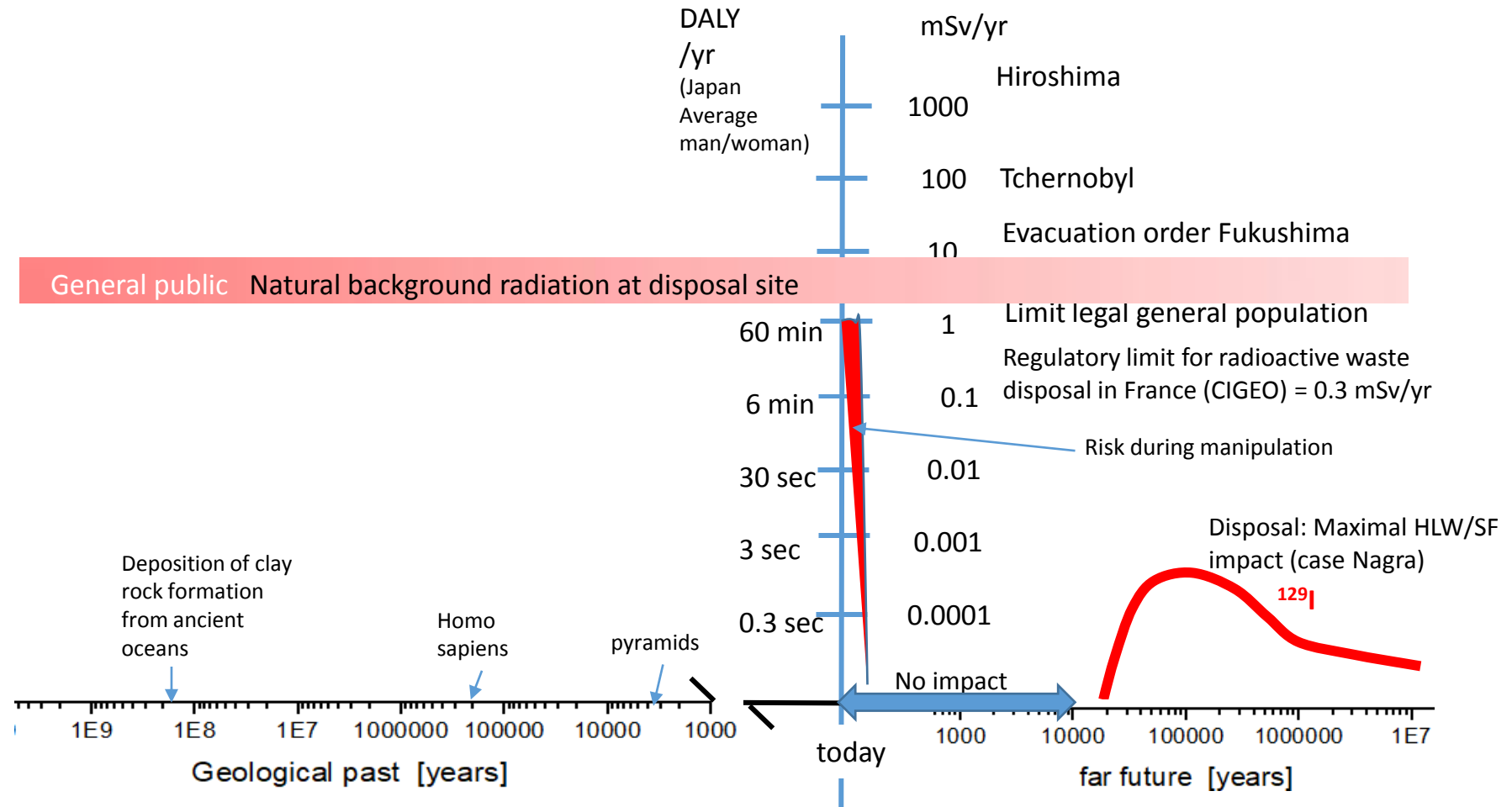


Residual glassy matrix, composition may be adapted to needs of resistance, strength and durability



Only geological disposal provides the long term safety needed for multiples generations

Radiological risks for the very far future: Example geological disposal in clay formations of spent fuel.



Risk from geologically disposed fuel debris may even be lower than that of spent fuel as the dose dominating ¹²⁹I (and probable ³⁶Cl) is already largely released in the accident and through cooling

Vitrified debris are unlikely to provide further safety gains compared to non vitrified containerized debris

Conclusions

Comparing the fuel debris with spent nuclear fuel, the 'washing' of the fuel debris over many decades by cooling water may remove a much larger fraction of the more mobile radionuclides (^{129}I , ^{36}Cl , ^{135}Cs ...) compared to that what is observed with spent nuclear fuel.

As the environmental impact of disposed spent nuclear fuel is largely linked to this more mobile nuclides, the direct disposal of fuel debris in suitable containers might be less restrictive or complicated than the direct disposal of spent nuclear fuel pursued in many countries.

Encapsulation of fuel debris in a glass matrix (vitrification) in a container may provide further handling and release stability and may retard the access of groundwater to the debris in case of disposal, but it is unlikely to reduce the long term risk further once disposed compared to none-vitrified debris

Confinement studies using simulated fuel debris may need to be pursued

A last word on past efforts to create a worldwide
nuclear legacy initiative

AN ATTEMPT TO START A GLOBAL INITIATIVE ...

Mobilisation of scientific community to work on nuclear legacy issues like in Japan is the base to build an IPCC like initiative, stopped after COVID crisis??

- To help speeding up ...
- To have a long term vision...
- To have a systems approach...
- To be interdisciplinary and transdisciplinary
 - Geosciences, radiochemistry, nuclear physics, medicine, toxicology, demography, history, sociology, landscape management...
 - joint problem framing by social and natural sciences
- To inform policies and regulations...
 - How to attain Agenda 2030 and..
 - the Sustainable Development Goals...
- To have an internationally coordinated approach...
- A number of meetings conducted in Tokai, Nantes, Tokyo with help of JAEA and the french



French Academy of Science, japaneze science council, ICSU (today ICS)

Nuclear legacy initiative

Limit the nuclear legacy burden for our children

