DE LA RECHERCHE À L'INDUSTRIE

RADIATION DAMAGE IN GLASSES

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DE LA RECHERCHE À L'INDUSTRIE

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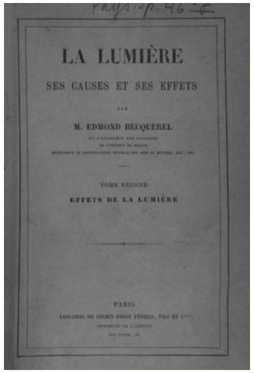
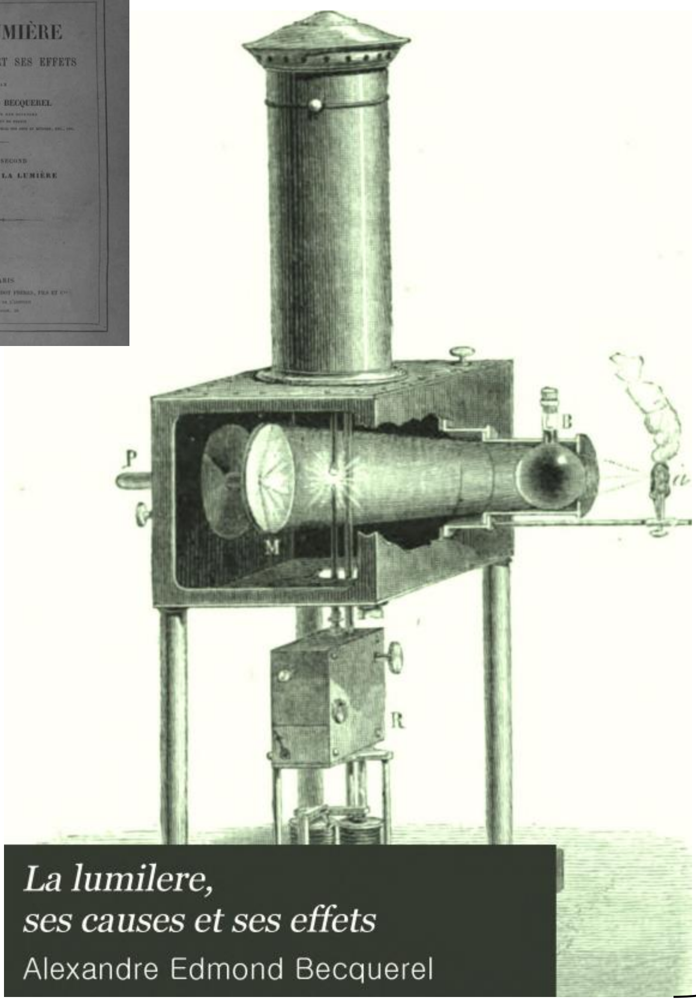
S.

Glass and radiation: an old story

Edmond Becquerel, *La lumière: ses causes et ses effets*, Vol. 2 (Paris, France: F. Didot, **1868**)





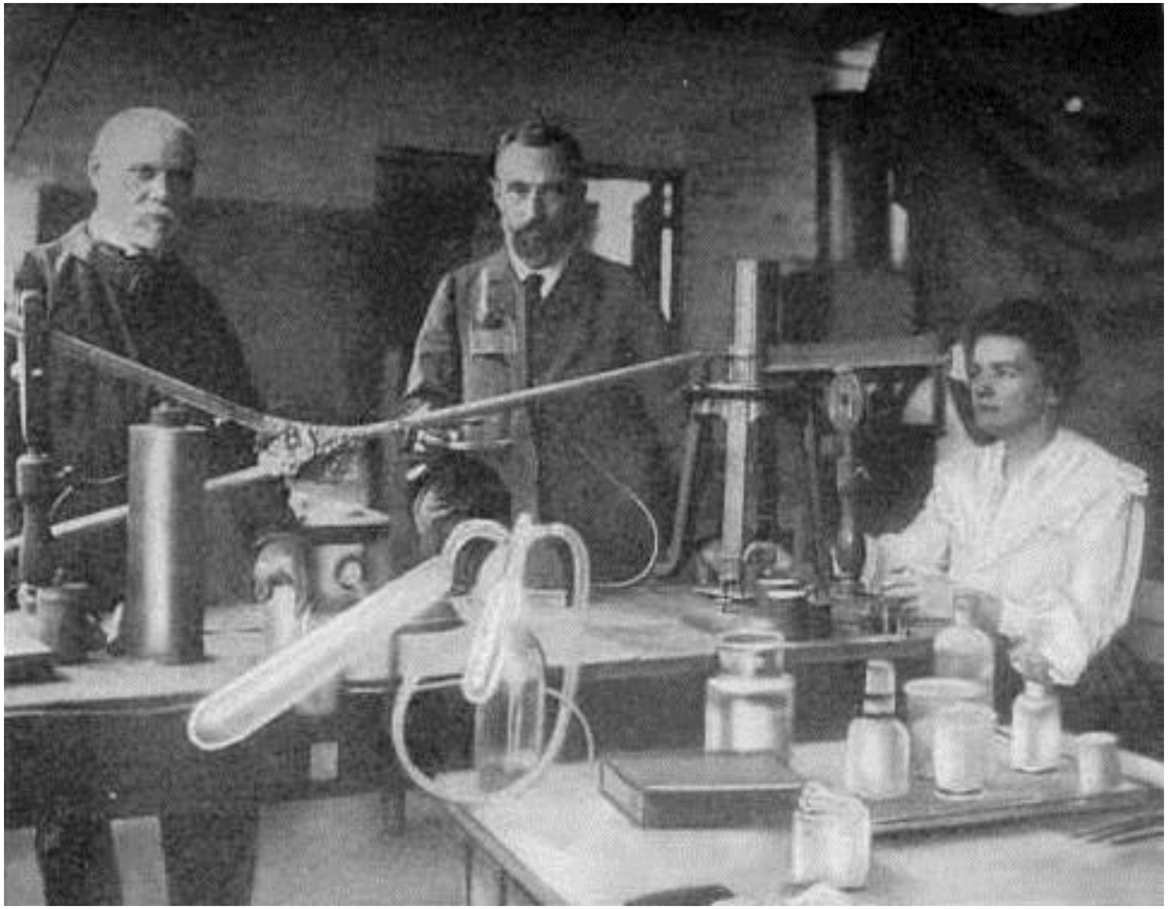
La lumilere, ses causes et ses effets
Alexandre Edmond Becquerel

- M. Faraday, 1824, modification of the glass color when exposed to sun light
- Pelouze, 1867, glass coloration observed when the glass contains Mn or Fe
- E. Becquerel, 1868, process involving a modification of the oxydation state of Mn, Fe
- P. Curie, 1899, Radium colors or blackens glasses

Le verre peut éprouver des changements de teinte à la lumière; ainsi Faraday (2) a observé que le verre teint en pourpre par le manganèse prend une teinte beaucoup plus prononcée quand il a été exposé aux rayons solaires. Pelouze (3), qui s'est beaucoup occupé des causes de coloration des verres, a reconnu que des verres ordinaires pouvaient se colorer à la lumière, mais quand ils renfermaient des sels de manganèse ou de fer, et une fois la coloration obtenue, l'action de la chaleur pouvait la faire disparaître;

(2) *Ann. de chimie et de physique*, 2e série, t. 25, p. 99 (1824)
(3) *Ann. de chim. et de phys.*, 4e série, t. 10, p. 194 (1867).

La coloration du verre à la lumière, d'après cela, semble être due à ce que le peroxyde de fer cède une partie de son oxygène au protoxyde de manganèse, qui passerait à un état d'oxydation supérieur; le recuit du verre, c'est-à-dire l'action de la température rouge sombre, produirait une réaction inverse, qui expliquerait la décoloration.



Pierre and Marie Curie in their laboratory, where radium was discovered.

Glass and radiation: an old story

Vol. 10. Part 1   March, 1925   Number 51

BULLETIN

OF THE

NATIONAL RESEARCH COUNCIL

RADIOACTIVITY

Report of Committee on X-Rays and Radioactivity
National Research Council

BY
A. F. KOVARIK AND L. W. MCKEEHAN

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THE NATIONAL ACADEMY OF SCIENCES
WASHINGTON, D. C.
1925

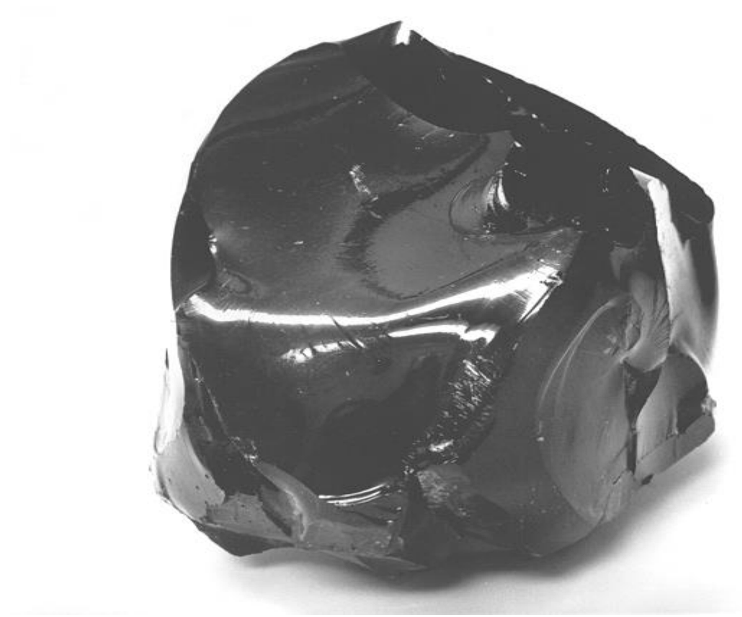
- Doelter, 1910, a review of the coloration of glasses by the action of rays from radium

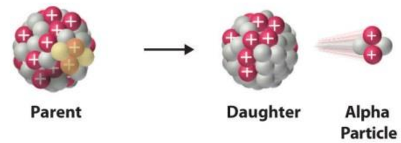
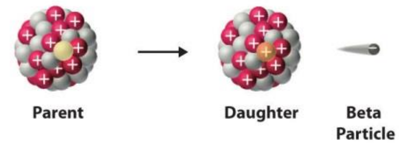
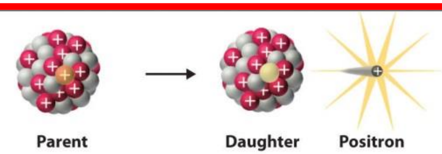
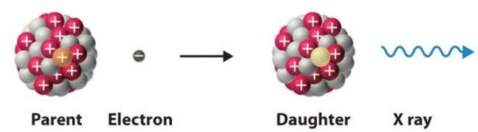
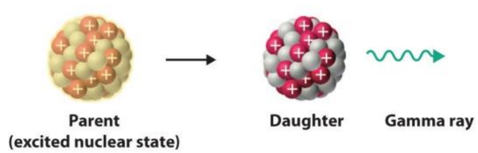
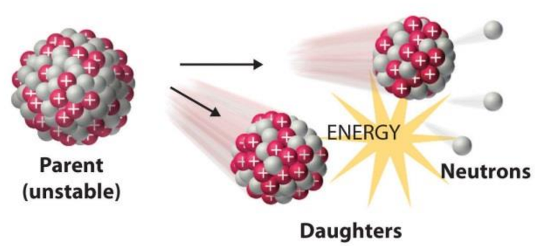
The early work on the coloration of glasses and minerals by the action of the rays from radium is reviewed in Doelter's memoir of 1910. Among

by α-rays, β-rays and γ-rays, (Crookes, Lind-Bardwell). In some cases the color produced is a surface effect and is probably the result of α-rays, which in forming haloes in mica and in coloring glass produce color only as far as they penetrate. The intensity of coloration reaches a limit with

Kara-Michailova). The rate of disappearance is greater at higher temperatures and Clarke calculated on the basis of his experiments with glass colored violet that instantaneous decoloration of the specimens would have taken place at from 500 to 600° C, which is also the annealing range for the glass he used, so that both decoloration and annealing may be connected with the same change of molecular aggregation. Bělař made

Nuclear Glass or GCM: What type of radiation?



Decay Type	Radiation Emitted	Generic Equation	Model
Alpha decay	${}^{4}_{2}\alpha$	${}^{A}_{Z}X \longrightarrow {}^{A-4}_{Z-2}X' + {}^{4}_{2}\alpha$	
Beta decay	${}^{0}_{-1}\beta$	${}^{A}_{Z}X \longrightarrow {}_{Z+1}^{A}X' + {}^{0}_{-1}\beta$	
Positron emission	${}^{0}_{+1}\beta$	${}^{A}_{Z}X \longrightarrow {}_{Z-1}^{A}X' + {}^{0}_{+1}\beta$	
Electron capture	X rays	${}^{A}_{Z}X + {}^{0}_{-1}e \longrightarrow {}_{Z-1}^{A}X' + \text{X ray}$	
Gamma emission	${}^{0}_{0}\gamma$	${}^{A}_{Z}X^{*} \xrightarrow{\text{Relaxation}} {}^{A}_{Z}X' + {}^{0}_{0}\gamma$	
Spontaneous fission	Neutrons	${}^{A+B+C}_{Z+Y}X \longrightarrow {}^{A}_{Z}X' + {}^{B}_{Y}X' + C{}^{1}_{0}n$	

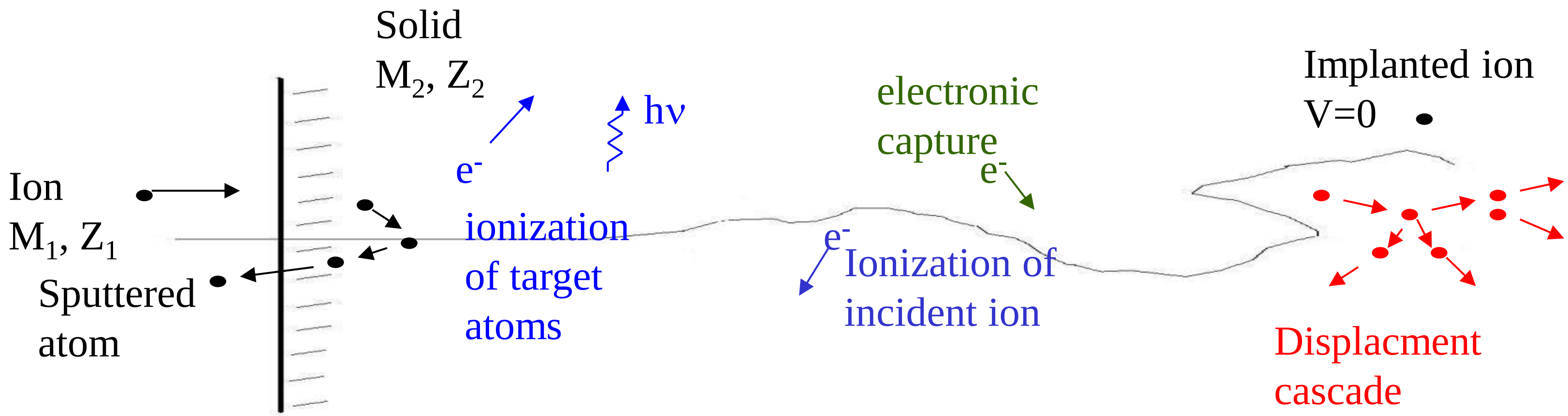
Minor actinides: mainly α decays

Fission products: mainly β decays

Most of alpha and beta decays

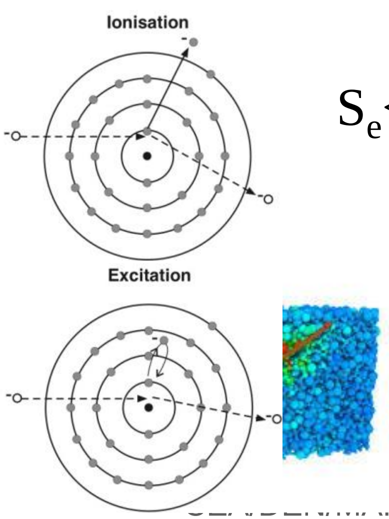
Interaction with matter

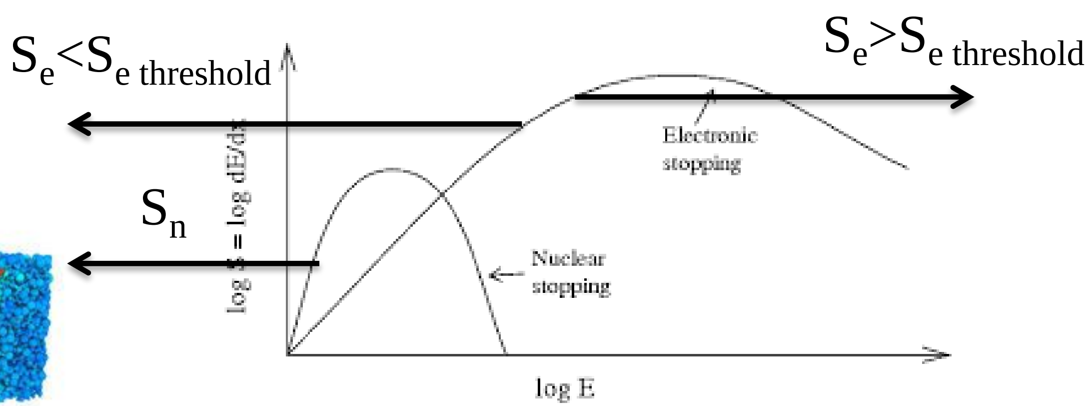
Due to the various decays: Emission of particles with high amount of energy

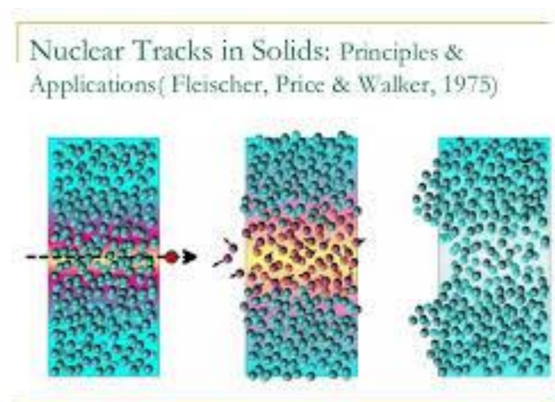


$Se = (dE/dx)_{elec}$ = Electronic energy loss due to collisions with electrons

$Sn = (dE/dx)_{nucl}$ = Nuclear energy loss due to collisions with atoms





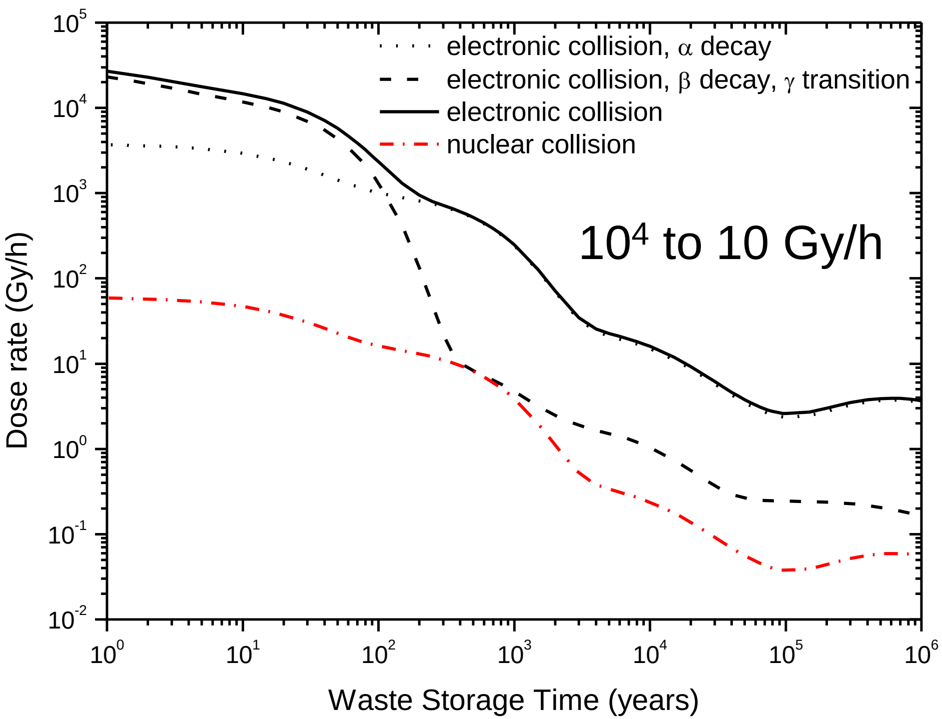


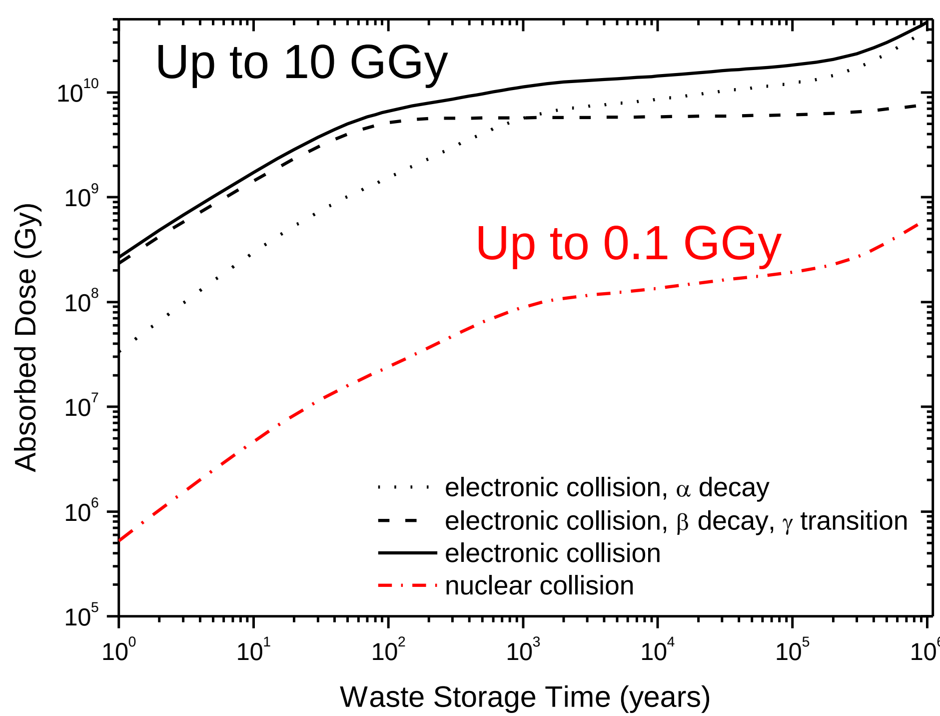
Interaction with matter

Important parameters to consider:

- **Dose rate** : absorbed energy per unit of mass of material per unit of time (Gy/s)

- **Dose** : absorbed energy per unit of mass of material (Gy = J/kg)

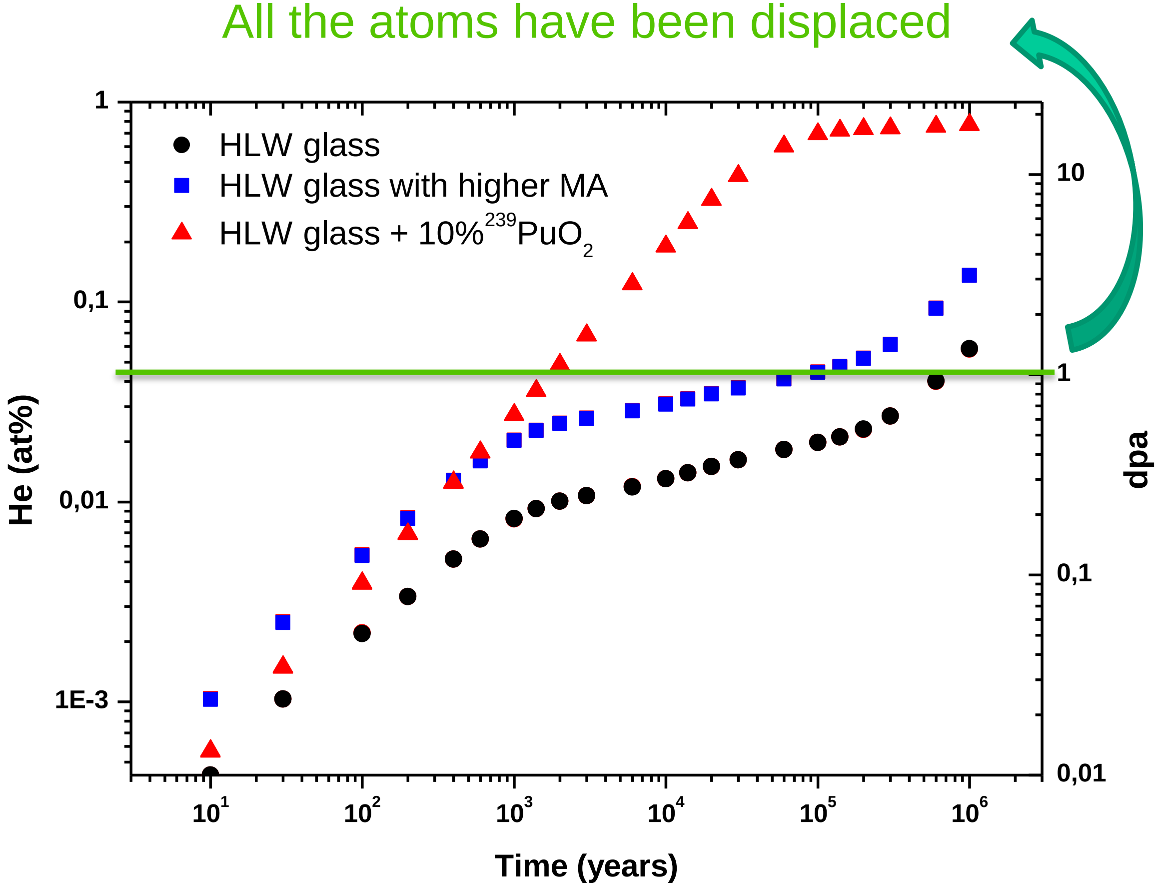




- Nuclear collisions, **dpa** = **displacements per atom**

$$\text{dpa = displacements per atom} = \frac{\text{Number of displaced atoms in volume from NRT equation}}{\text{Number of materials atoms in same volume}}$$

All the atoms have been displaced



$$N_d(T_d) = \begin{bmatrix} 0 & , & T_d < E_d \\ 1 & , & E_d < T_d < 2E_d/0.8 \\ \dfrac{0.8T_d}{2E_d} & , & 2E_d/0.8 < T_d < \infty \end{bmatrix}$$

$$T_D = F_{D,n} = E_0 - F_{D,e}$$

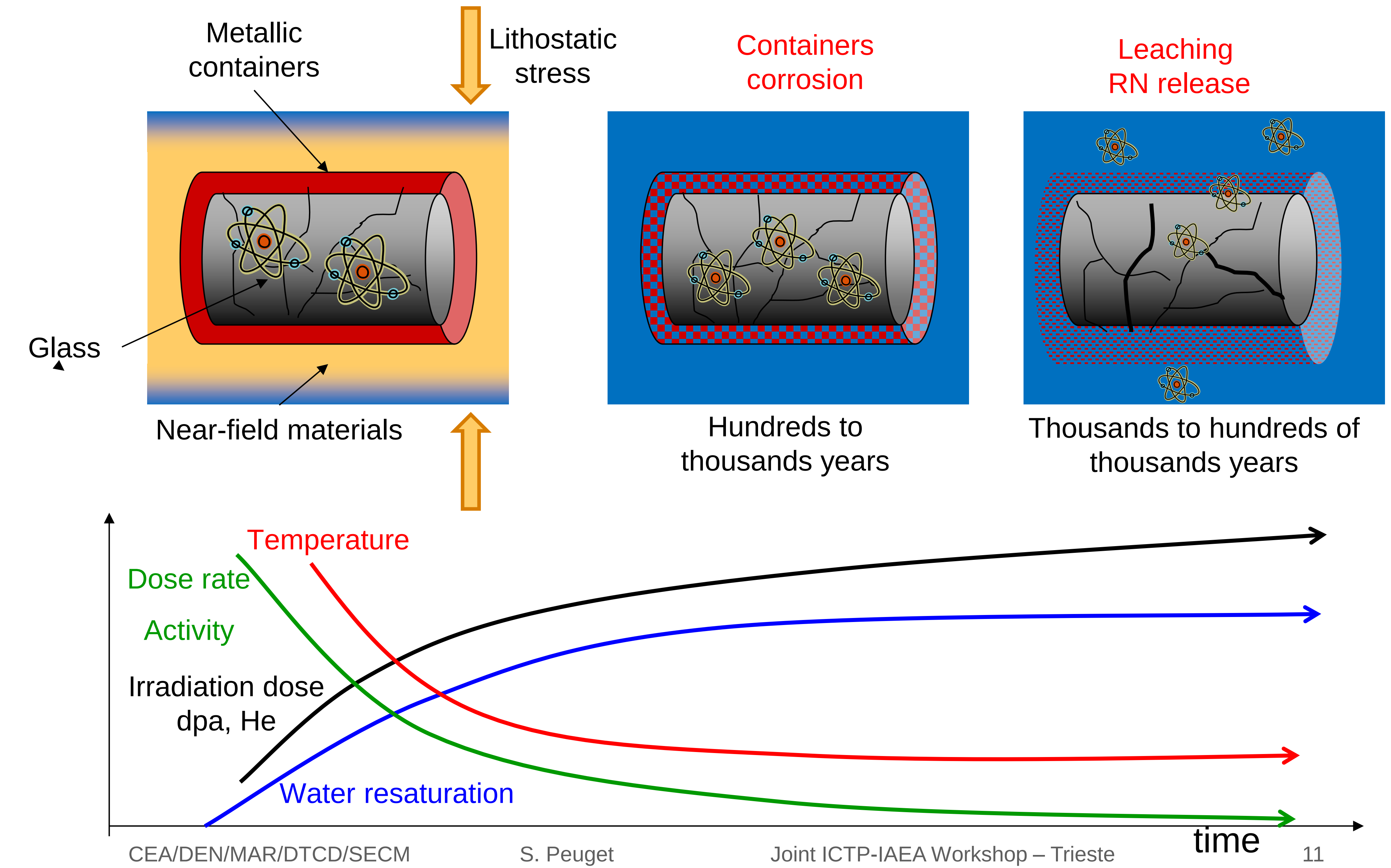
T_D=Energy available for damage production

E_0=Energy of the particle

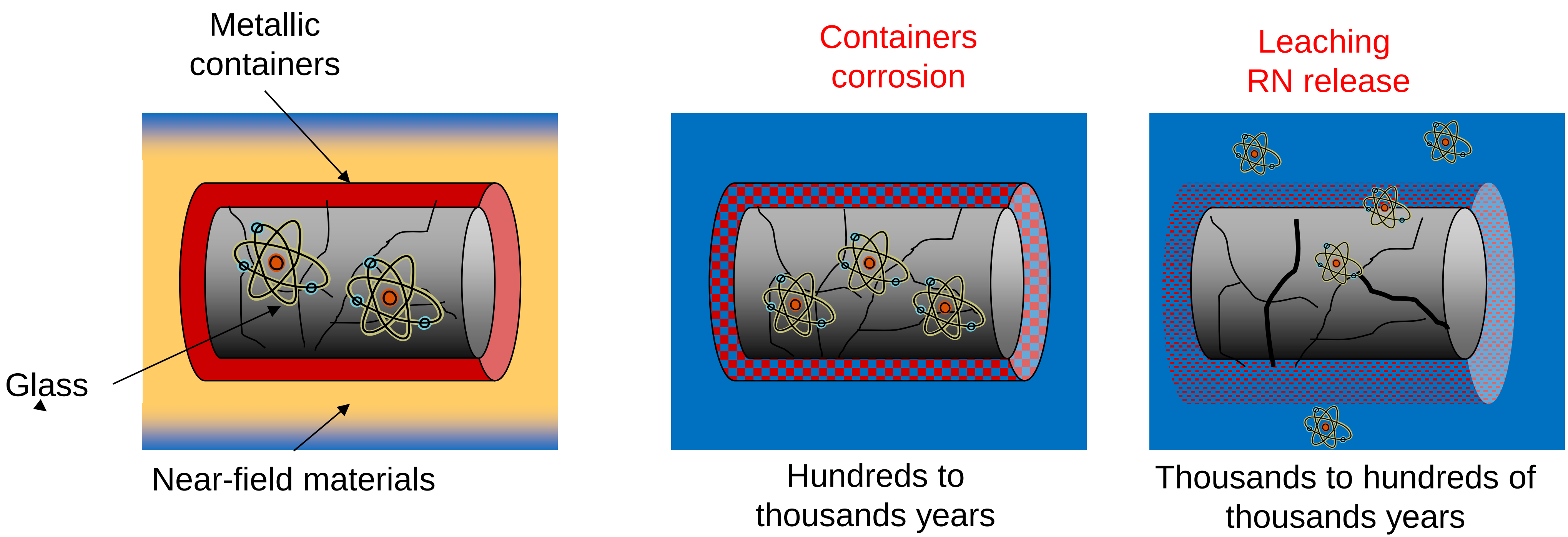
$F_{D,e}$=Energy lost to electronic stopping

E_d=Threshold displacement energy

Nuclear Glass or GCM: a complex ageing scenario



Nuclear Glass or GCM: a complex ageing scenario



Q1: Stability of the metastable glassy state under irradiation (dose and dose rate) ?

Q2: Waste mechanical degradation? Cracking due to Rad Effects and He generation?

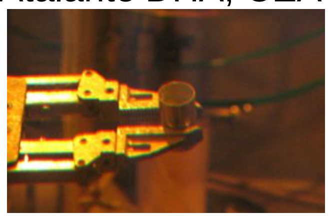
Q3: Effect of radiation on the confinement properties? Leaching behavior? Coupling with the surrounding materials?

Methodology to study radiation effects at CEA

- **Accelerate the time scale**
- **Dissociate the effects of beta and alpha decays (*electronic / nuclear stopping power*) and *helium generation***
- **Evaluate the effects on the confinement properties and the glass structure**

Propose some models to explain the glass behavior under irradiation

Atalante DHA, CEA



1. Actinide doped glasses

^{244}Cm, ^{239}Pu, ^{241}Am…

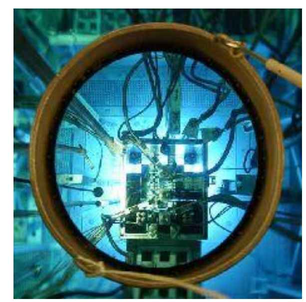
JANNuS Saclay, Orsay, GANIL, SIRIUS

1. External irradiation

with electrons or light and heavy ions

He Au, Kr

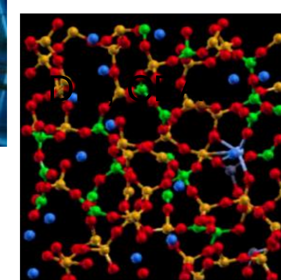
OSIRIS, CEA



3. In pile irradiation : $^{10}B(n,\alpha)^{7}Li$

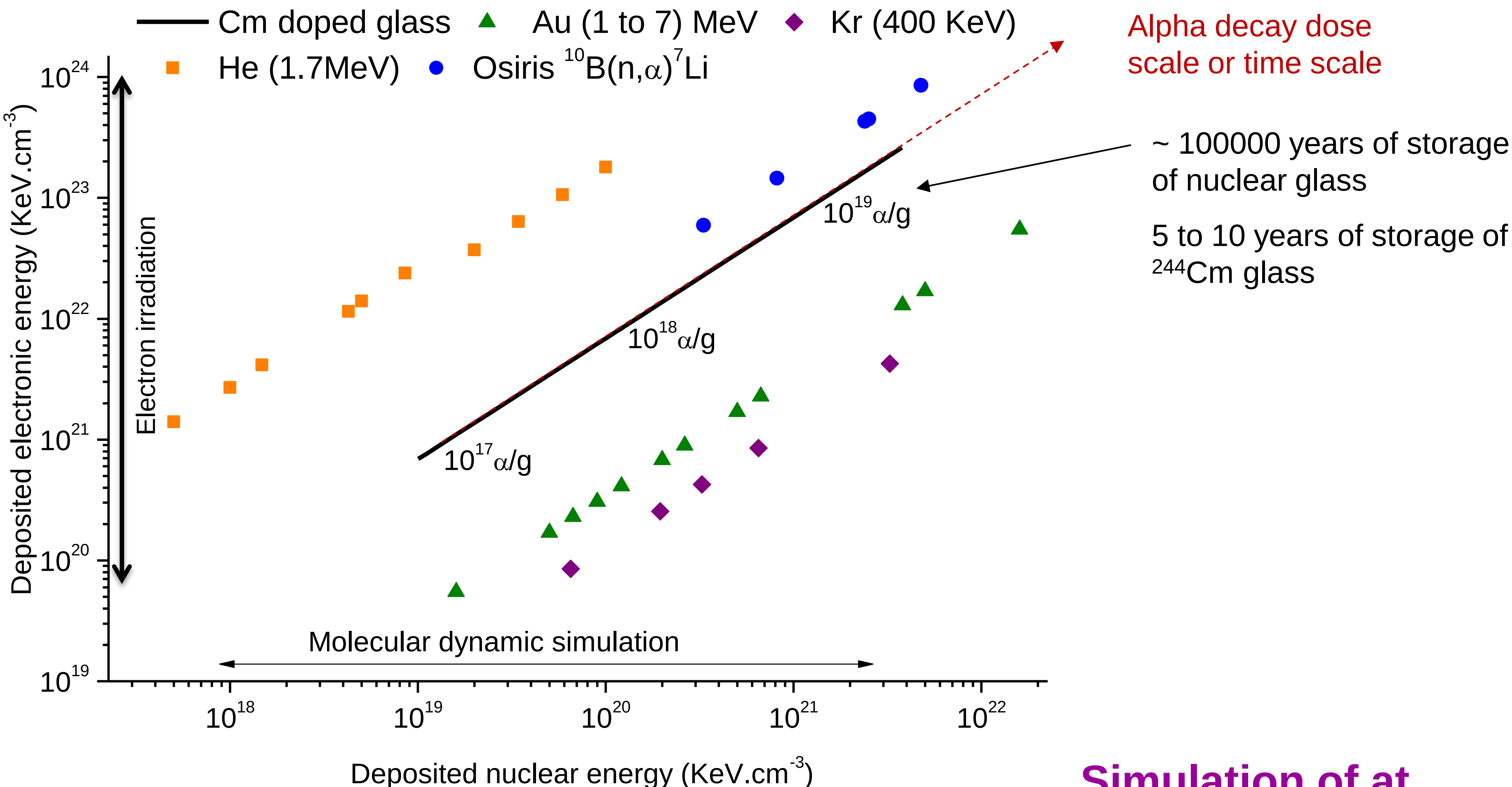
4. Molecular dynamic modeling of ballistic effects

MD, CEA



Comparison

Methodology to study radiation effects at CEA



Simulation of at least 100000 years of disposal by various methods !

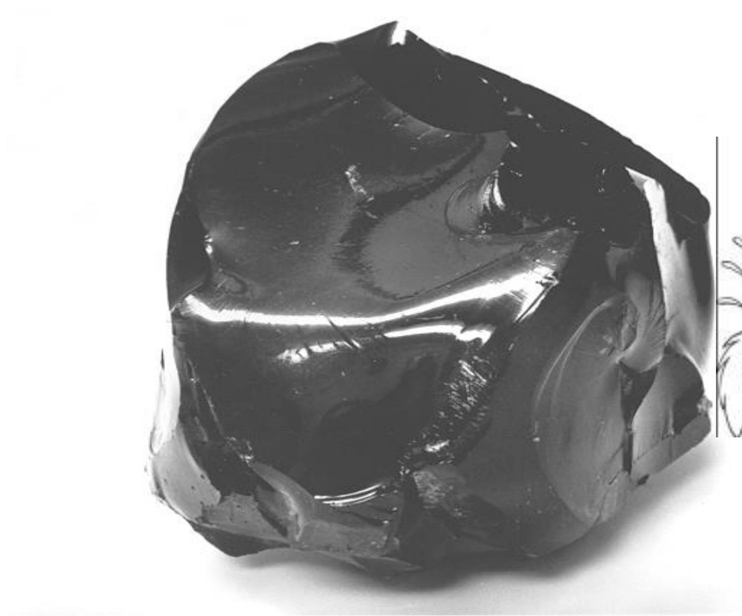
Light ions irradiations (He) : mainly electronic interactions

Heavy ions irradiations (Kr, Au) : mainly nuclear interactions

Doped glasses and OSIRIS irradiation : electronic and nuclear interactions

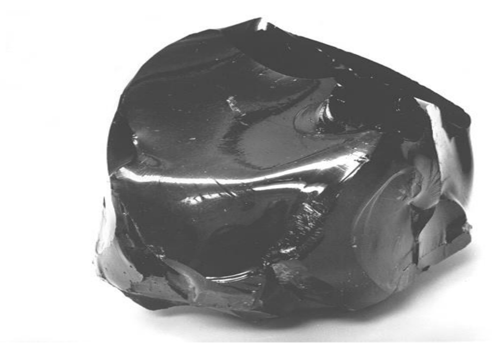
Molecular Dynamics : only nuclear interactions

Nuclear Glass or GCM under irradiation: What do we know?



Q1: Stability of the metastable glassy state ?

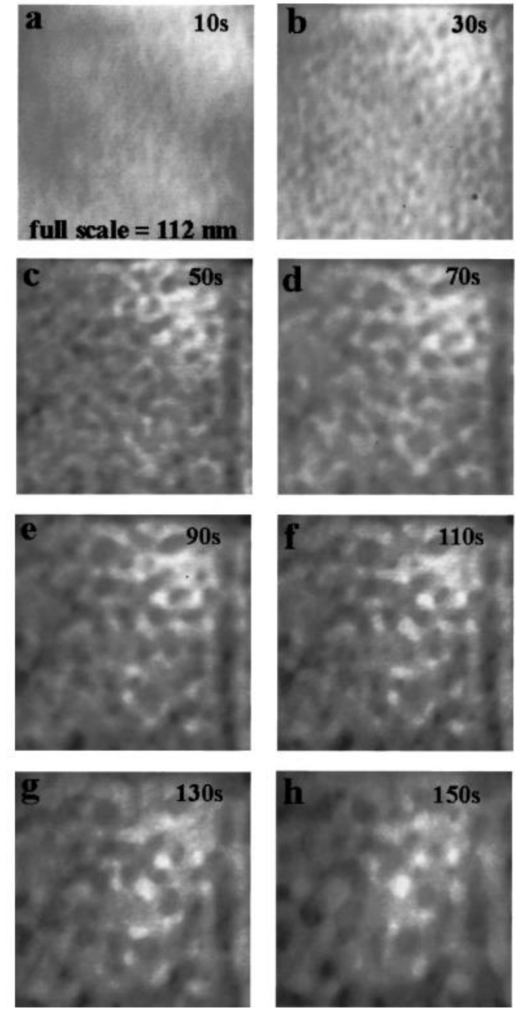
- How radiation can affect the glassy state?
- Can irradiation favor or reduce the crystallization, the phase separation of a glass?
- Can irradiation induce a radiolytic decomposition of the glass? Oxygen bubbles…
- What is the effect of the dose and dose rate on these processes? How to extrapolate to the storage conditions?



Q1: STABILITY OF THE METASTABLE GLASSY STATE ?

Phase separation, bubble formation ? e$^-$ irr. very high dose rate: 10^9 x greater

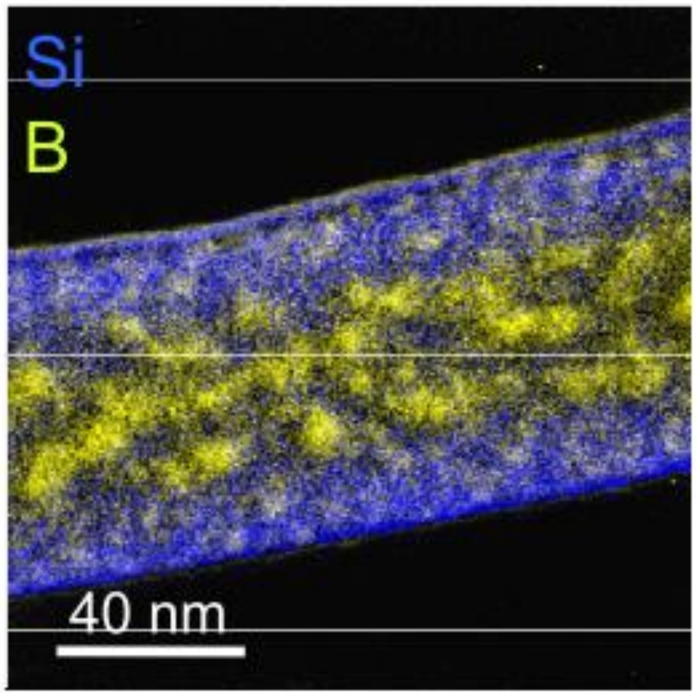
Ca-Borosilicate Glass



Observed under electron irradiation TEM studies

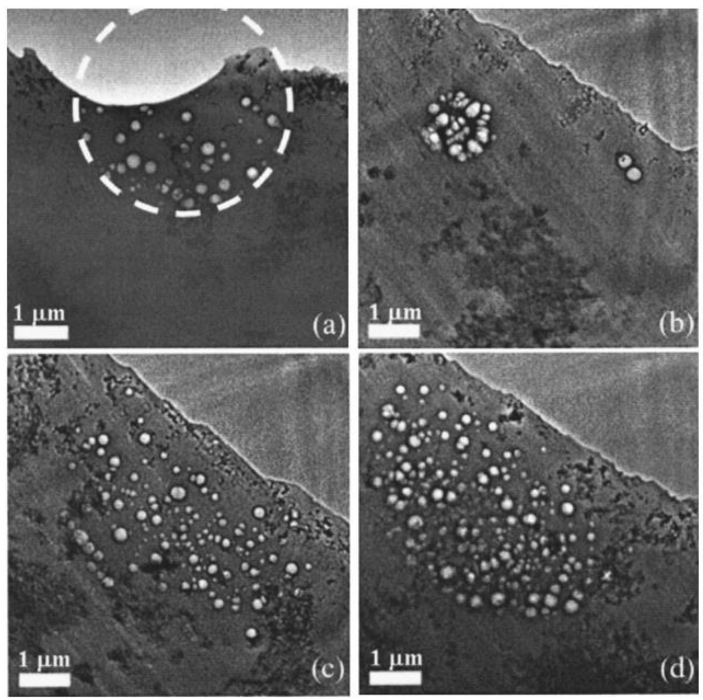
Highly dependent of the glass composition

Na-Borosilicate Glass



Sun, Microscopy and Analysis 106 (2005)

Li-Borosilicate Glass



*Ollier, JAP **99** (2006)*

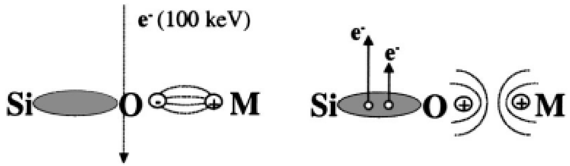
*Jiang, JAP **92** (2002)*

Q1: STABILITY OF THE METASTABLE GLASSY STATE ?

Phase separation, bubble formation ? <u>e⁻ irr. very high dose rate: 10^9 x greater</u>

Several contributions:

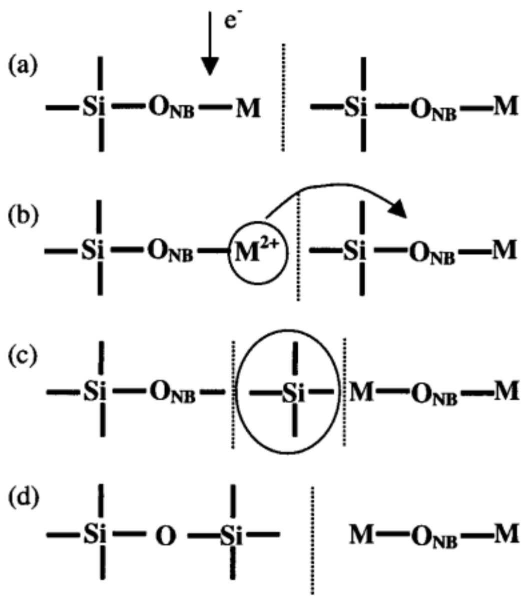
- **Bond breaking, alkali migration** *Jiang, JAP **92** (2002)*



- **Modification of the glass chemical composition**

(Electron Stimulated Desorption) *Sun, NIMB 218 (2004)*
Mir, JNCS 453 (2016)

Favors oxygen bubbles and phase separation

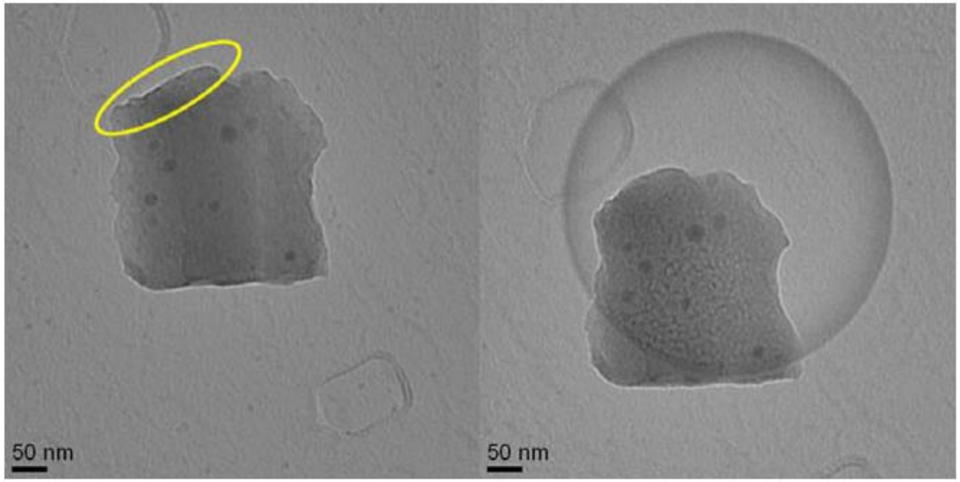


Q1: STABILITY OF THE METASTABLE GLASSY STATE ?

Phase separation, bubble formation ? <u>**e^- irr. very high dose rate: 10^9 x greater**</u>

Several contributions:

- **Modification of the glass viscosity**
  - **Very high dose rate (10 orders of magnitude higher than expected in HLW glass)**
  - **Bond breaking**
  - **Chemical composition changes**



Favors oxygen bubbles and phase separation

$$\eta_R(T) = \eta(T)/\left[1+\alpha_e I_e\left[1+C\exp\left(D/RT\right)\right]\right]$$

η(T) viscosity of an non-irradiated material,
α_e efficiency of electron beam bond breaking and annihilation
$A_e I_e$ dimensionless electron flux density

Ojovan, Mater. Res. Soc. Symp. Proc. Vol. 1193 (2009)
MÖbus, JNM 396 (2010)

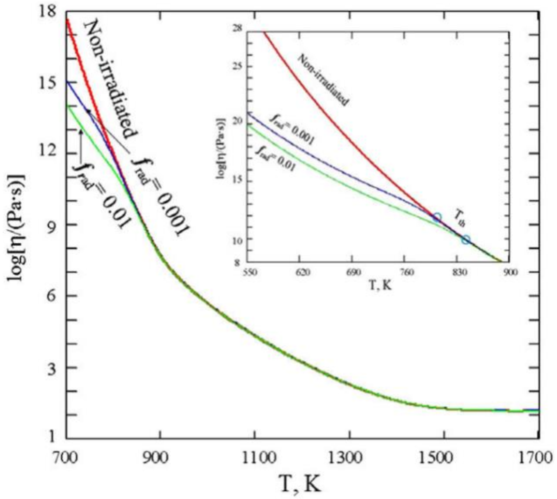


Fig. 9. Viscosities of non-irradiated and electron beam irradiated amorphous soda lime silicate system $70SiO_2 21CaO 9Na_2O$ as a function of temperature for two dimensionless electron flux densities ($f_{rad} = \alpha_e I_e$) 0.01 and 0.001.

Q1: STABILITY OF THE METASTABLE GLASSY STATE ?

Phase separation, bubble formation ? e⁻ irr. « lower » dose rate 10^4 x greater

Irradiation by e⁻ @ 2.5 MeV   up to 4,5GGy

- **Surface phenomena :**

ESD of Na^+ ions in the first µm at the surface

Modification of the glass composition

Tendancy of phase separation in the first µm

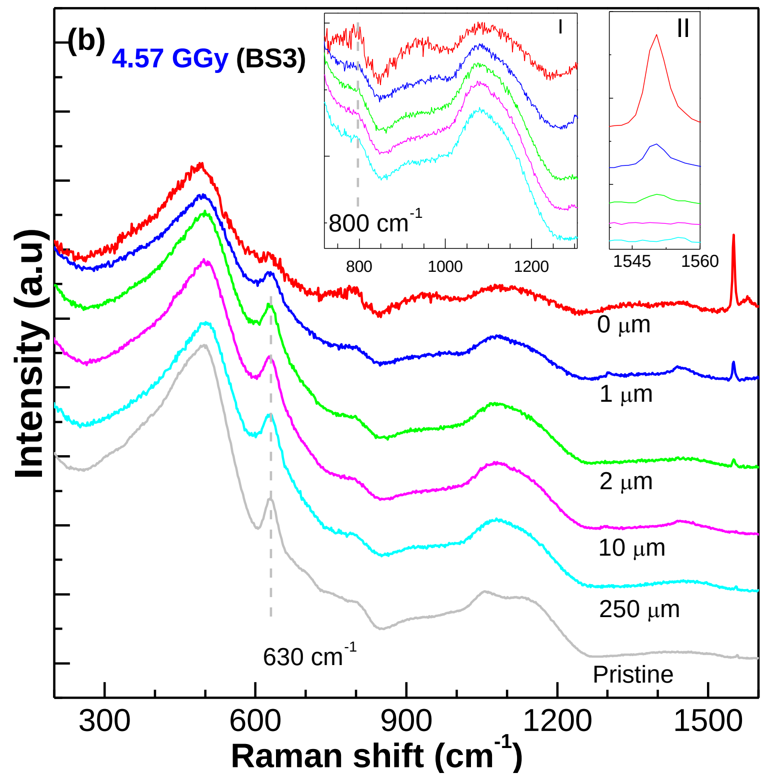
More important for simple glass

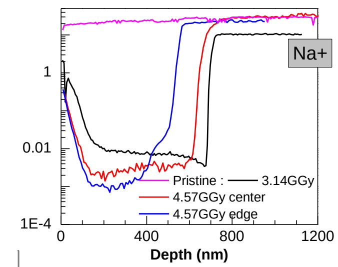
- **Bulk :**

No phase separation in the bulk,

Decrease of boron coordination and increase of NBO around Si and B atoms

Mir et al, JNCS 2016





Q1: STABILITY OF THE METASTABLE GLASSY STATE ?

Summary:

Phase separation, bubble formation in nuclear glasses: under beta irradiation

- **Only observed under TEM studies and very sensitive to the glass composition**

10 orders of magnitude higher dose rate than expected in HLW glass

- **No phase separation observed with electron accelerator based studies**

- **Depletion of alkali atoms at the irradiated surface**

Is it representative of long term ageing of nuclear glass?

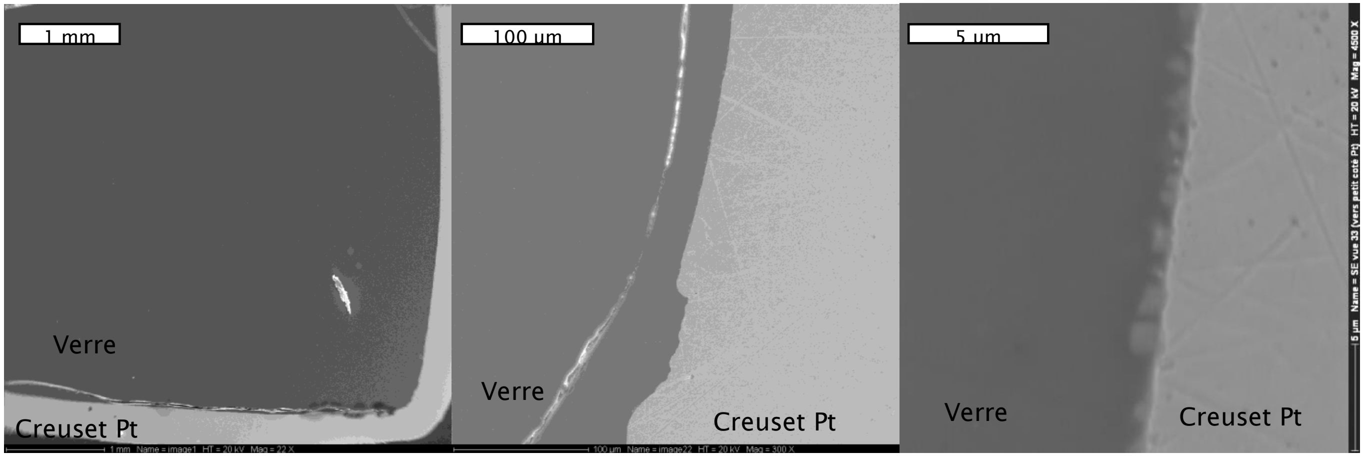
Need to characterize old radioactive glasses

30 years of storage time:

~3GGy ~half of the dose @ 1000years

STABILITY OF THE METASTABLE GLASSY STATE ?

Phase separation, bubble formation ? alpha decays irradiation

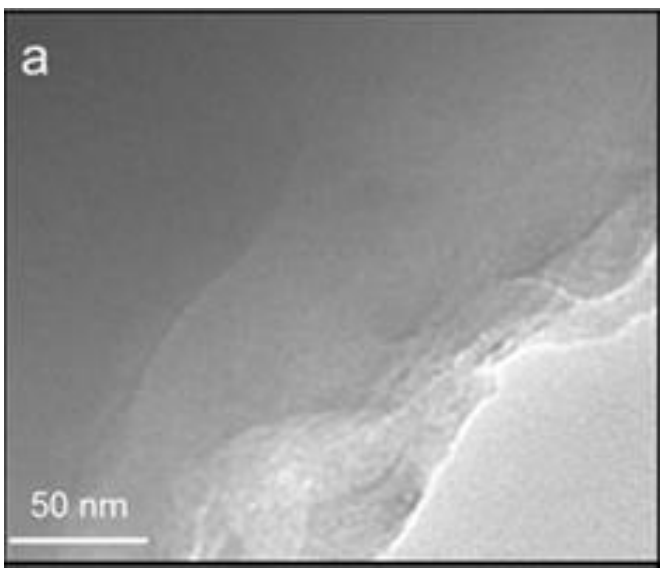


(Around 100000 years of storage)

S. Peuget et al. JNM 44 (2014)

^{244}Cm SON 68 glass : SEM (CEA Marcoule), alpha decay dose $2x10^{19}\alpha/g$

TEM (ITU Karlsruhe), alpha decay dose $8x10^{18}\alpha/g$

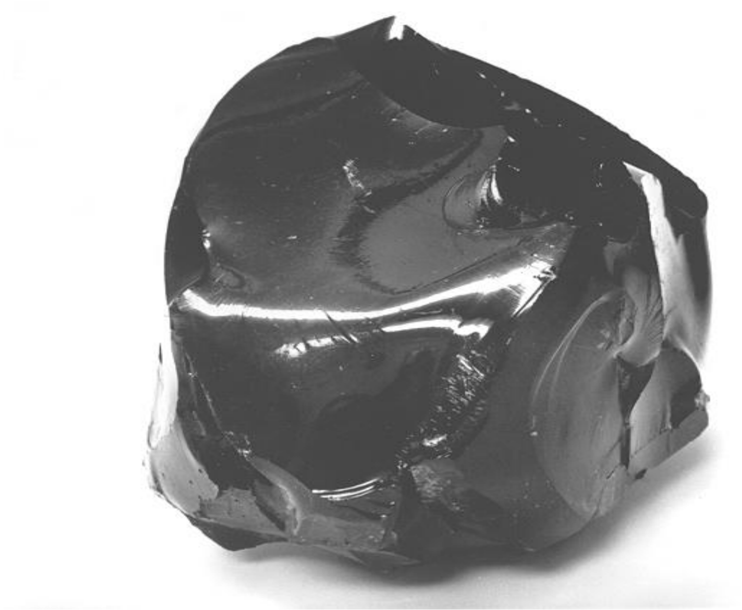


Homogeneous microstructure, without bubbles, phase separation or crystallization

Very good stability of the glassy state

ACTINET i3
Integrated Infrastructure Initiative for Actinide Science

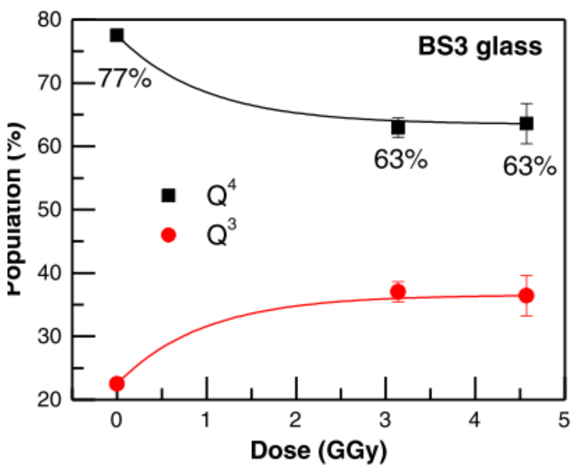
Characteristics of the irradiated glassy state?

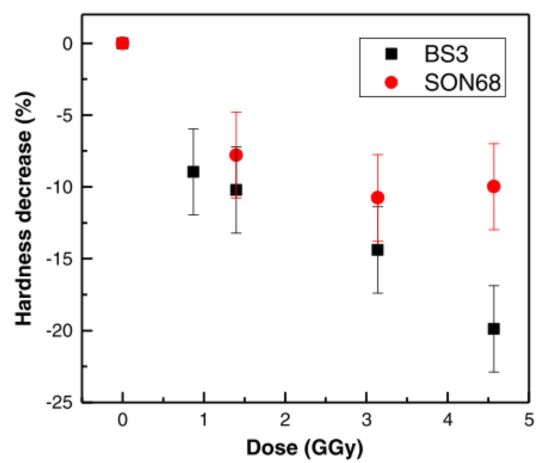


Characteristics of the irradiated glassy state? e^- irradiation

Main effects observed under electron irradiation:

- ✓ Formation of punctual defects and molecular oxygen
- ✓ Changes in oxidation states (Fe^{2+}/Fe^{3+}, $Ln^{2+}/Ln^{3+}/Ln^{4+}$…)
- ✓ Changes in boron coordination number and glass polymerization index
- ✓ Enhanced defect and atomic mobility (enhanced alkali migration)
- ✓ A saturation effect with dose: stabilization after 3 to 4GGy





Mir et al, JNCS 453 (2016)

- ✓ Complex glasses are less modified than simple glasses

Characteristics of the irradiated glassy state? e^- irradiation

Why complex glasses are less modified than simple glasses?

SON68 *[Jacquet-Francillon, Radiochimica Acta 25 (1978), Ollier, J.Non-Cryst. Solids 323 (2003)]*

SiO_2 *[Shelby, J. Appl. Phys. 51 (1980) 2561]*

Borosilicates *[Boizot, NIMB 166 (2000), JNCS 283 (2001), Mohapatra NIMB 269 (2011), Chen, Chin. Phys. B 22 (2013)]*

Origine of the evolution of simple glasses?

Ionizing radiation

↓

Exciton creation (e^-/h^o)

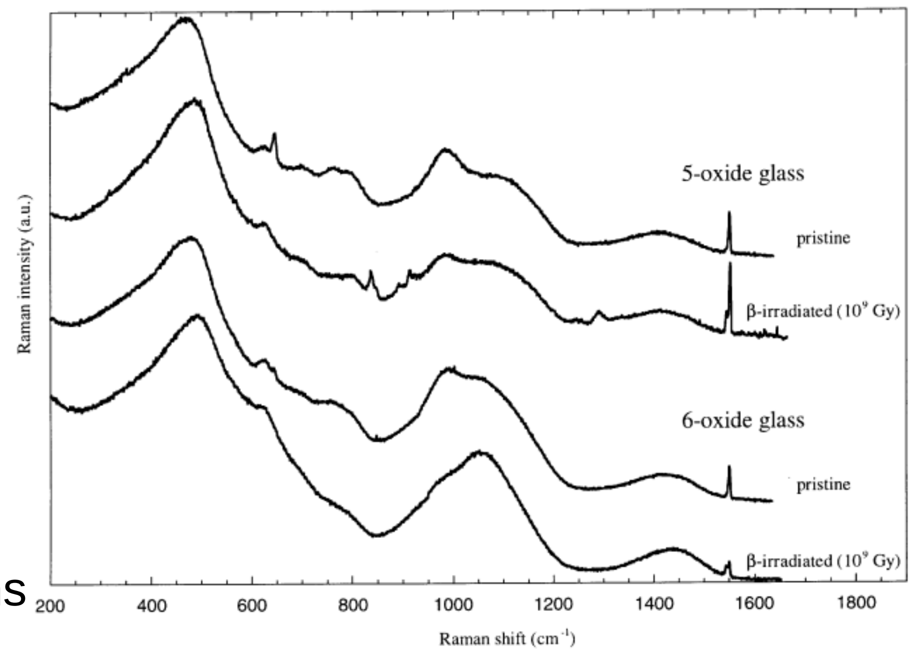
↓

Ponctual defects, alkali migration

Accumulation with dose…

↓

Structural and macroscopic properties evolutions



Characteristics of the irradiated glassy state? e⁻ irradiation

Why complex glasses are less affected than simple glasses?

Effect of incorporation of transition élements (Fe, Cr), lanthanides, mixed alkaline

Olivier. JNCS 351 (2005)

Ionizing radiation

↓

Exciton (e^-/h^o) but interaction with the transition elements

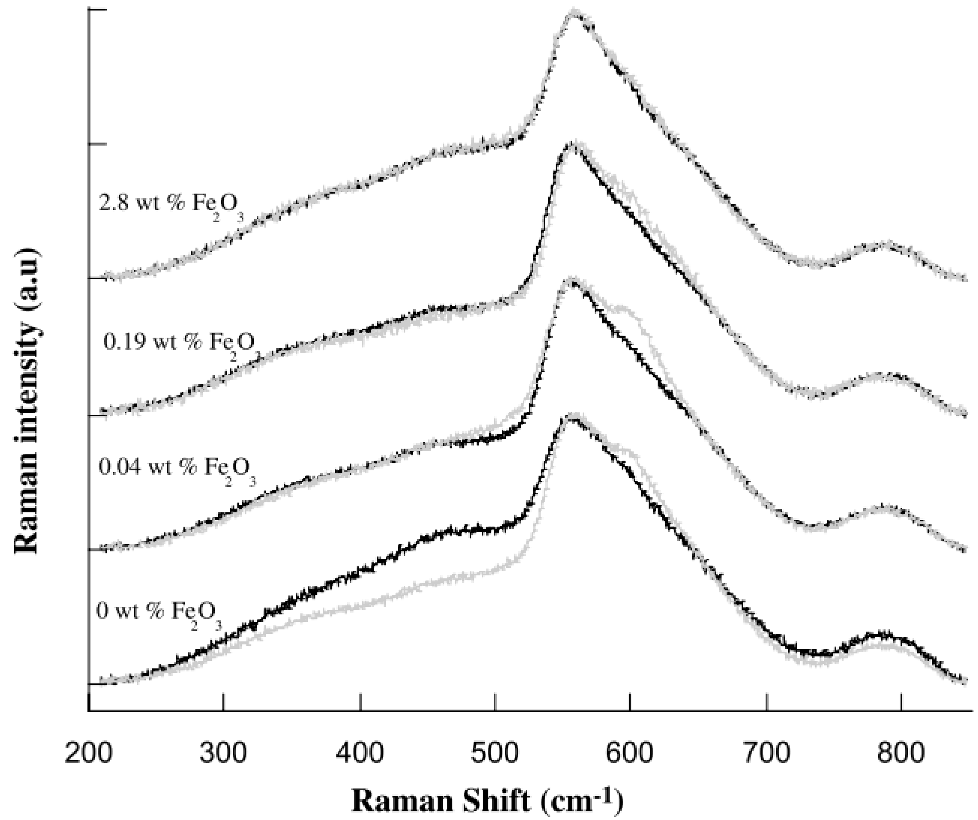
Equilibrium:

$Fe^{3+} + e^- \longrightarrow Fe^{2+}$

$Fe^{2+} + h^o \longrightarrow Fe^{3+}$

↓

Decrease or vanishing of structural evolution

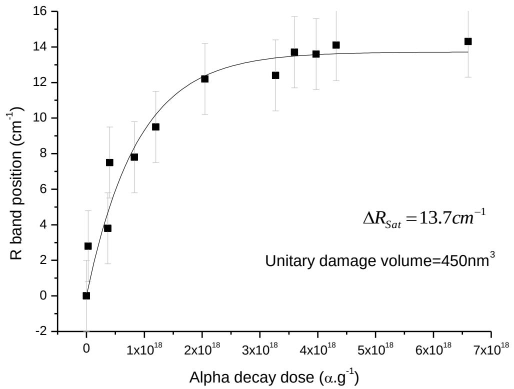


The glass complexity (transition elements, lanthanides, mixed alkalines …) increases the glass resistance toward ionizing radiation

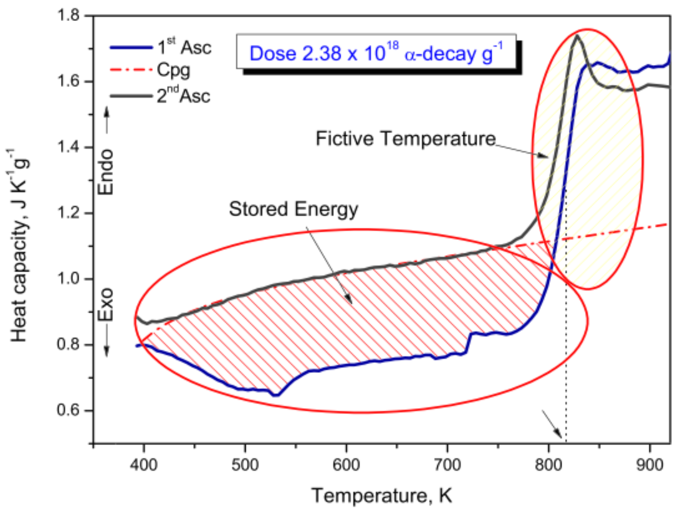
Characteristics of the irradiated glassy state? α irradiation

Main effects observed under alpha decay irradiation:

- ✓ **A saturation effect with dose, a new glass structure is reached after around 4×10^{18} α/g (Nuclear dose ~30 MGy)**



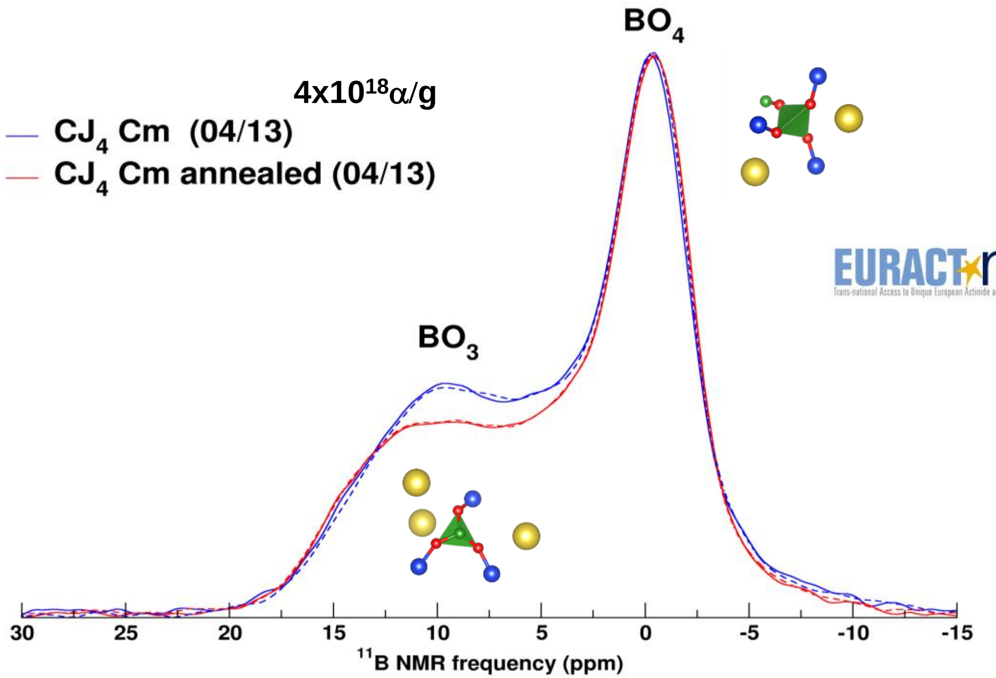
- ✓ **No effect of the dose rate in the relevant range**
- ✓ **Changes at both Short Range Order and Medium Range Order**
  - ✓ Changes in boron coordination number and glass polymerization index
  - ✓ Changes in ring statistic, angle distribution

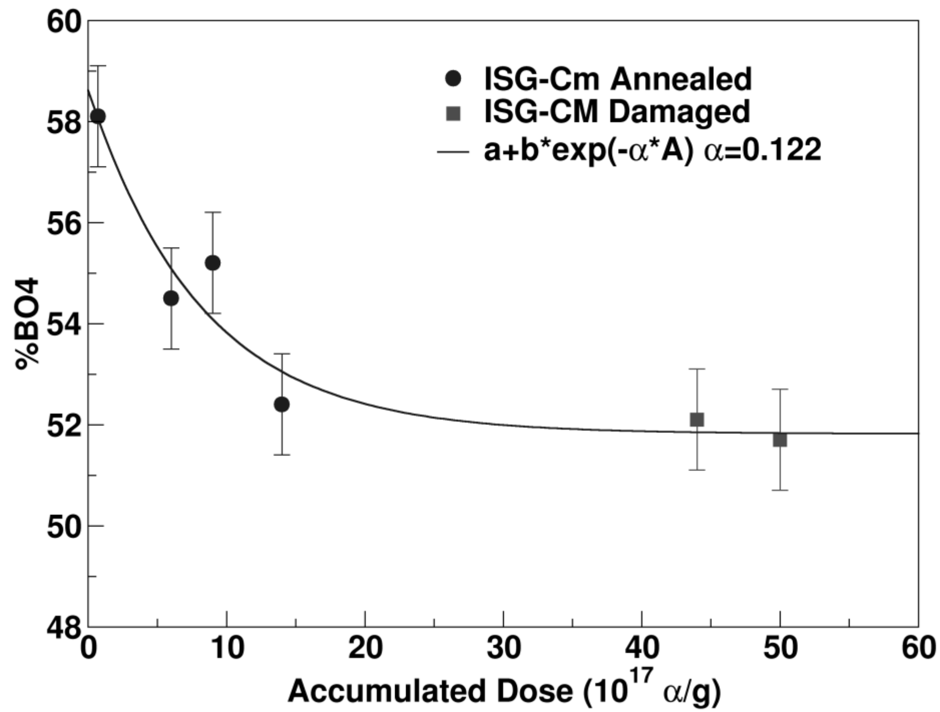


- ✓ **A higher fictive temperature after irradiation**
- ✓ **Stored energy of ~100J/g**
- ✓ **Complex glasses are less modified than simple glasses**

Characteristics of the irradiated glassy state? α irradiation

Main effects observed under alpha decay irradiation: SRO





^{244}Cm ISG glass

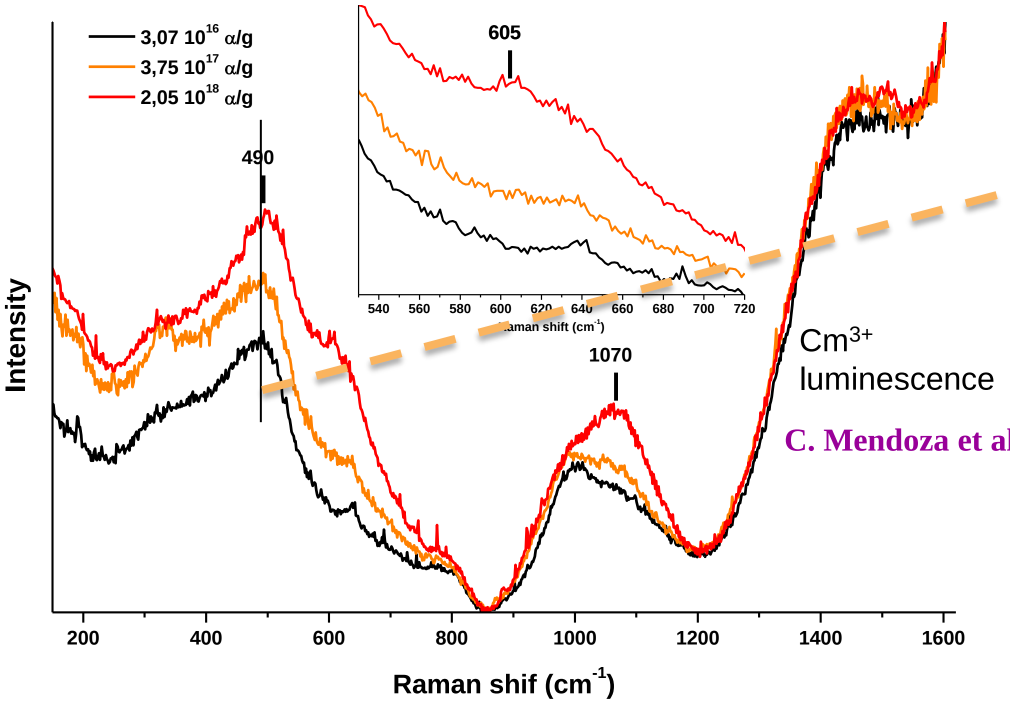
T. Charpentier et al. Scientific Reports 6:25499 (2016)

Partial conversion of BO_4 into BO_3

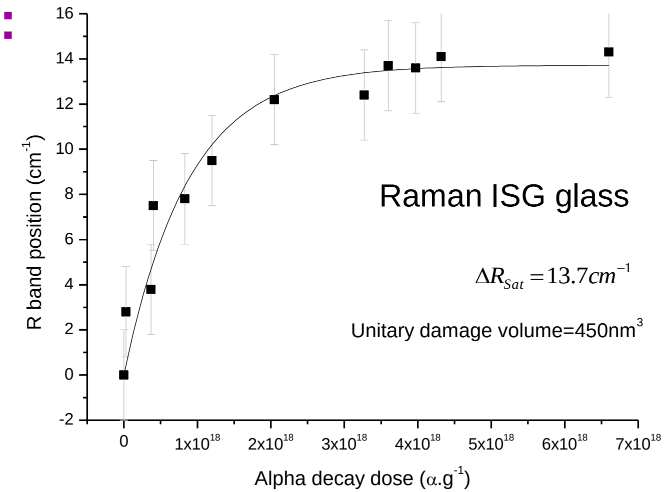
Characteristics of the irradiated glassy state? α irradiation

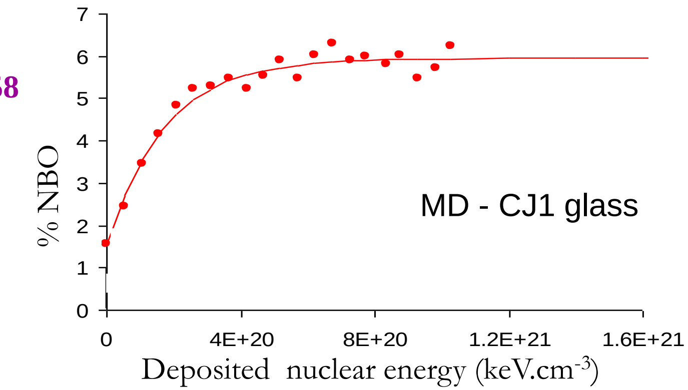
Main effects observed under alpha decay irradiation:

Raman spectroscopy, Cm doped ISG



C. Mendoza et al. Proc. Chem. 7 (2012) 58





J.-M. Delaye et al, J. Non-Cryst. Solids 357 (2011) 2763

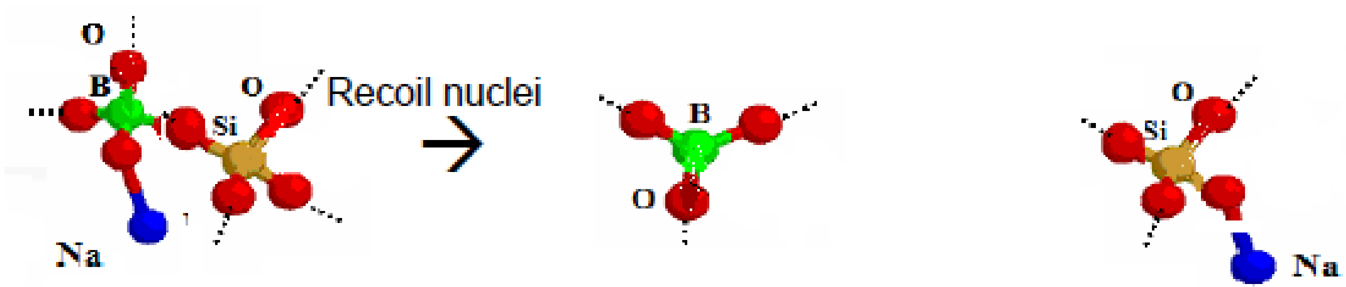
- Increase of Q3 contribution in ISG glass : <u>more NBO</u>
- Slight shift of the vibration band around $500 cm^{-1}$

  <u>Decrease of the mean angle between silica tetrahedra</u>
- New D2 band on ISG Cm doped glass: <u>3 members silica rings</u>
- Stabilization of the silicon local environment after around 4×10^{18} α/g

Characteristics of the irradiated glassy state? α irradiation

Modification of the Short Range Order

Increase of trigonal boron, increase of NBO



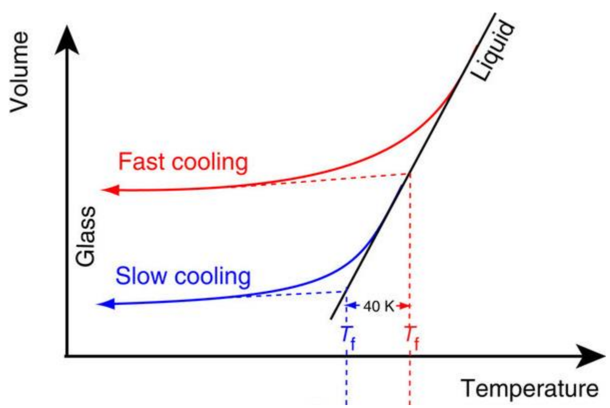
Modification of the Medium Range Order

Ring statistic modification, increase of glass disorder and Si/B mixing

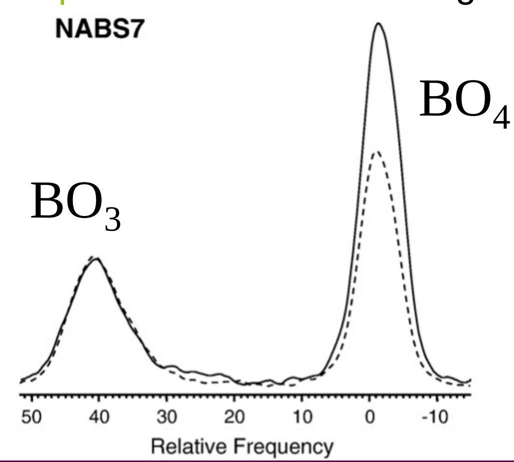
Effects similar to those induced by thermal quenching of a molten glass

$BO_4 \xrightarrow{\nearrow T} BO_3 + NBO$

Decrease of the glass density



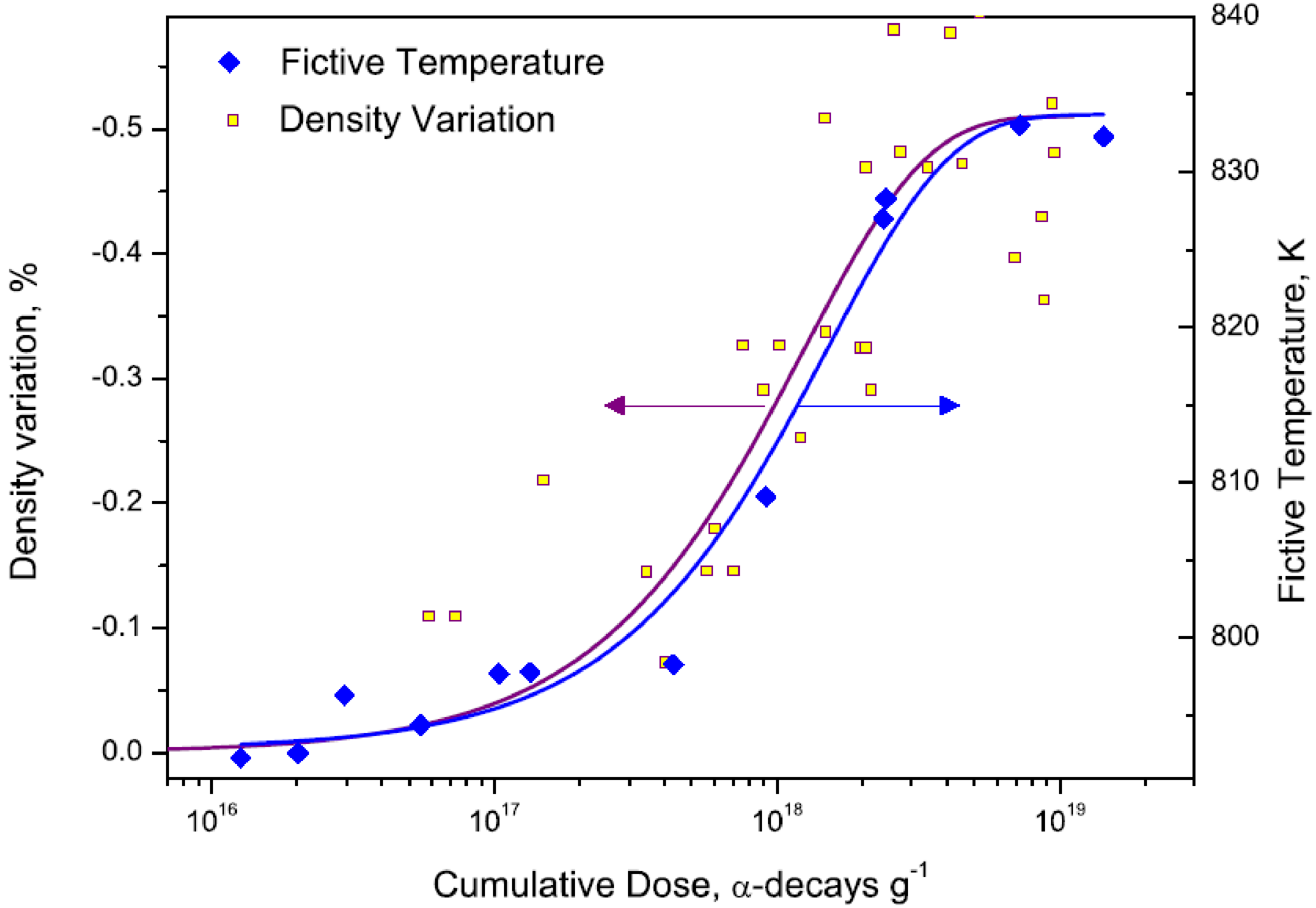
^{11}B NMR on quenched and annealed glass



Wu and Stebbins JNCS 356 (2010)

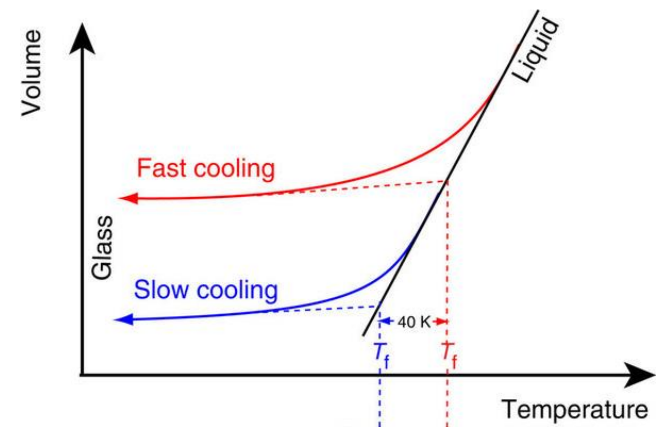
Characteristics of the irradiated glassy state? α irradiation

DSC on ^{244}Cm doped SON68 glass (ITU, actinet-i3 project)



Maugeri et al, J. Am. Ceram. Soc. 95 (2012) 2869

ACTINET i3
Integrated Infrastructure Initiative for Actinide Science



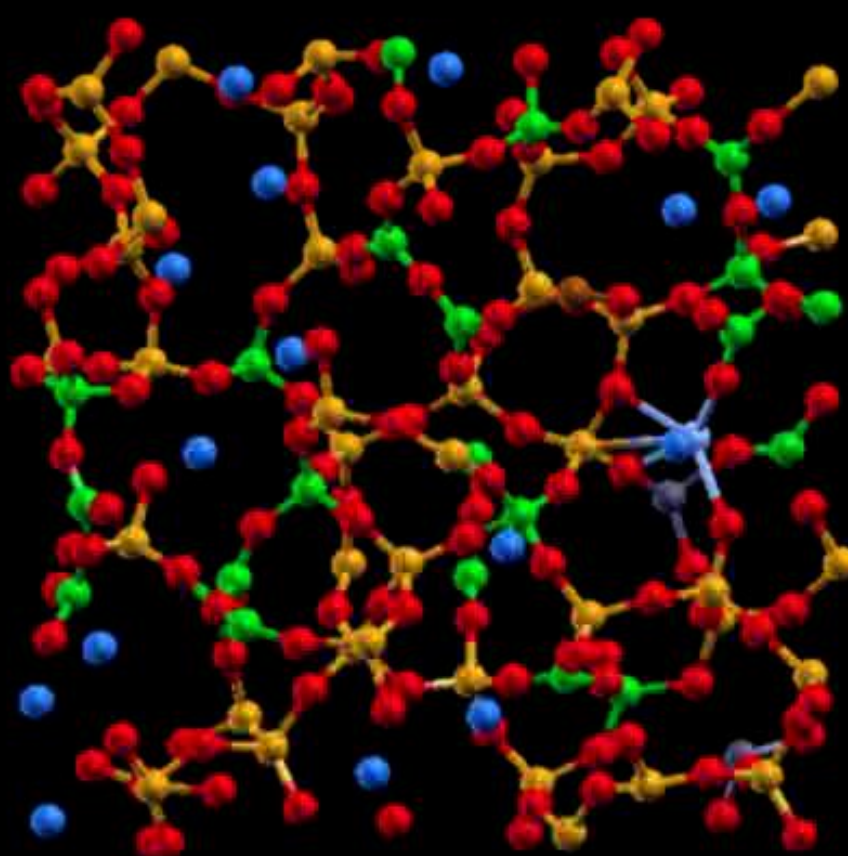
Increase of the glass fictive temperature with alpha decay dose

Formation a new structure similar to a fast quenched glass

Characteristics of the irradiated glassy state? α irradiation

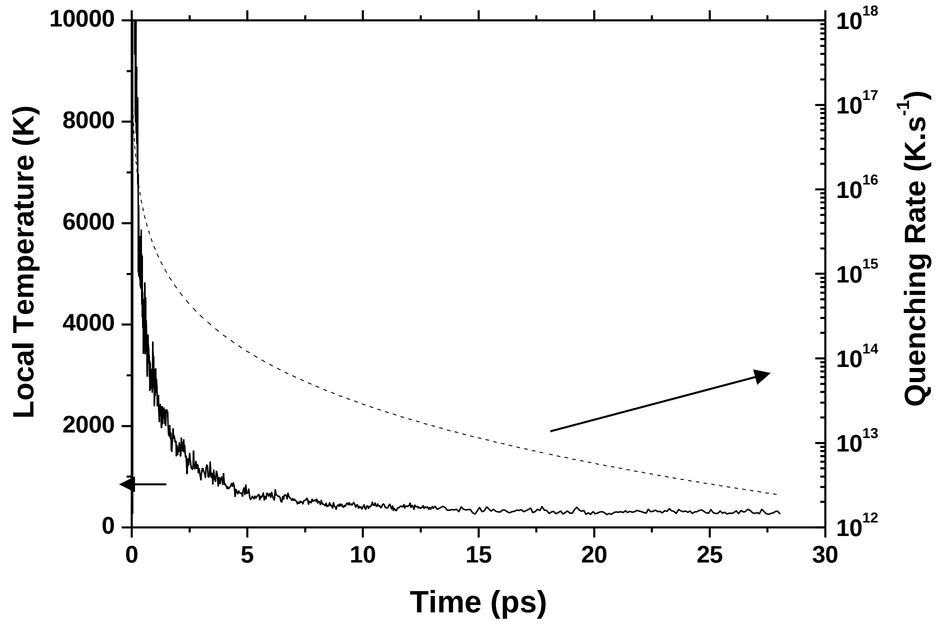
What happen in the displacement cascade induced by a recoil nuclei?

JM Delaye, PRB 61 (2000) 14481



1. Balistic phase
2. Thermal phase

J.M Delaye lecture 2



Golden = Si
Green = B
Blue = Na
Red = O

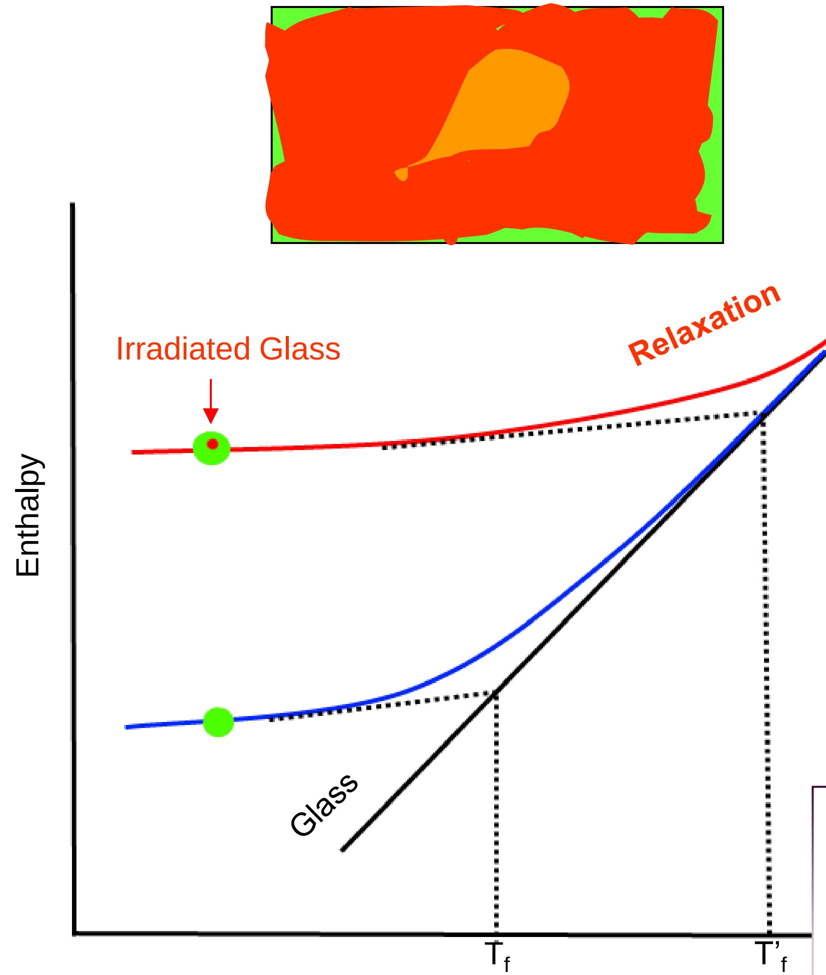
Very high quenching rate of the disordered state induced by the displacement cascade

« Supervitrification »

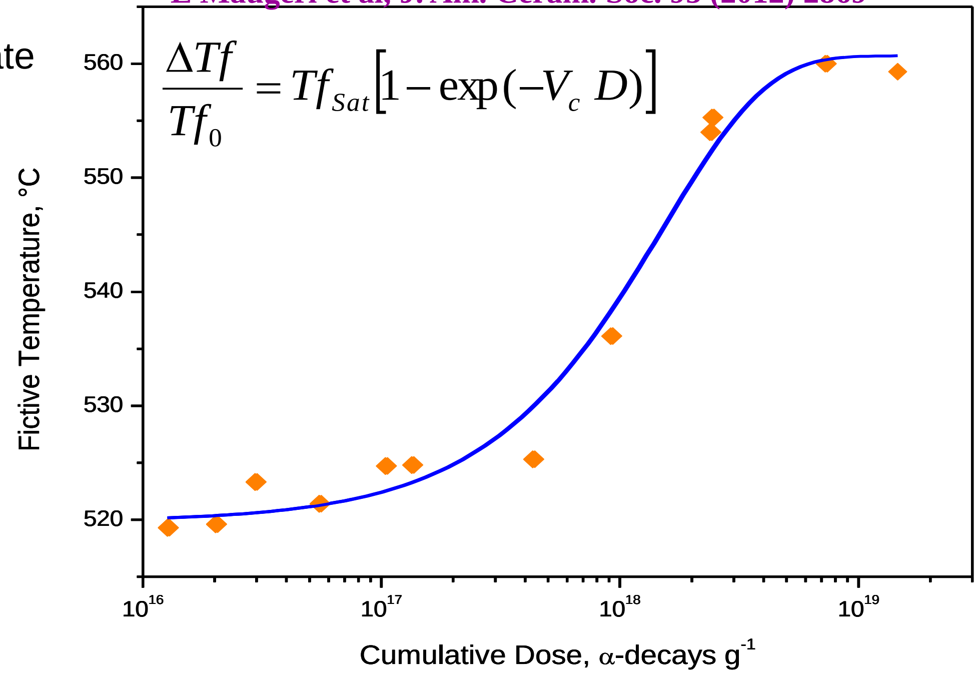
Understanding of glass behavior under alpha decays

1. Balistic step : disordered state
2. Relaxation step : very important quenching rate

Irradiated zone has a higher fictive temperature



E Maugeri et al, J. Am. Ceram. Soc. 95 (2012) 2869



Model of accumulation of ballistic disordering fast quenching events: "supervitrification"

Stabilization of a new glass structure when all the volume has been damaged once

Open questions:

Which step control the irradiated state?

Energy deposition step, quenching step?

Any rôle of alpha particle?

A.H. Mir lecture

Nuclear Glass or GCM under irradiation: What do we know?

Q1: Stability of the metastable glassy state ?

Except at very high dose rate (not relevant), no phase separation or devitrifiction observed

Depletion of alkali atoms at the surface of some electron irradiated glasses, Is it representative of the disposal conditions?

For all irradiation conditions, formation of a new glassy structure with slight modifications of SRO and MRO, higher fictive temperature, stored energy is significant

The glass chemical complexity has a positive effect on the radiation changes

Effect of beta and alpha decays mainly studied separately up to now, but the ageing scenario under irradiation is more complexe

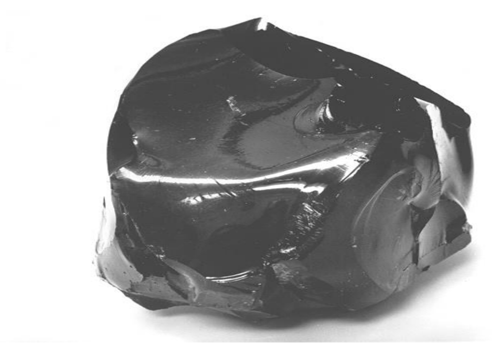
A.H. Mir lecture

→ **Coupling effect between beta and alpha and temperature to evaluate**

Q2: Waste mechanical degradation?

Can irradiation induce a cracking of the material?

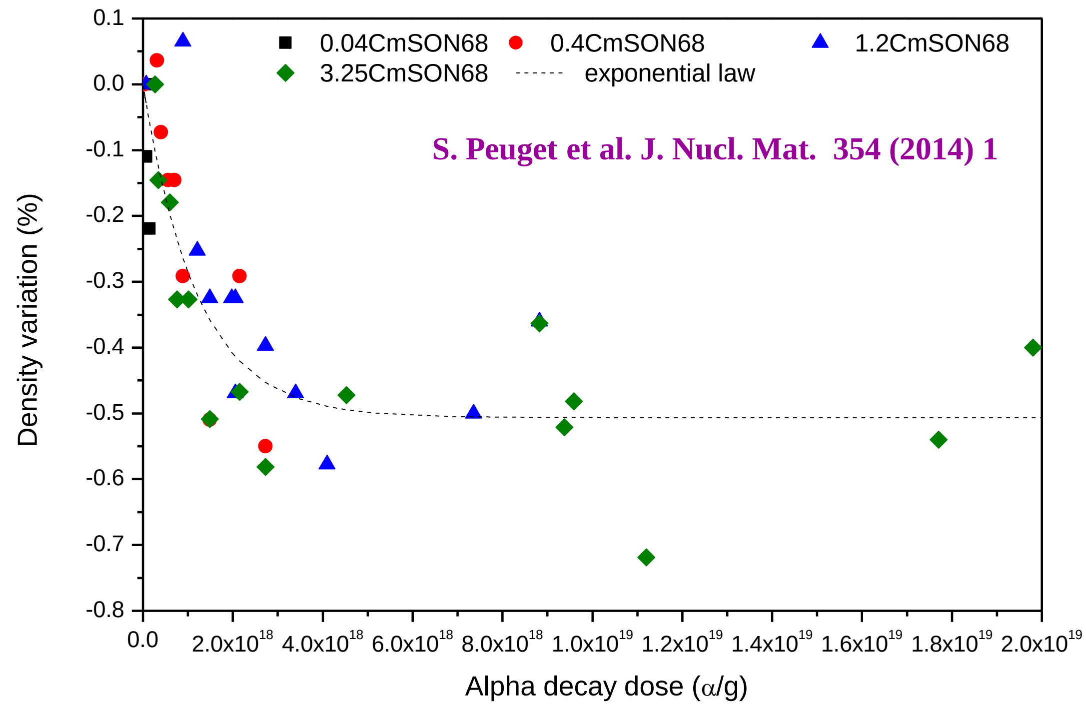
- Due to important swelling under irradiation?
- Due to bubble formation (He bubbles generated by alpha decays)
- Degradation of the mechanical properties?



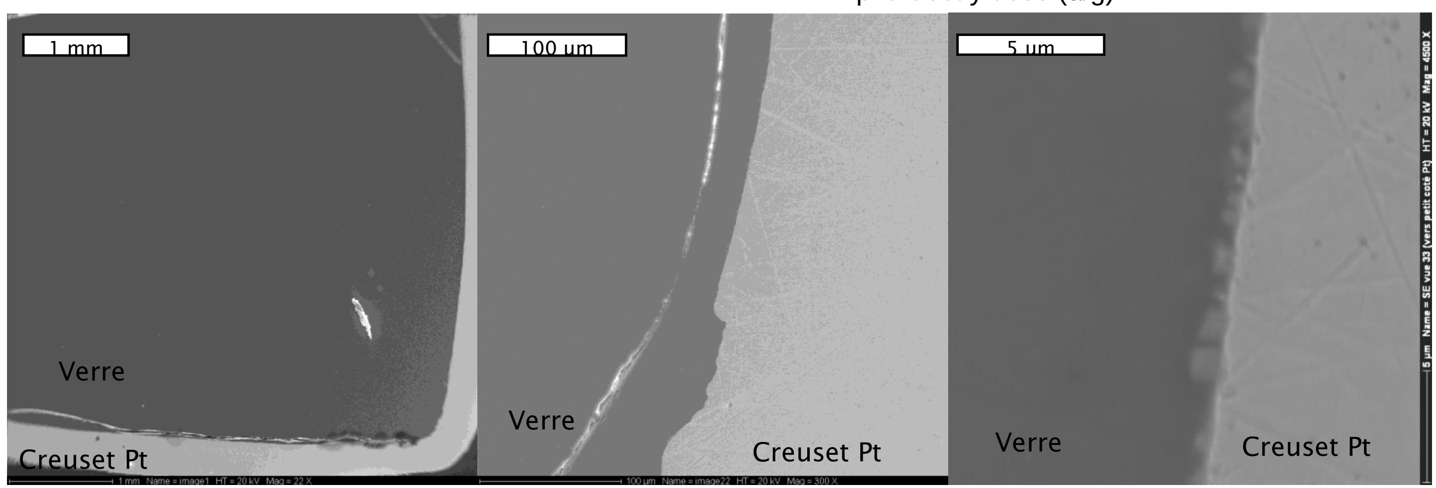
Q2: WASTE MECHANICAL DEGRADATION?

Important swelling of Homogeneous glass?

Slight variation of the glass density



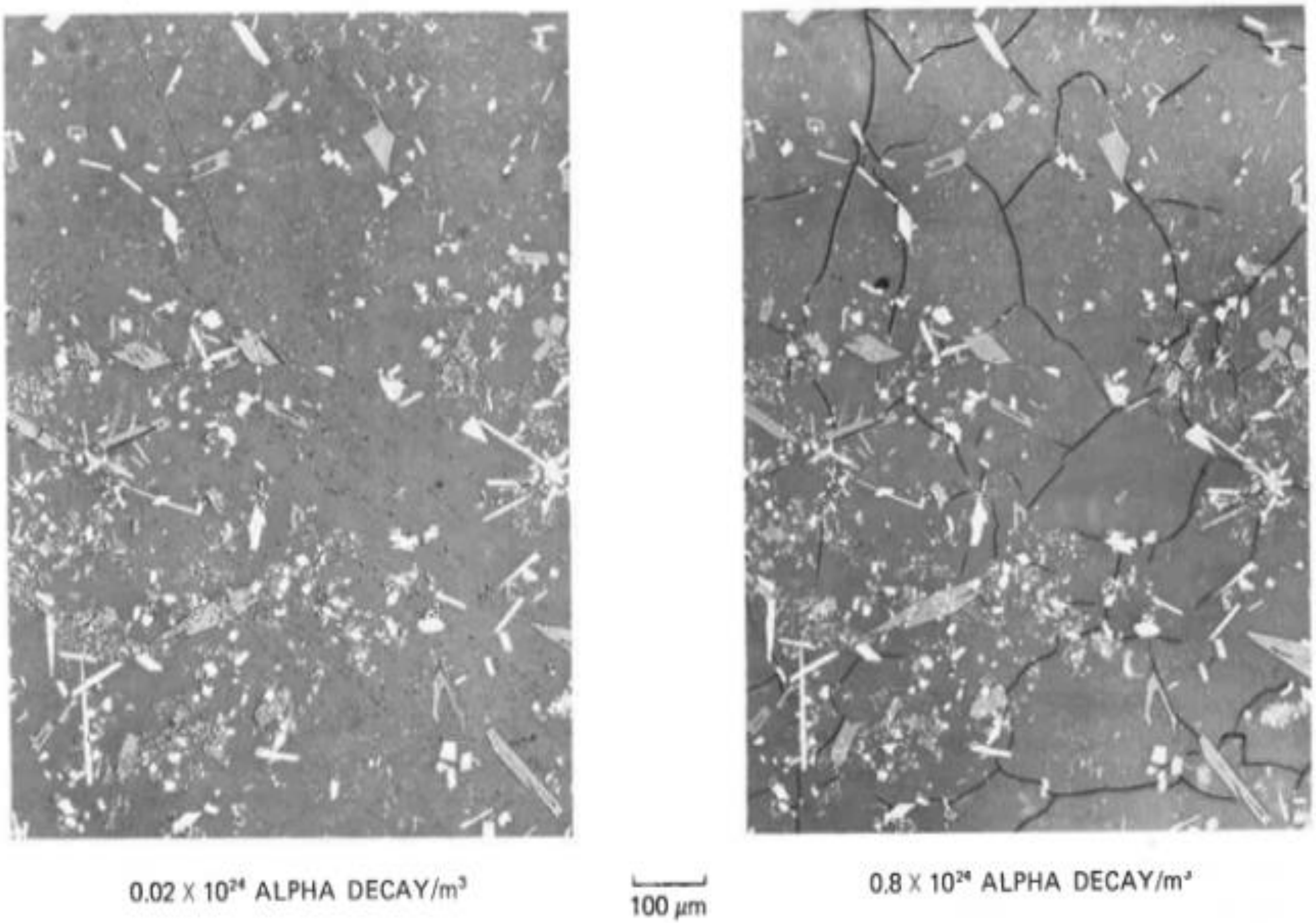
Low swelling level, no microcraking



Q2: WASTE MECHANICAL DEGRADATION?

Waste form degradation ? GCM?

Microcracking observed on some GCM



W. J. Weber and F. P. Roberts, Nuclear Technology, vol. 60.,178-198.

Amorphization of the crystalline phases: high swelling level of crystalline phase

To go further in GCM development:

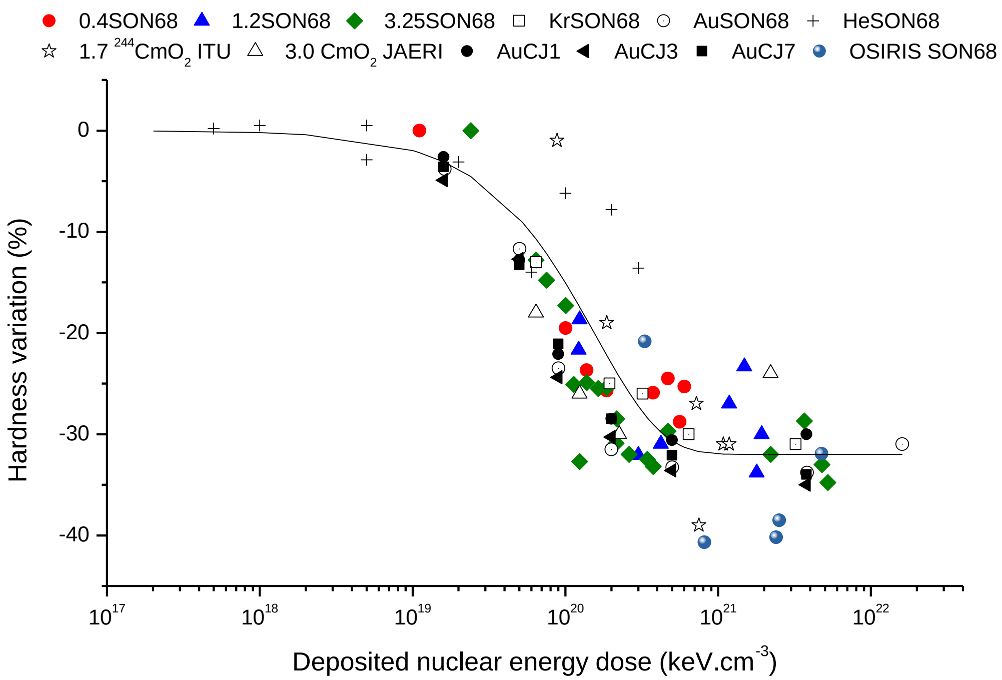
Need to understand and master the origin of radiation induced cracking

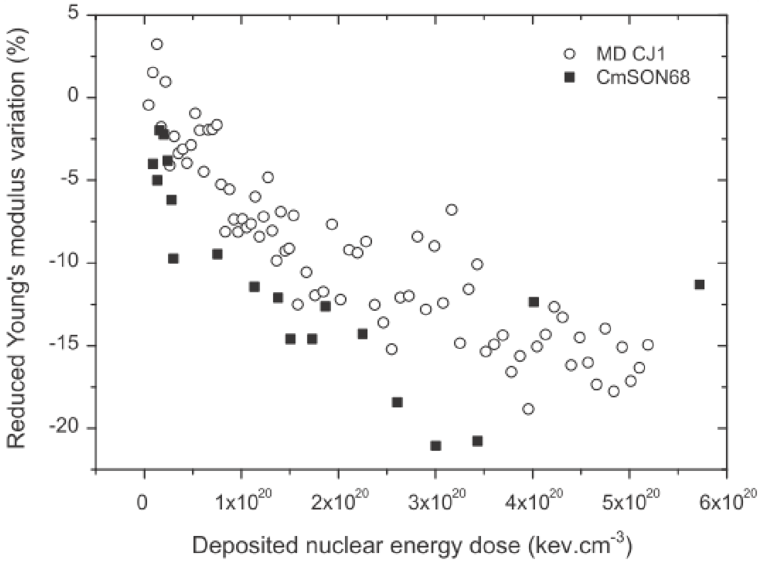
Evaluation of the impact of type of phase, density and size of crystalline phases

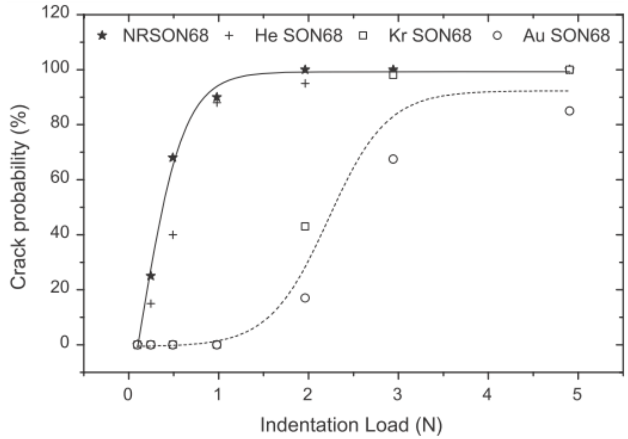
Q2: WASTE MECHANICAL DEGRADATION?

Degradation of the mechanical properties?

Decrease of Hardness, Young Modulus, increase of fracture toughness







No significant degradation of the mechanical properties

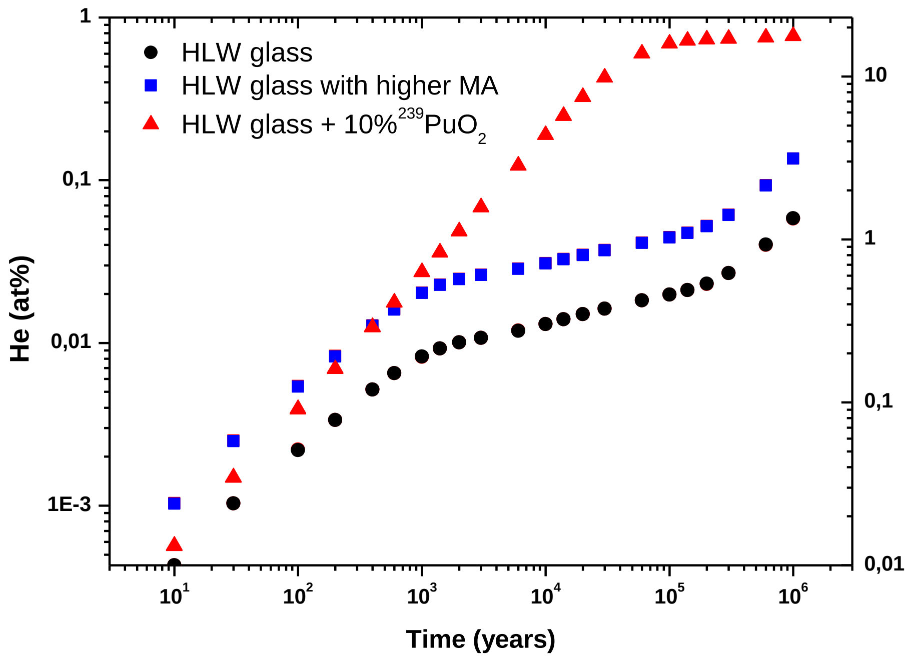
Even slightly better, fracture toughness increase …

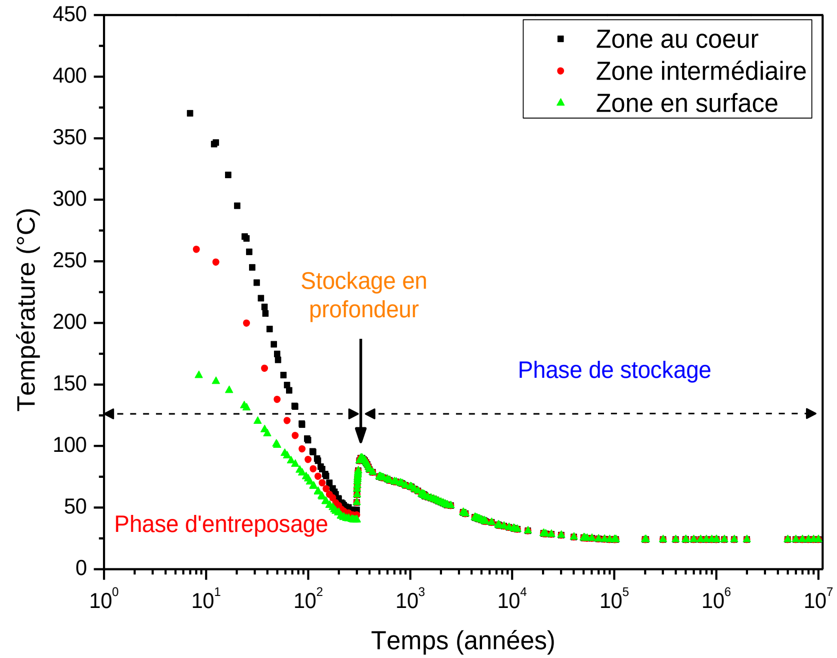
Origin associated to structural changes under irradiation

J.M. Delaye lecture 2

Q2: WASTE MECHANICAL DEGRADATION?

Is there any risk of formation of pressurized He bubbles in a nuclear glass?





- **Helium incorporation mechanism in the glassy network ?**
- **Solubility limit ? Helium bubble formation?**
- **Helium diffusion mechanism?**
- **Impact of radiation damage on these mechanisms?**

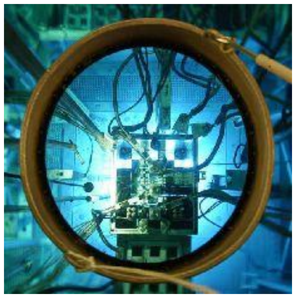
Q2: WASTE MECHANICAL DEGRADATION?

He - METHODOLOGY

Irradiation in OSIRIS reactor ($^{10}B(n,\alpha)^{7}Li$)

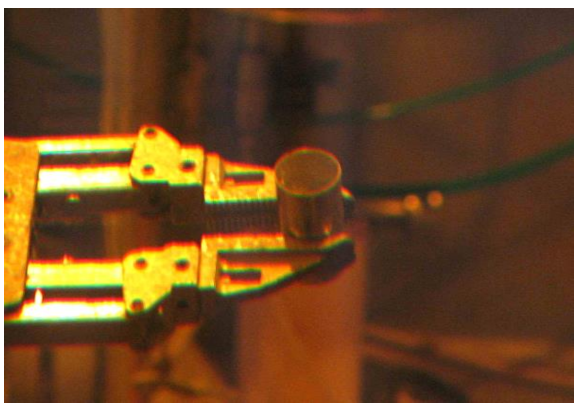
$[He]_{max}$: $2{,}2 \times 10^{20}$ at./cm^3
dpa: ~ 1-2

CEA Saclay
OSIRIS



Cm doped glass (alpha decays)

$[He]_{max}$: $4{,}4 \times 10^{19}$ at./cm^3
dpa: 1



He Infusion (P, T)

Equilibium gas/solid

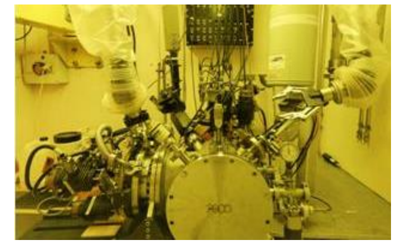
$[He]_{max}$: $3{,}5 \times 10^{18}$ at./cm^3
dpa: 0

CEA Marcoule

$^{3}He^{+}$ implantation :

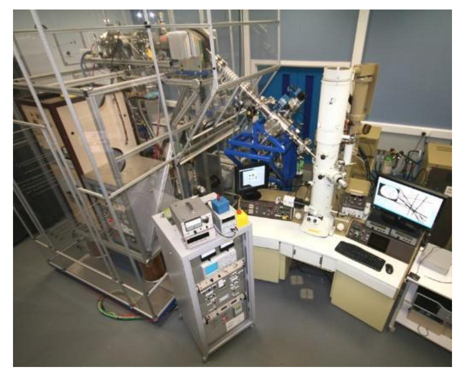
$[He]_{max}$: $4{,}3 \times 10^{21}$ at./cm^3 (local)
dpa: 11 (local)

CEMHTI Orléans, LEEL Saclay
NRA $d(^{3}He,p)\alpha$



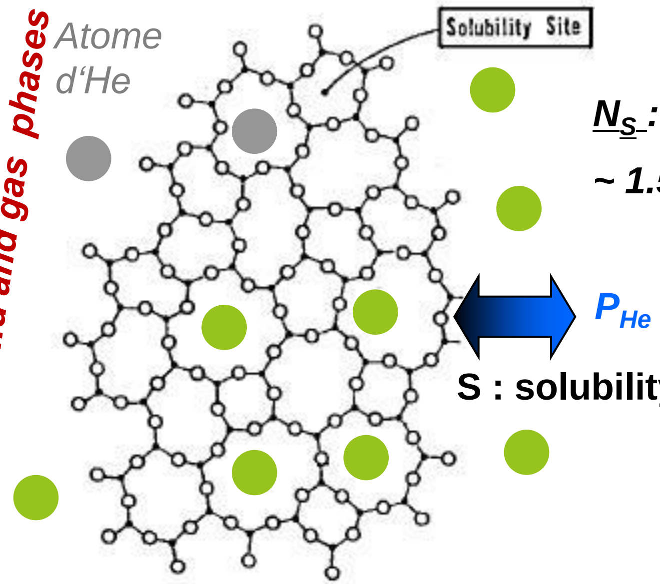
A.H. Mir lecture

Jannus Orsay, MIAMI Huddersfield
in-situ TEM



Q2: WASTE MECHANICAL DEGRADATION? He

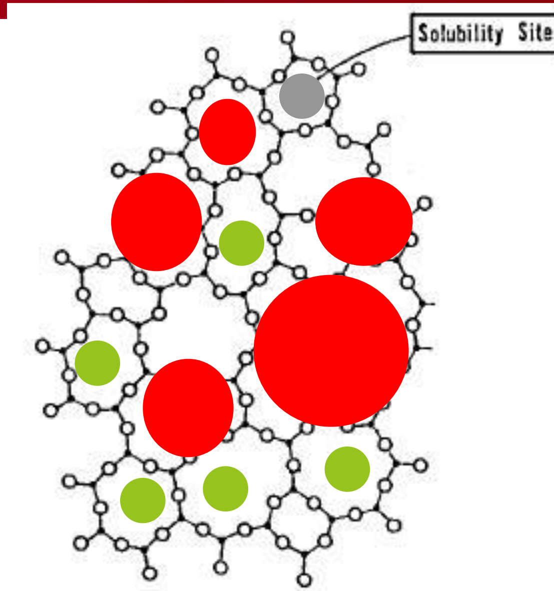
Solubilisation : equilibrium between solid and gas phases



***N*$_S$: density of solubility sites**

~ 1.5 to 3 at%

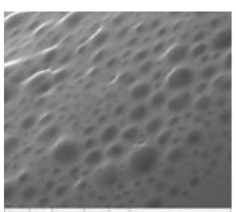
S : solubility constante

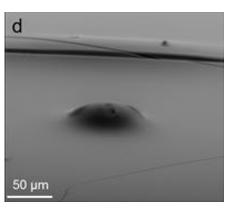


Incorporation (desintegration, implantation) : out of equilibrium

[He] < Ns

- Helium solubilized in the glass free volume
- Bubble formation for T>Tg (~550°C) **only if defects already exist at the glass surface**





1. [He] < 0,1at%
   - He solubilized in the free volume

Commercial glass

2. 0,1at%<[He] < 3 at%
   - He solubilized in the free volume
   - First bubbles …
   - Importance of temperature and radiation?

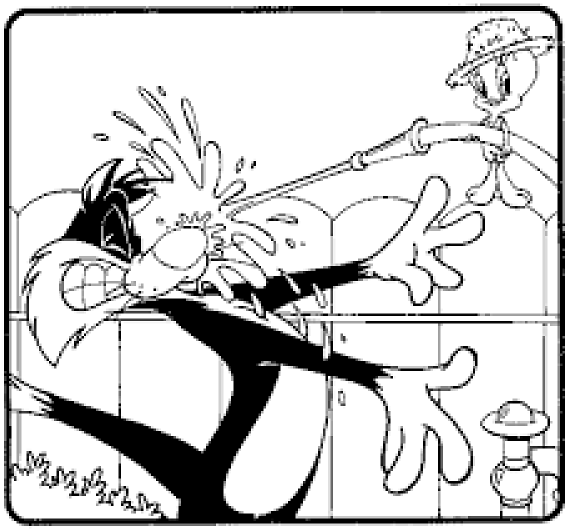
Pu glass

A.H. Mir talk

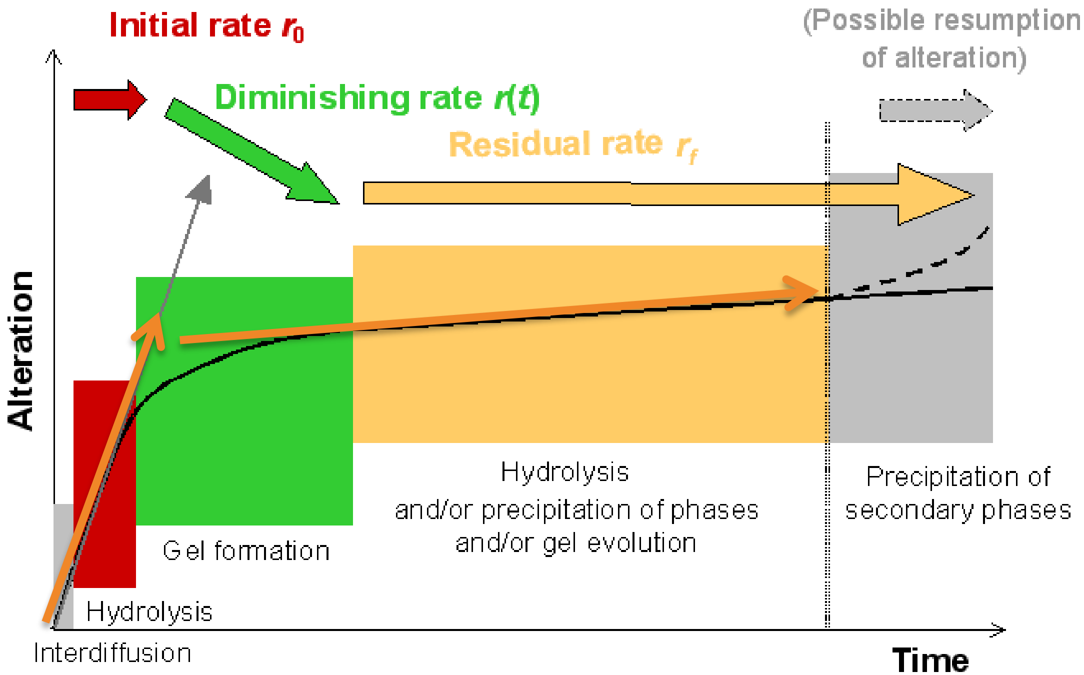
Disposal conditions

3. [He] > Ns
   - Bubble formation at low temperature

Q3: Effect of radiation on the confinement properties?
Leaching behavior? Importance of the surrounding materials?



Q3: EFFECTS OF RADIATION ON THE LEACHING BEHAVIOR?



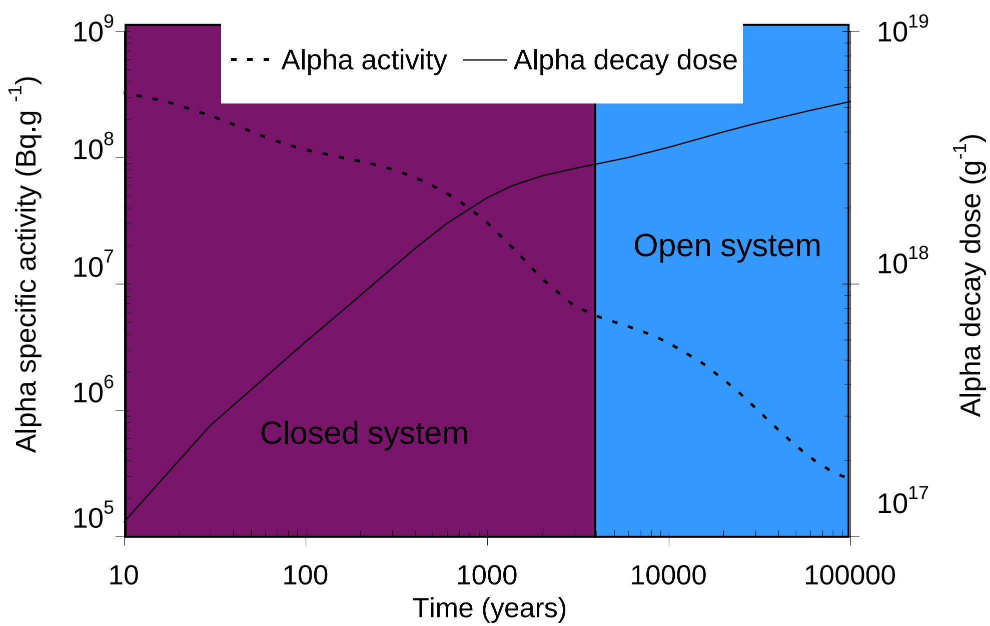
Two main steps to study:

1. **Impact on r_0**
2. **Impact on residual rate, r_r**

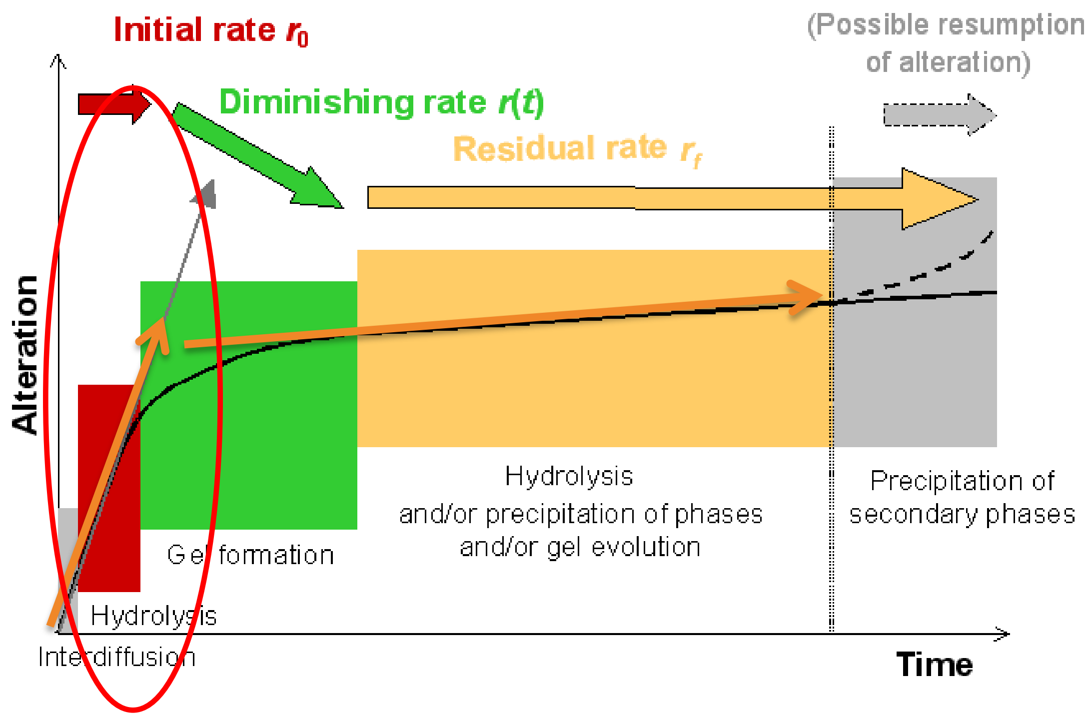
Two parameters to study:

1. **Dose rate**
2. **Dose**

Experiments on radioactive and externally irradiated SON68 glasses



Q3: EFFECTS OF RADIATION ON THE LEACHING BEHAVIOR?



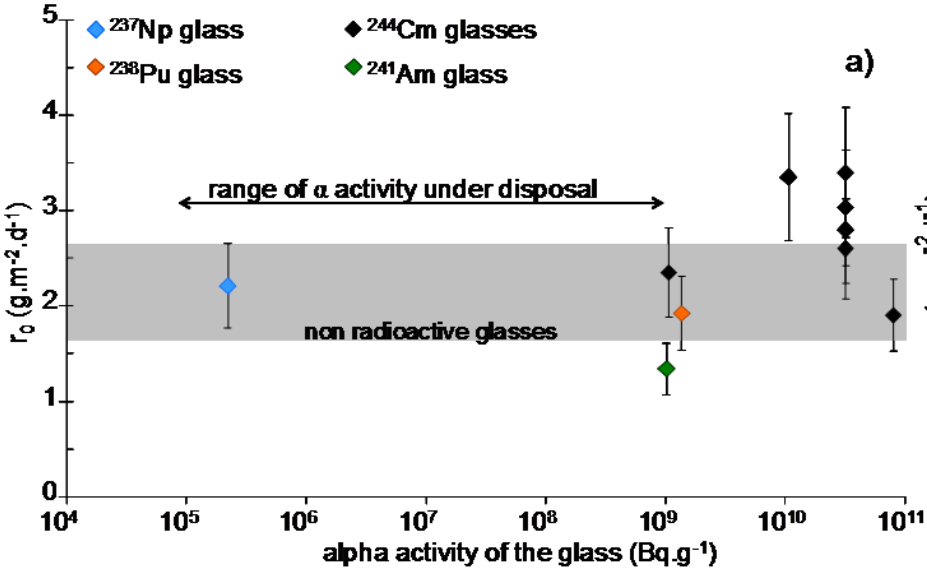
1. Impact on r_0

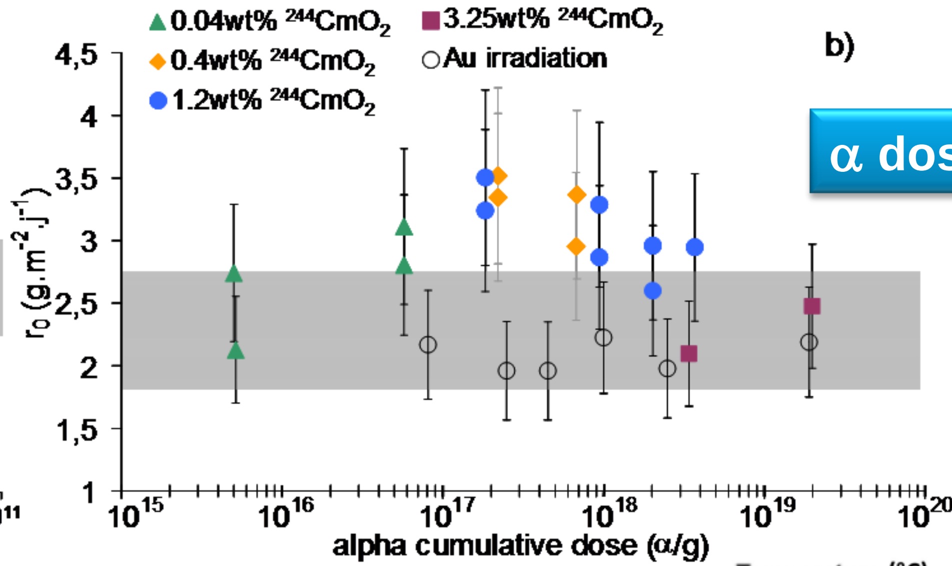
Two parameters to study:

1. **Dose rate**
2. **Dose**

Initial alteration rate, r_0: hydrolysis step

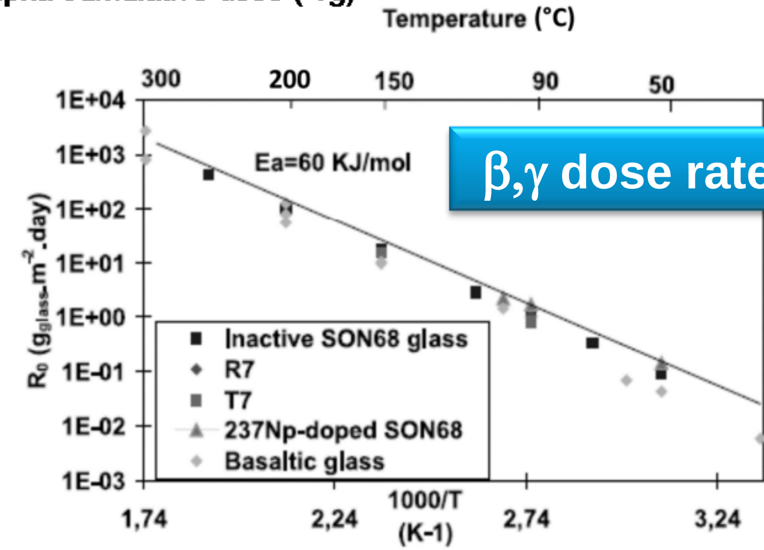
Soxhlet test with chemical analysis of the leachates





α dose

α dose rate



β,γ dose rate

No significant effect of α and β,γ dose rate on r_0

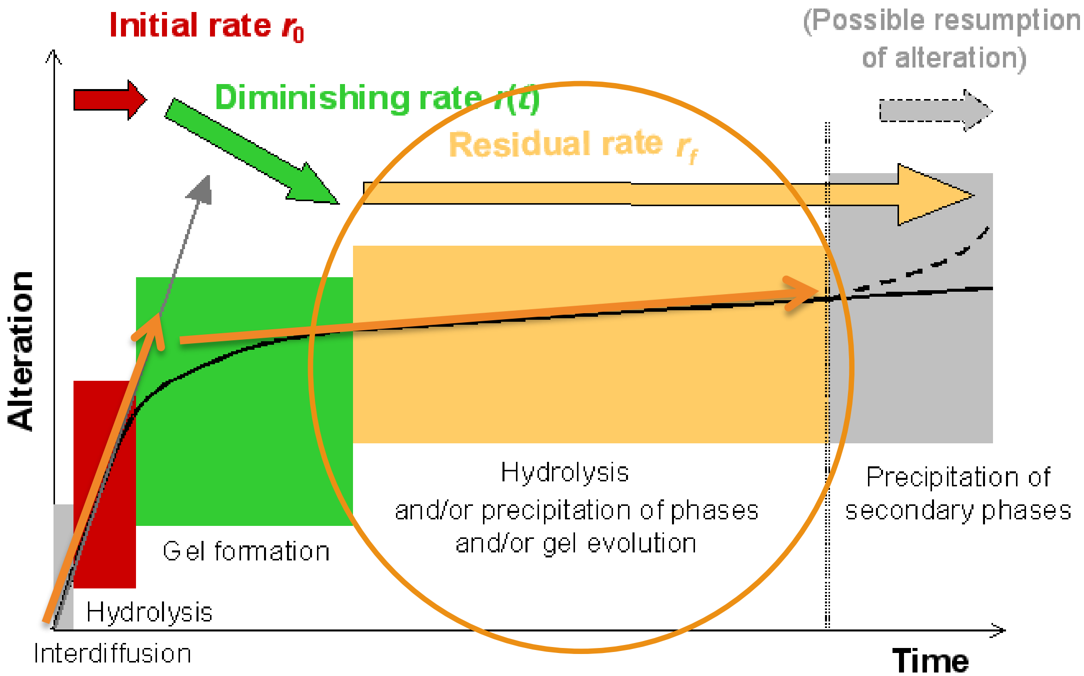
No significant effect of α dose on r_0

No significant effect of β,γ dose (up to 1GGy) on r_0

S. Peuget et al. JNM. 362 (2007) 374

T. Advocat et al. JNM 298 (2001)

Q3: EFFECTS OF RADIATION ON THE LEACHING BEHAVIOR?



2. Impact on residual rate, r_r

Two parameters to study:

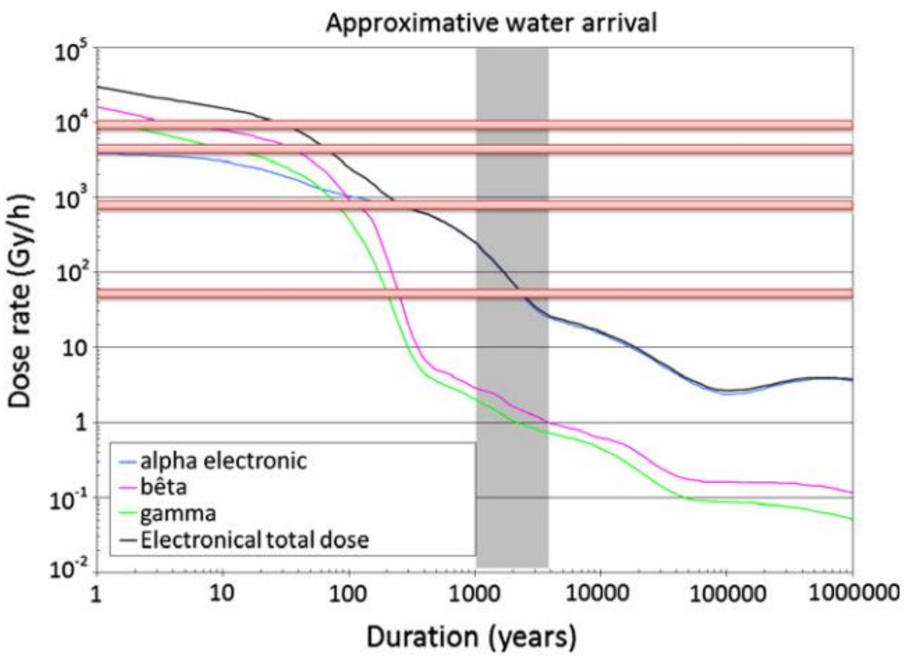
1. **Dose rate**
2. **Dose**

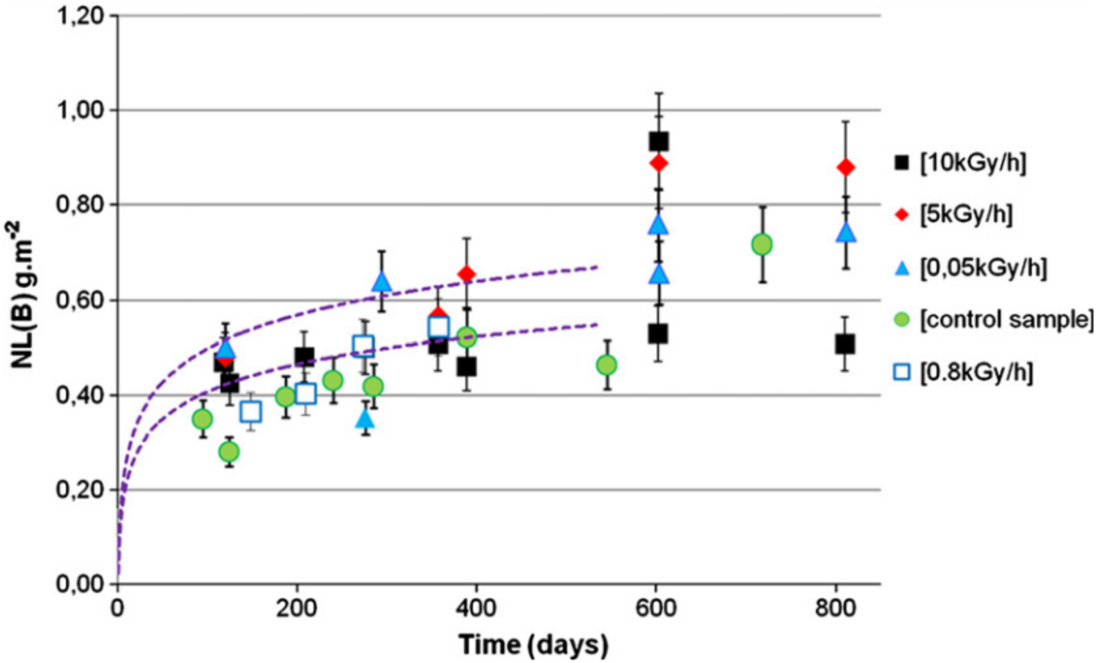
Q3: EFFECTS OF RADIATION ON THE LEACHING BEHAVIOR?

SON68 glasses leached under γ irradiation in pure water

90°C, static leaching test S/V=20cm^{-1}

(Brigitte irradiation facility, SCK-CEN, Belgium)





S. Rolland, M. Tribet, JNM 433 (2013) 382

Similar alteration phenomenology with same r_r as for non-radioactive glass

Similar alteration products (PRI: phylosilicates, porous gel, dense area, pristine glass)

No significant effect of γ dose rate on r_r in pure water

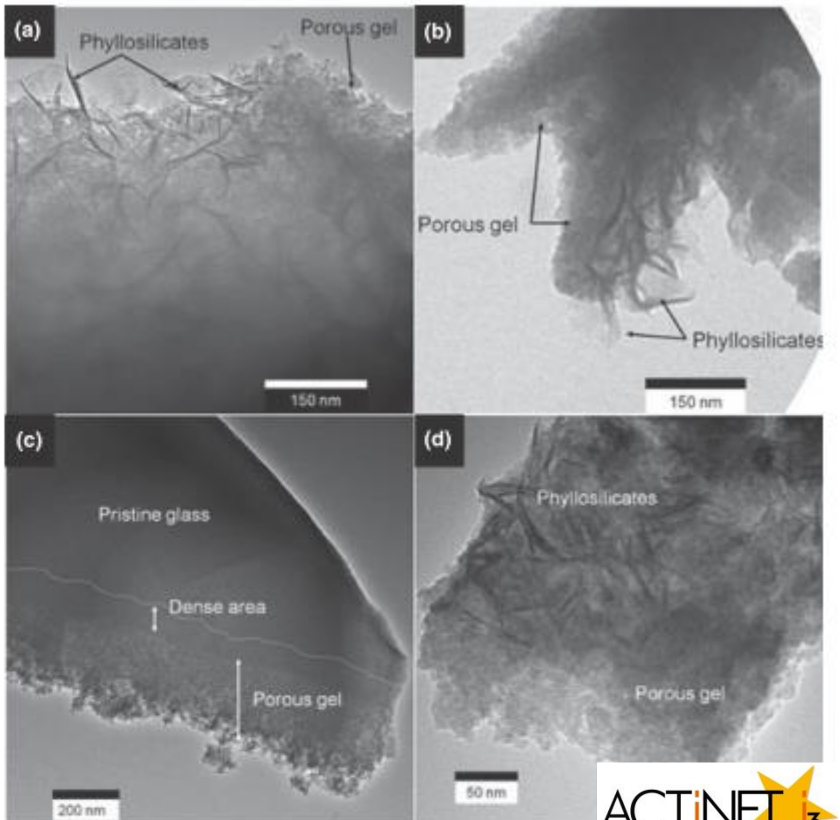
Q3: EFFECTS OF RADIATION ON THE LEACHING BEHAVIOR?

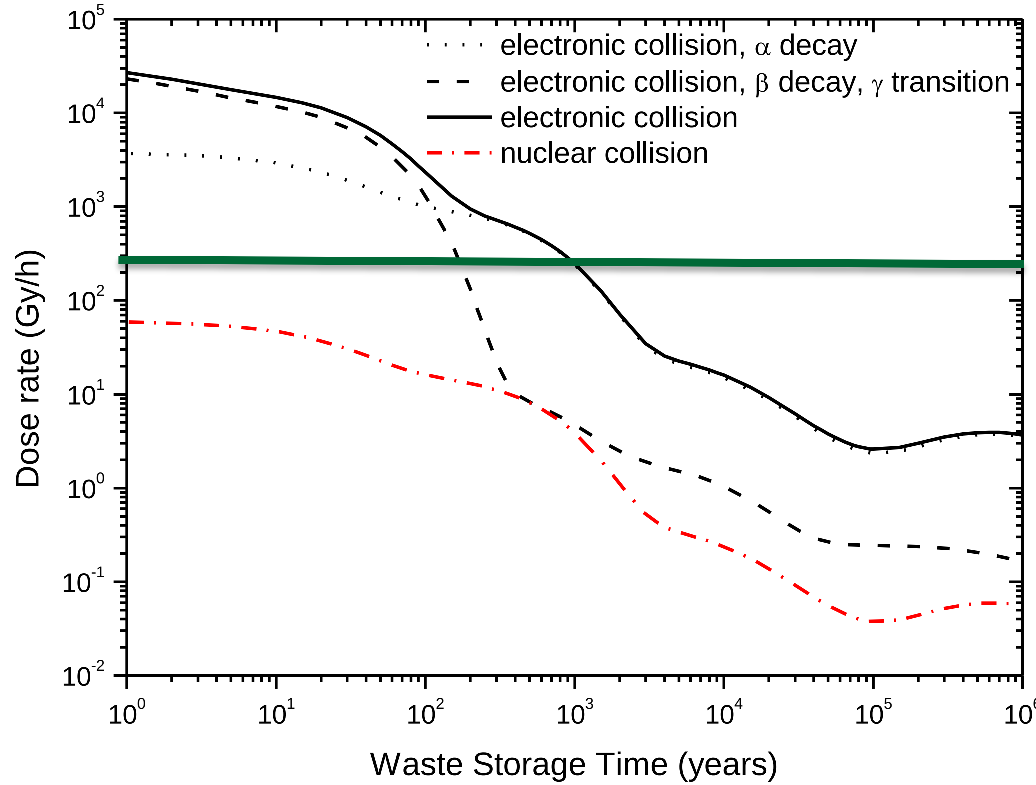
^{239}Pu doped SON68 glass leached in pure water, α dose rate ~ 1000 years of disposal

90°C, static leaching test S/V=20cm^{-1}

TEM CEA

TEM ITU





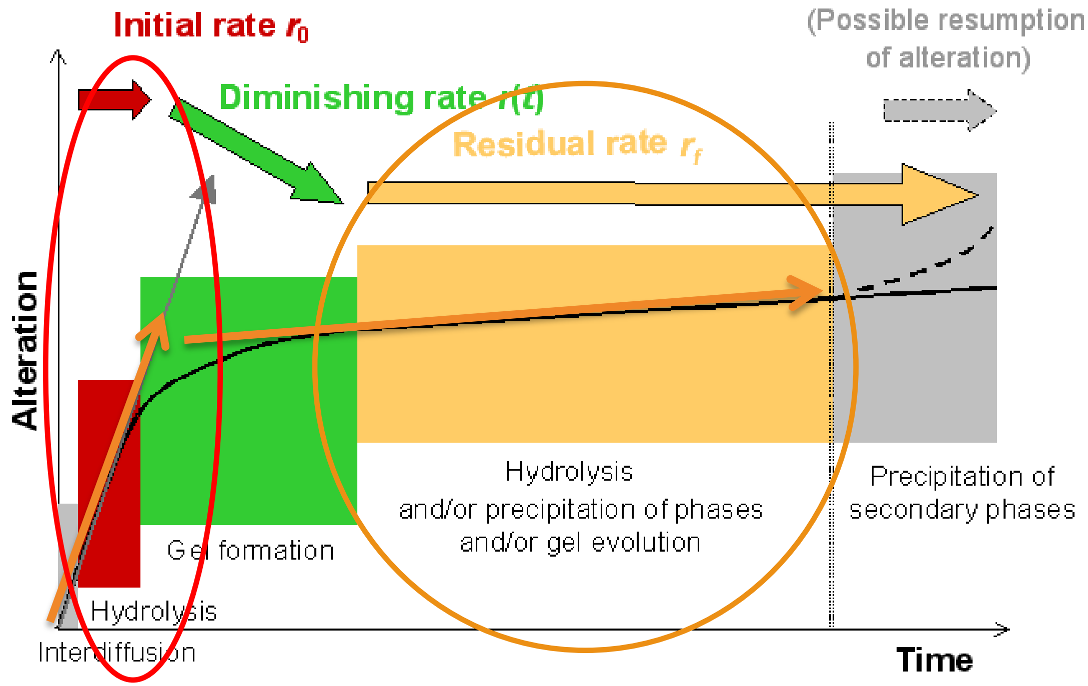
S. Rolland, M. Tribet, Int. J. Appl. Glass Science 4 (2013) 295

Similar alteration phenomenology with same r_r as for non-radioactive glass

Similar alteration products (PRI: phylosilicates, porous gel, dense area, pristine glass)

No significant effect of α dose rate @ 1000 years on r_r

Q3: EFFECTS OF RADIATION ON THE LEACHING BEHAVIOR?



Two main steps to study:

1. **Impact on r_0**
2. **Impact on residual rate, r_r**

Two parameters to study:

1. **Dose rate**
2. **Dose**

No significant effect of $\alpha\beta\gamma$ dose rate on r_0 and r_r (in pure water)

No significant effect of α dose on r_0

Only one study on nucl dose on r_r, M. Tribet talk β,γ dose not studied up to now…

Coupling dose and dose rate on the leaching behavior, to evaluate …

Effect of radiation with the surounding materials, to evaluate …

Conclusion and prospects on Rad. Eff. in nuclear glass

- A lot of work on beta and alpha decays …. but studied separately

- Continous progress in the understanding of radiation effects, beta and alpha decays, He behaviour
  - ✓ Some models are available and need to be to tested
  - ✓ Understanding of glass composition effects is still not so clear

- Complex aging irradiation conditions with mutli-irradiation sources and complex thermal history ⟶ a new step to overcome

- No glass natural analog with fission products or actinides … validation of long term ageing?
  - Characterization of old radioactive glasses (radiation and transmutation)
  - Characterization of alpha doped glasses on longer accumulation time

- Effects of complex irradiation scenario on long term leaching rate to focus on

DE LA RECHERCHE À L'INDUSTRIE

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Thank you for your attention !!!

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