Impact of different irradiation scenarios on the long-term behavior of ISG glass

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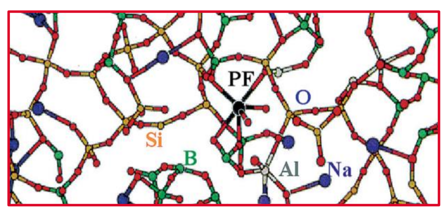
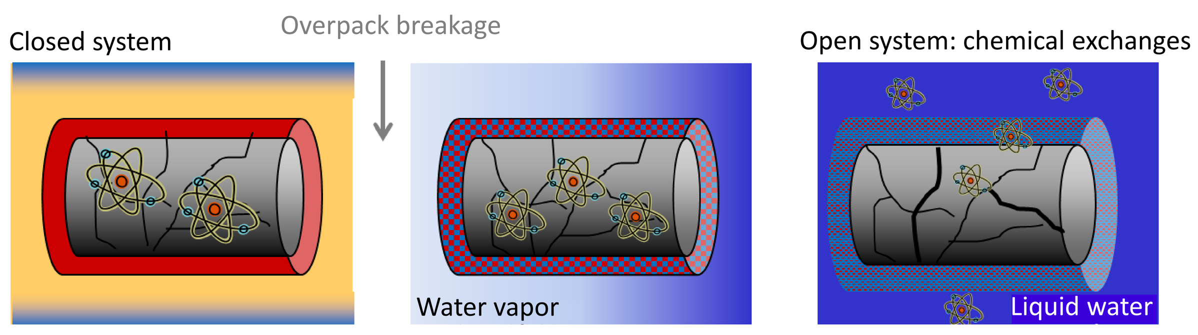
Context

High-level nuclear wastes => borosilicate glass



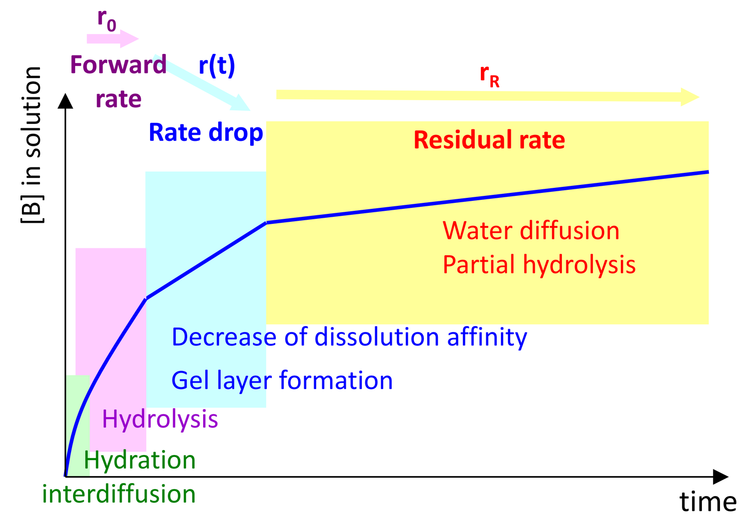
Nuclear fuels are reprocessed and the high level wastes are vitrified in a borosilicate glass. They are intended for a long-term disposal in a deep geological repository.

Long-term evolution in deep geological disposal



Firstly, the glass will be in a closed system and will evolve through **self-irradiation.**[1-3]
Then when the overpack breaks, water vapor will begin to corrode the glass.
Finally, the clay will be resaturated by groundwater, which will **alter** the glass.
The impact of irradiation on glass alteration needs to be studied.

Alteration kinetics under saturated water

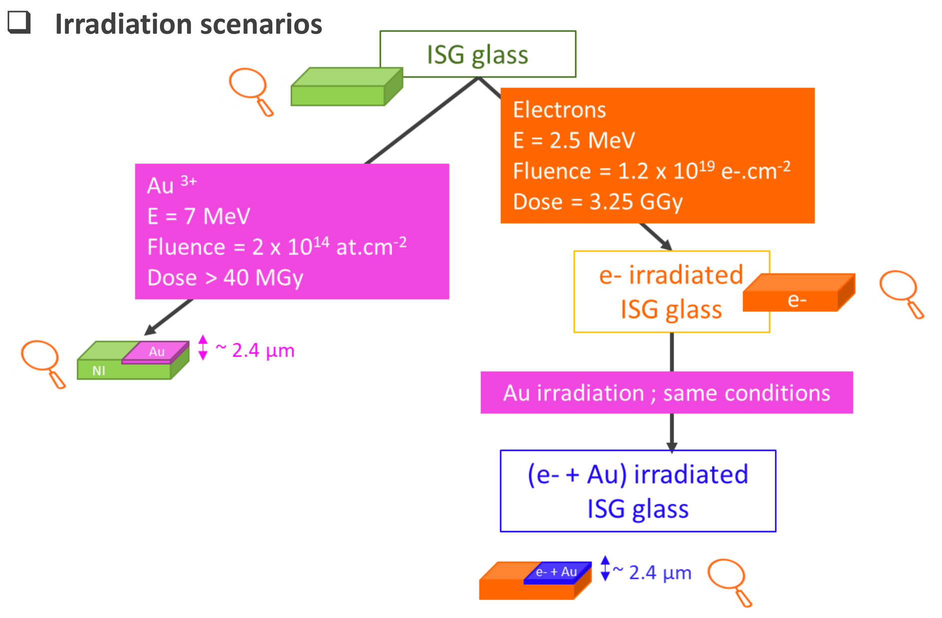


Materials and methods

Material

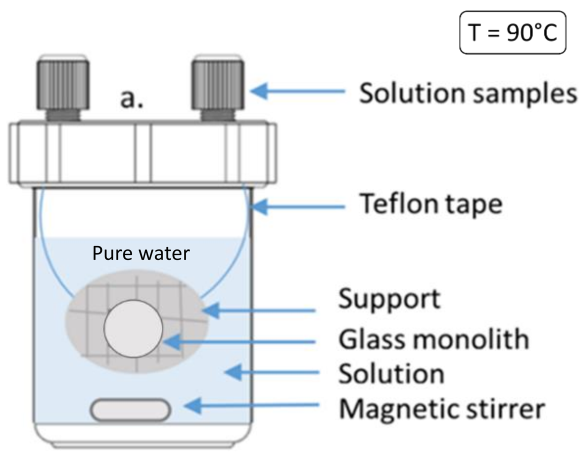
- **ISG glass (International Simple Glass): 6 oxydes borosilicate glass**
  SiO_2, B_2O_3, Na_2O, Al_2O_3, CaO, ZrO_2

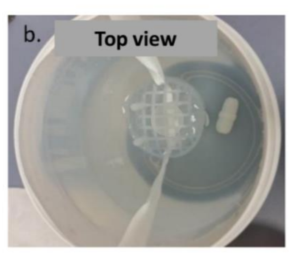
- **Irradiation scenarios**



Forward dissolution rate

- **Leaching device**

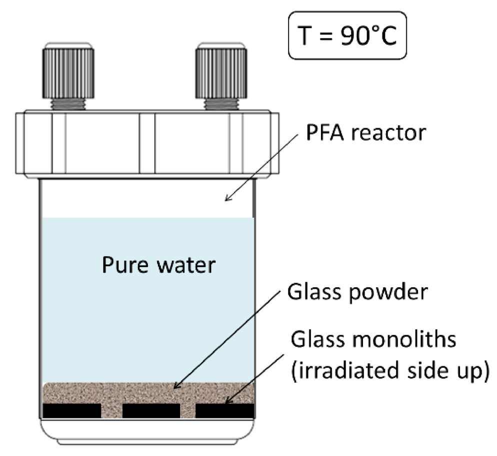


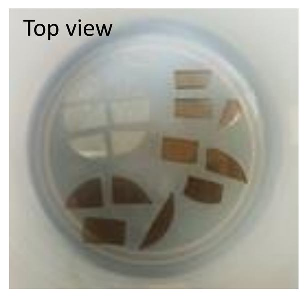


- **Solution analysis** (UV and ICP-AES) on regular samplings

Residual dissolution rate

- **Leaching device**



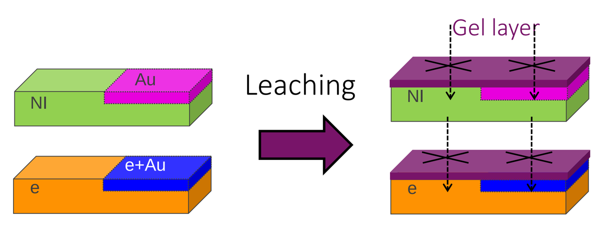


Characterization

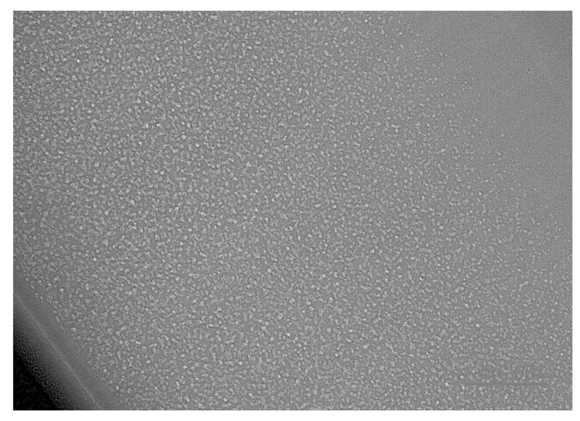
Glass coupons were regularly sampled and characterized by X-Ray Reflectometry, ToF-SIMS and TEM on FIB thin foils.

Glass alteration is monitored through the thickness of the altered layer (gel).

Cryo-TEM coupled with image processing (ImageJ) is used to characterize the porosity of the gel layer.

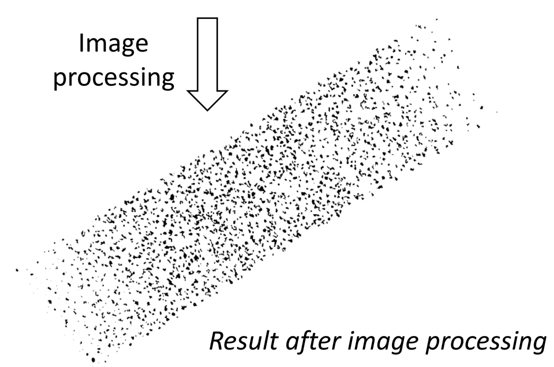


Cryo-TEM image processing



Cryo-TEM image in defocusing mode

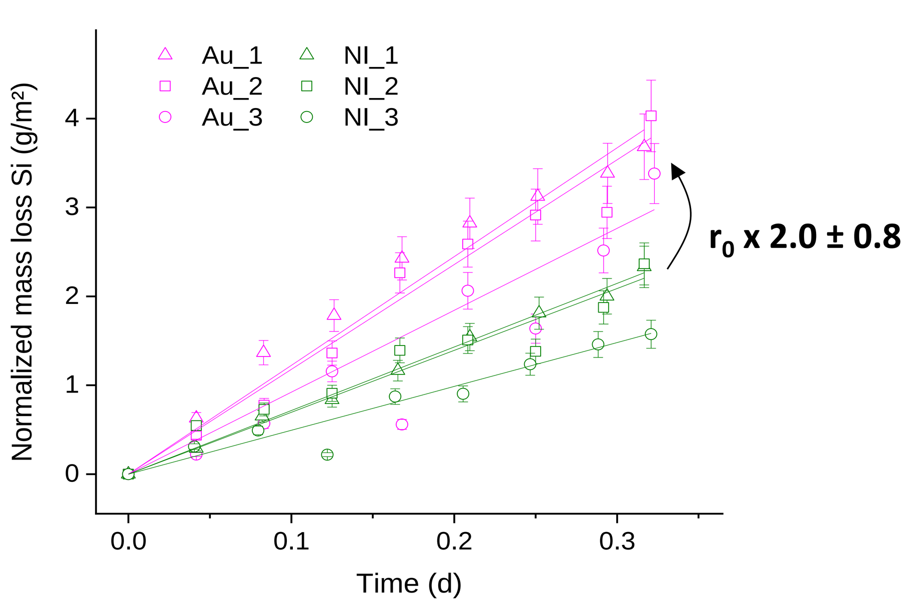
Image processing



Result after image processing

Alteration kinetics

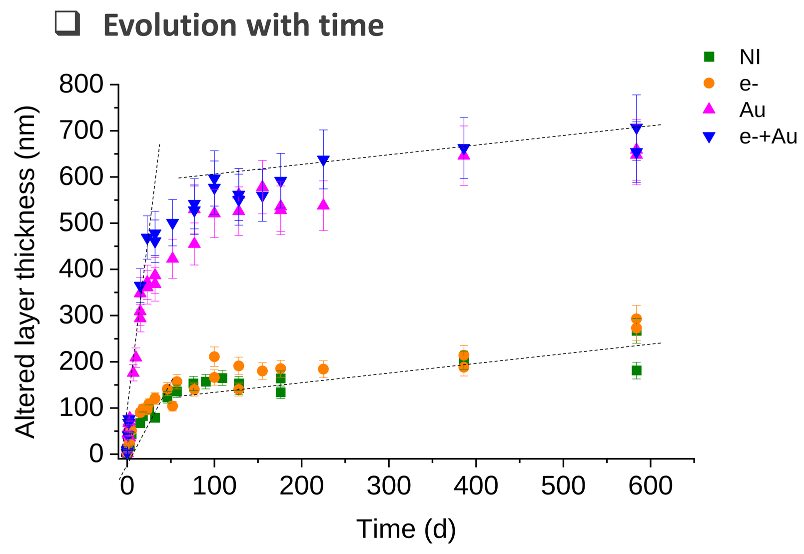
Forward alteration rate [4]



Au irradiation increases the forward alteration rate of ISG glass by a factor 2.

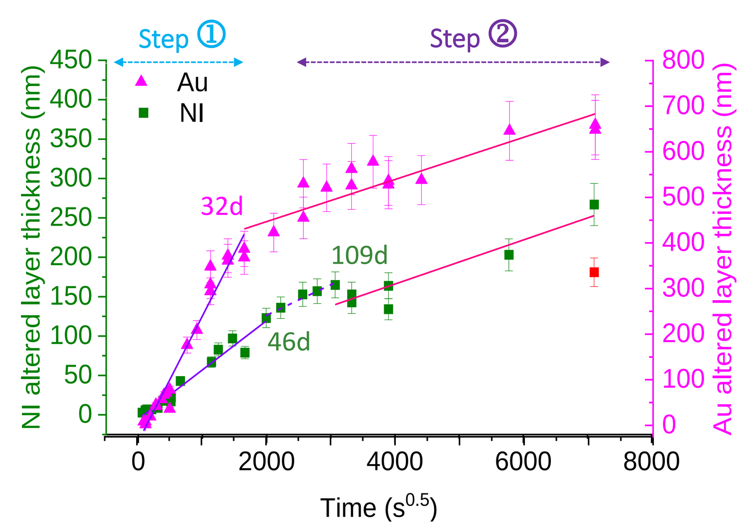
Residual alteration rate [5]

- **Evolution with time**



2 groups: NI ≈ e- << Au ≈ e-+Au
2 steps: First weeks: r (NI/e-) << R (Au/e-+Au)
Longer time: r_R (NI/e-) ≈ R_r (Au/e-+Au)

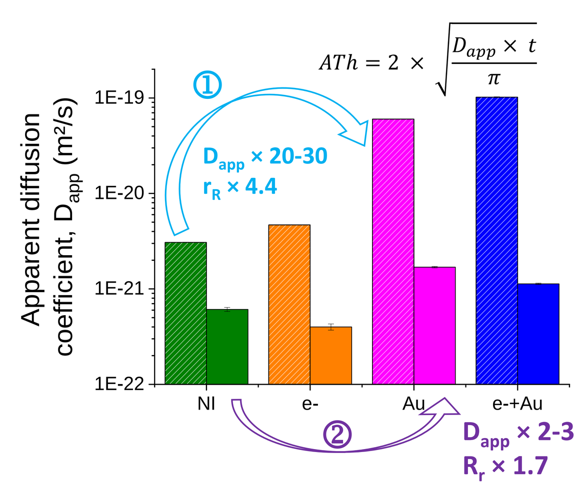
- **Evolution with square root of time**



2 Linear parts => 2 diffusive processes?
Earlier and more abrupt transition for Au (and e- + Au)
More progressive transition for NI (and e-)

**Results for e- and e- + Au not shown because similar to that of NI and Au*

- **Apparent diffusion coefficient**



① First diffusive step
Great impact of Au irradiation on D_{app}
NI ≈ e- << Au ≤ e- +Au

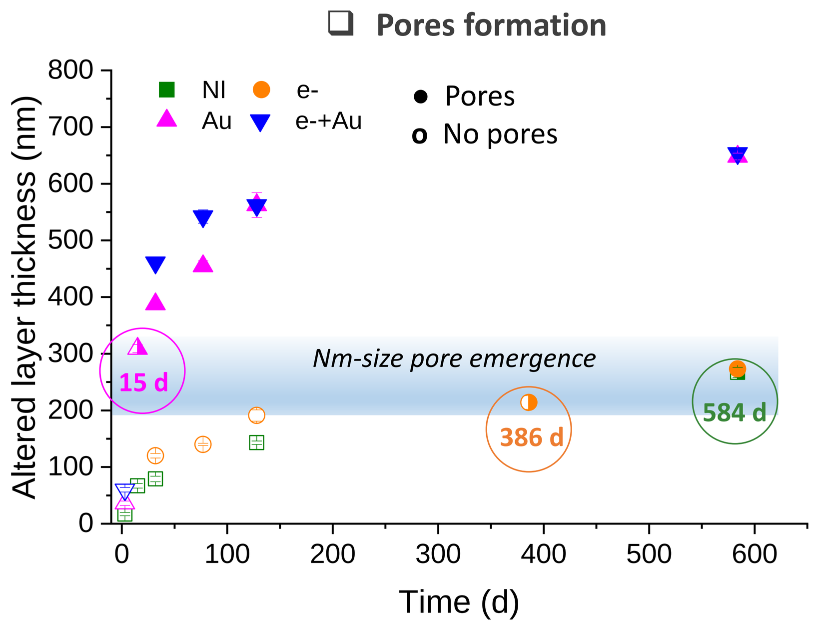
② Second diffusive step
Lower impact of Au irradiation on D_{app}
NI ≈ e- < Au ≈ e- +Au

ISG glass leaching behavior is impacted by the initial structure of the glass (mostly the first weeks)

Solid characterization

Gel porosity [5]

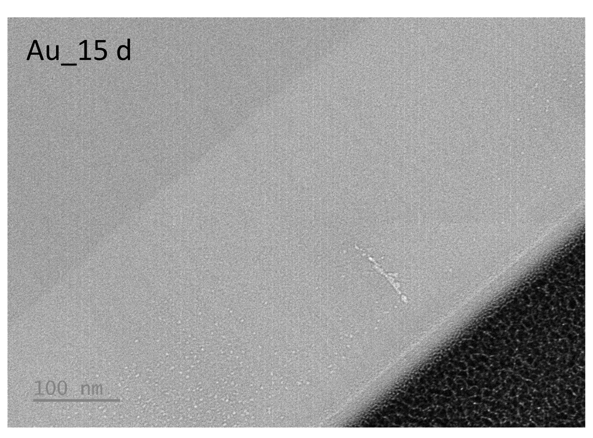
- **Pores formation**



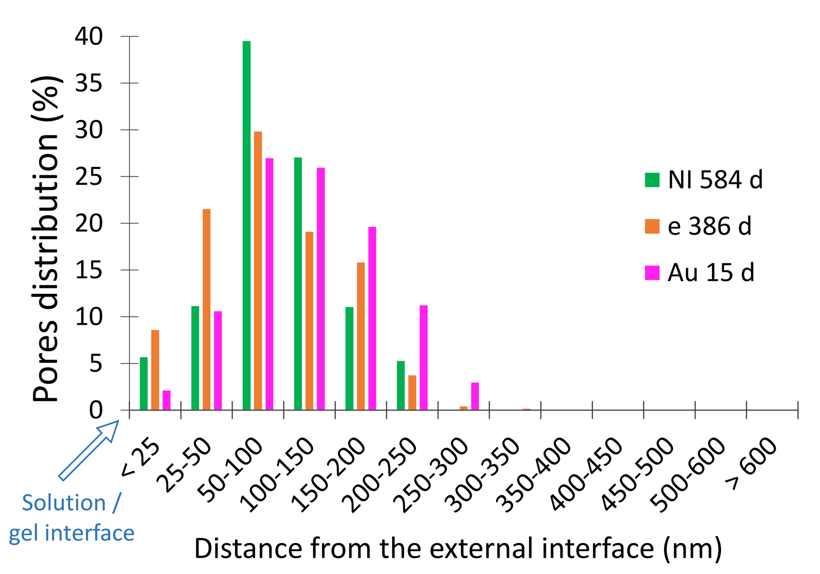
The first pores appear at different times:
- Between 15 – 32 days for Au et e-+Au glasses
- Between 386 – 584 days for e- et NI glasses

But at a similar thickness of the altered layer

- **Spare pores distribution**

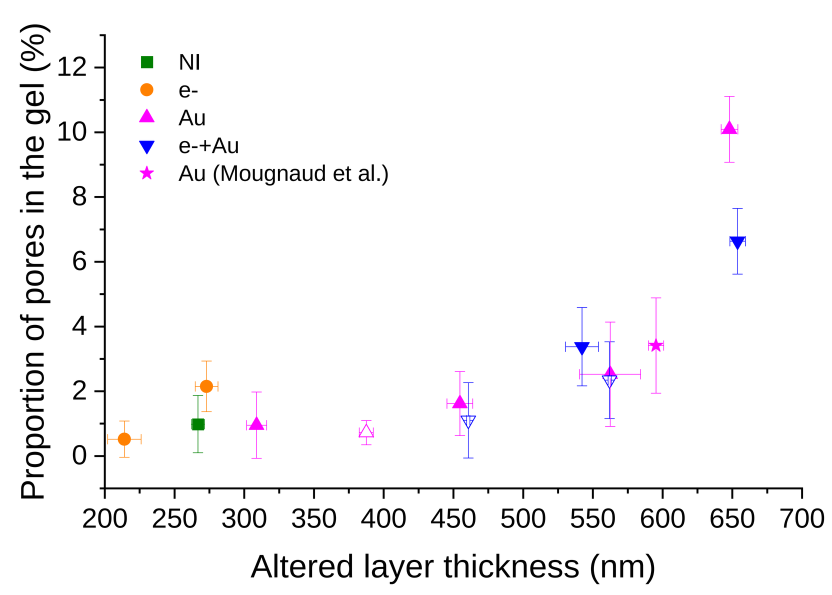


When they appear, pores are not homogeneously distributed. Their diameter is about 2 nm whatever the scenario and time.



Similar size and distribution of pores when they appear.
Whatever the scenario, porosity appears for the same thickness of the altered layer.

- **Pores proportion**



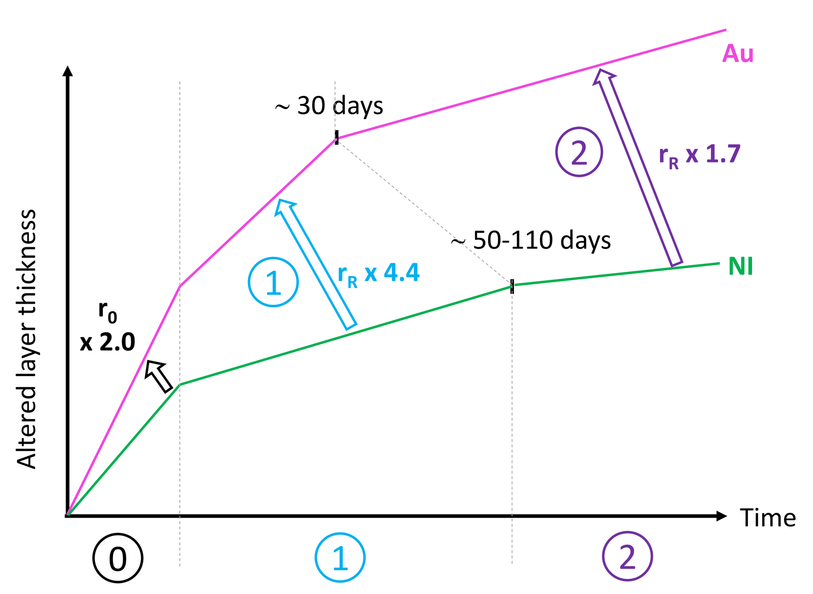
Increase in the pores proportion with the thickness of the alteration layer.

Conclusions

Au-irradiation has a stronger impact on glass alteration than e-irradiation, with no additionnal effect from sequential e+Au irradiation.

The forward alteration rate is increased by 2, while the residual alteration rate is increased by 4.4 in the first diffusive step and by 1.7 in the second one.

The appearance of pores in the gel seems to depend on its thickness, not its age. Gel characteristics (microstructure, composition, pore size and distribution) and alteration mechanisms are similar whatever the irradiation scenario.



References
[1] Gillet C. et al. (2022) Journal of Nuclear Materials, 572 (154079)
[2] Peuget et al. (2018) npj Materials Degradation 2(1)
[3] Mir. et al. (2017) Journal of Nuclear Materials, 489 pp. 91-98
[4] Gillet C. et al. *Impact of gold ions irradiation on the initial alteration rate of the International Simple Glass*, Journal of Nuclear Materials, submitted
[5] Gillet C. (2022) Thesis, Montpellier University

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