EFFECTS OF RADIATION ON GLASS WASTEFORMS

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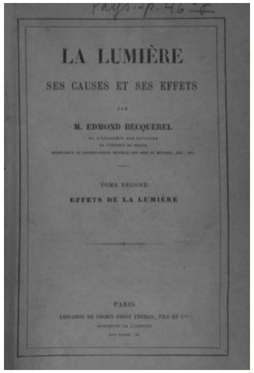
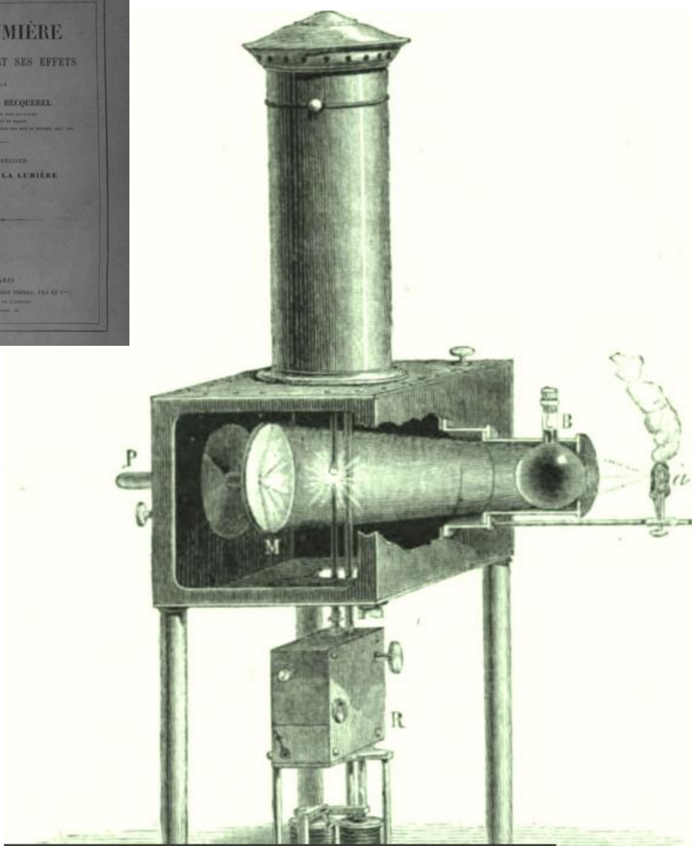
Glass and radiation: an old story

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





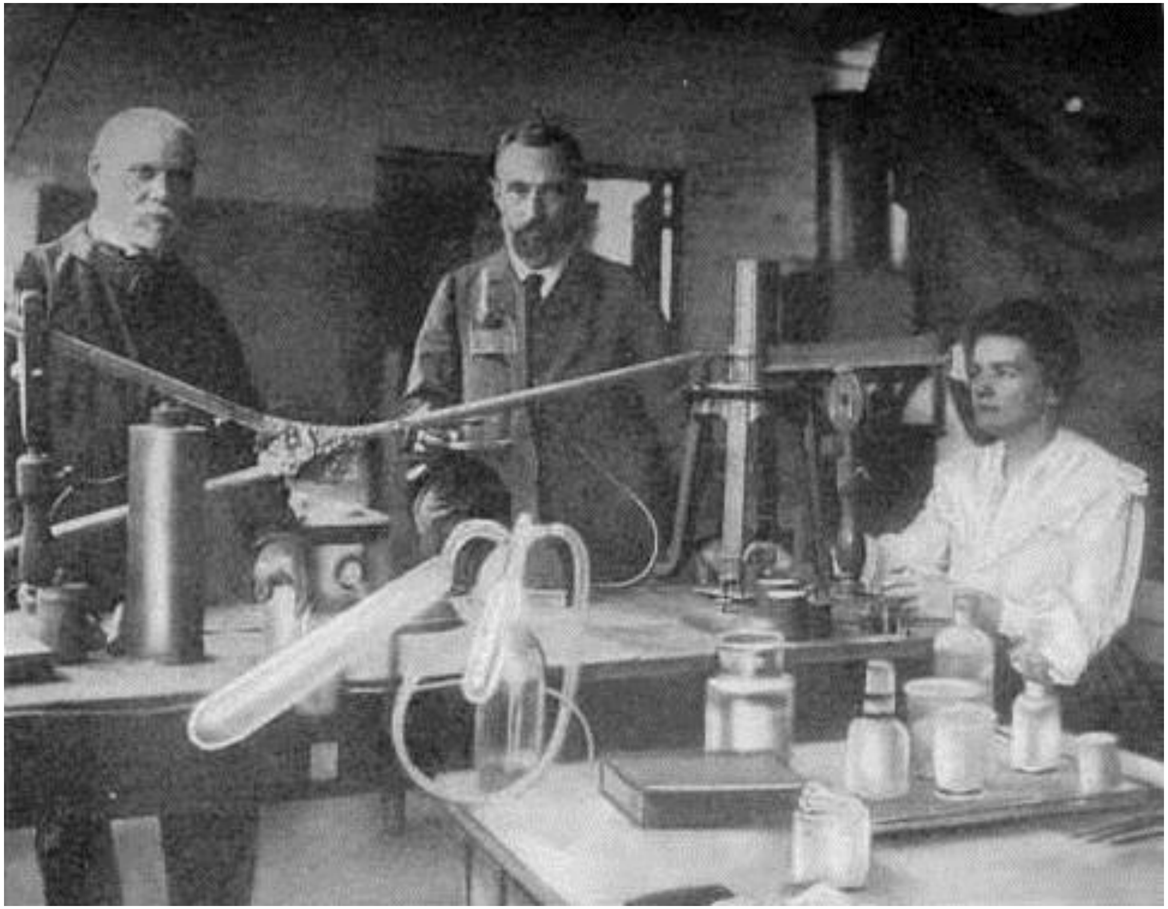
La lumilere, ses causes et ses effets
Alexandre Edmond Becquerel

- 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

PUBLISHED BY THE NATIONAL RESEARCH COUNCIL OF THE NATIONAL ACADEMY OF SCIENCES WASHINGTON, D. C. 1925

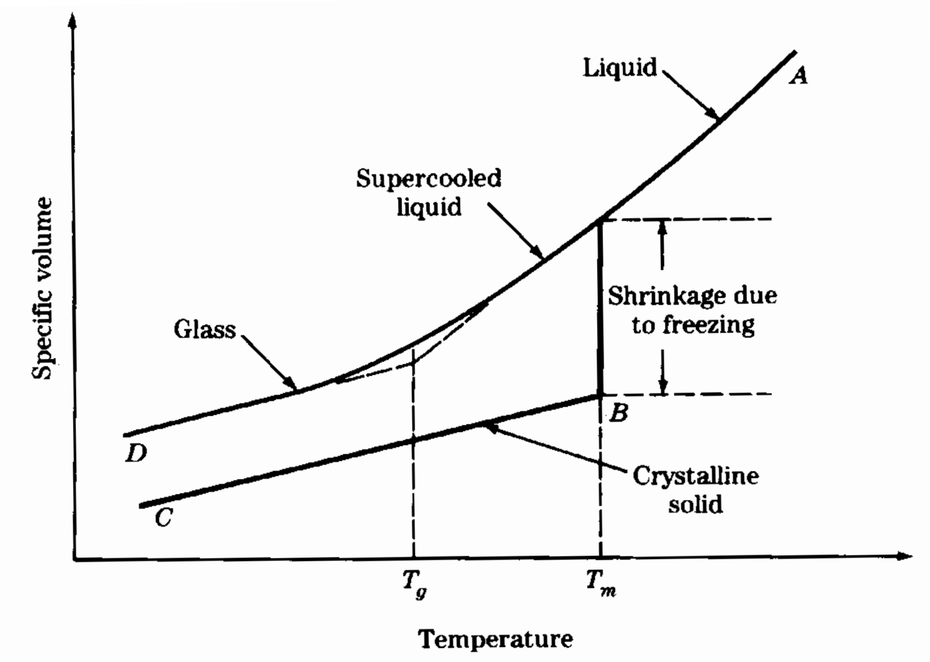
- Doelter, 1910, a review of the coloration of glasses by the action of rays from radium

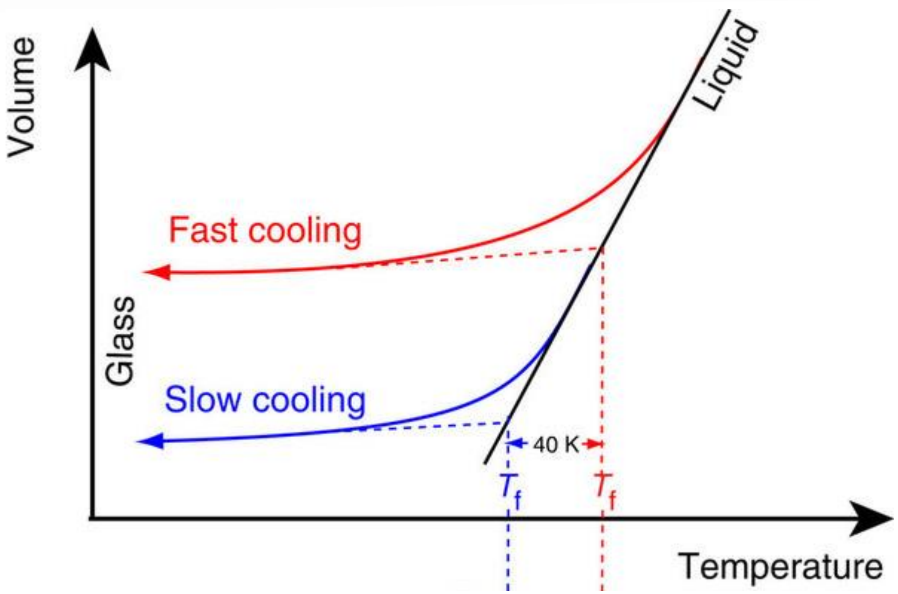
The early work on the coloration of glasses and minerals by the action of the rays from radium is reviewed in Doelter's memoir of 1910. Among

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

Glassy state





A glass (or vitreous solid) is a solid formed by rapid melt quenching.

A glass is an amorphous solid that exhibits a glass transition phenomena at $T_{g.}$

Viscosity

Relaxation time $$\tau = \frac{\eta}{G}$$

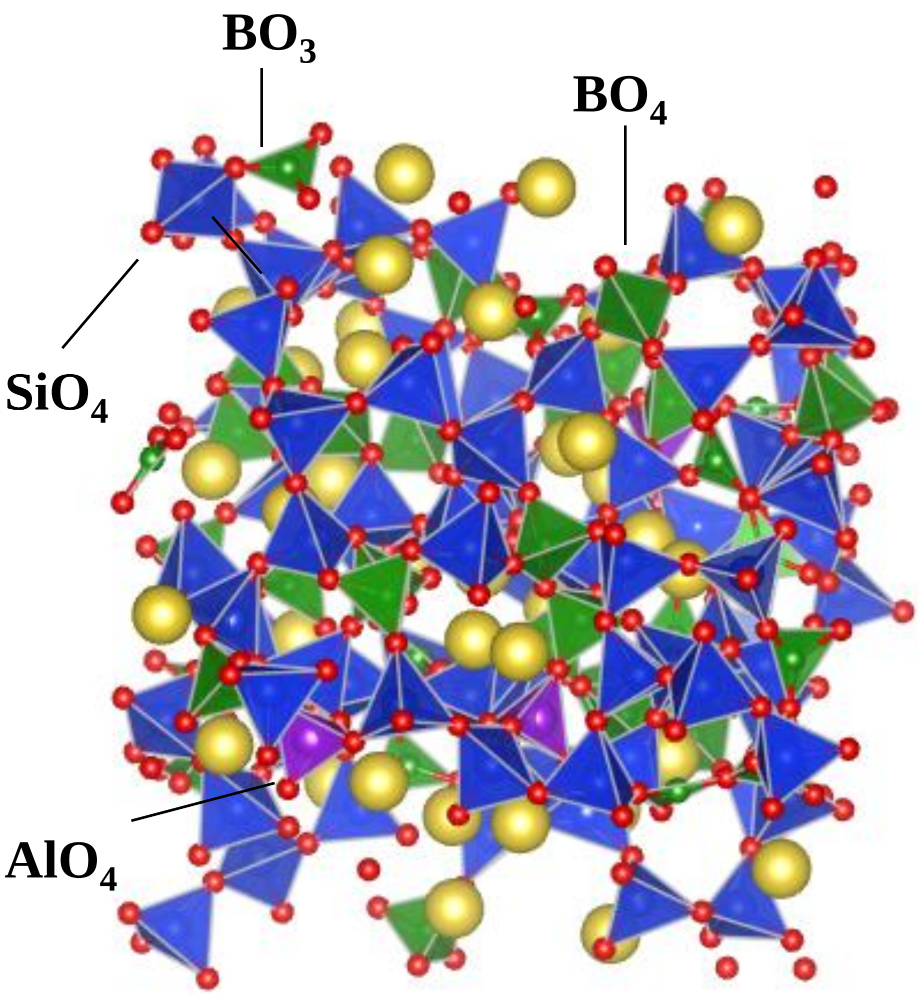
Shear Modulus

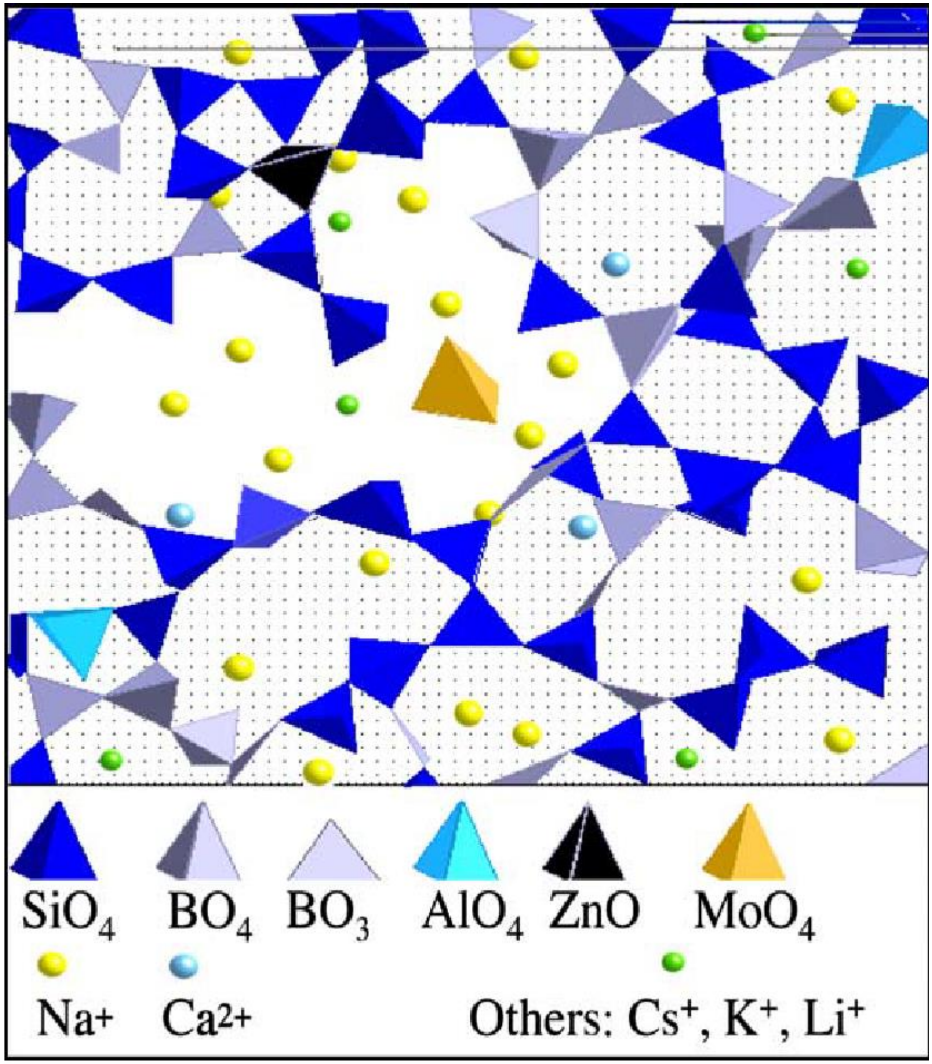
Glass properties depend on:

- **Chemical composition**
- **Thermal history during elaboration process**

Glassy state

- **Short Range Order, SRO : Yes Polyhedra**
- **Medium Range Order, MRO : Yes Angle, Ring statistic**
- **Long Range Order, LRO : No**



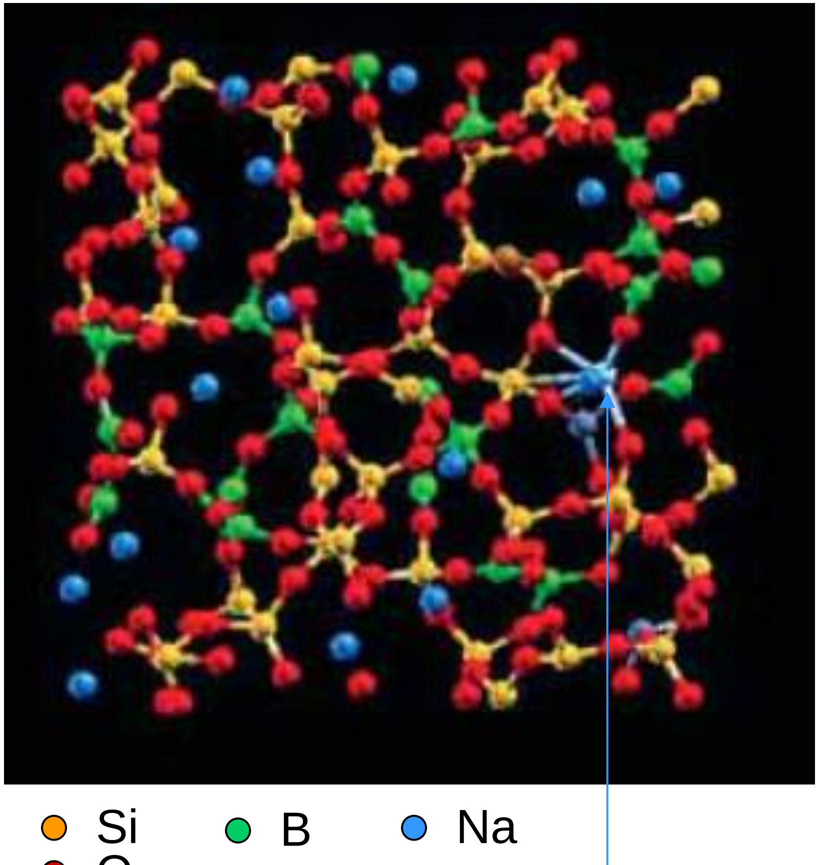


Complex oxides glasses : French Nuclear Glass

Oxide glass with around 30 oxides

Sodium alumino-borosilicate glass

L. Cormier, J.M. Delaye, D. Ghaleb, G. Calas, PRB 61 (2001) 14495

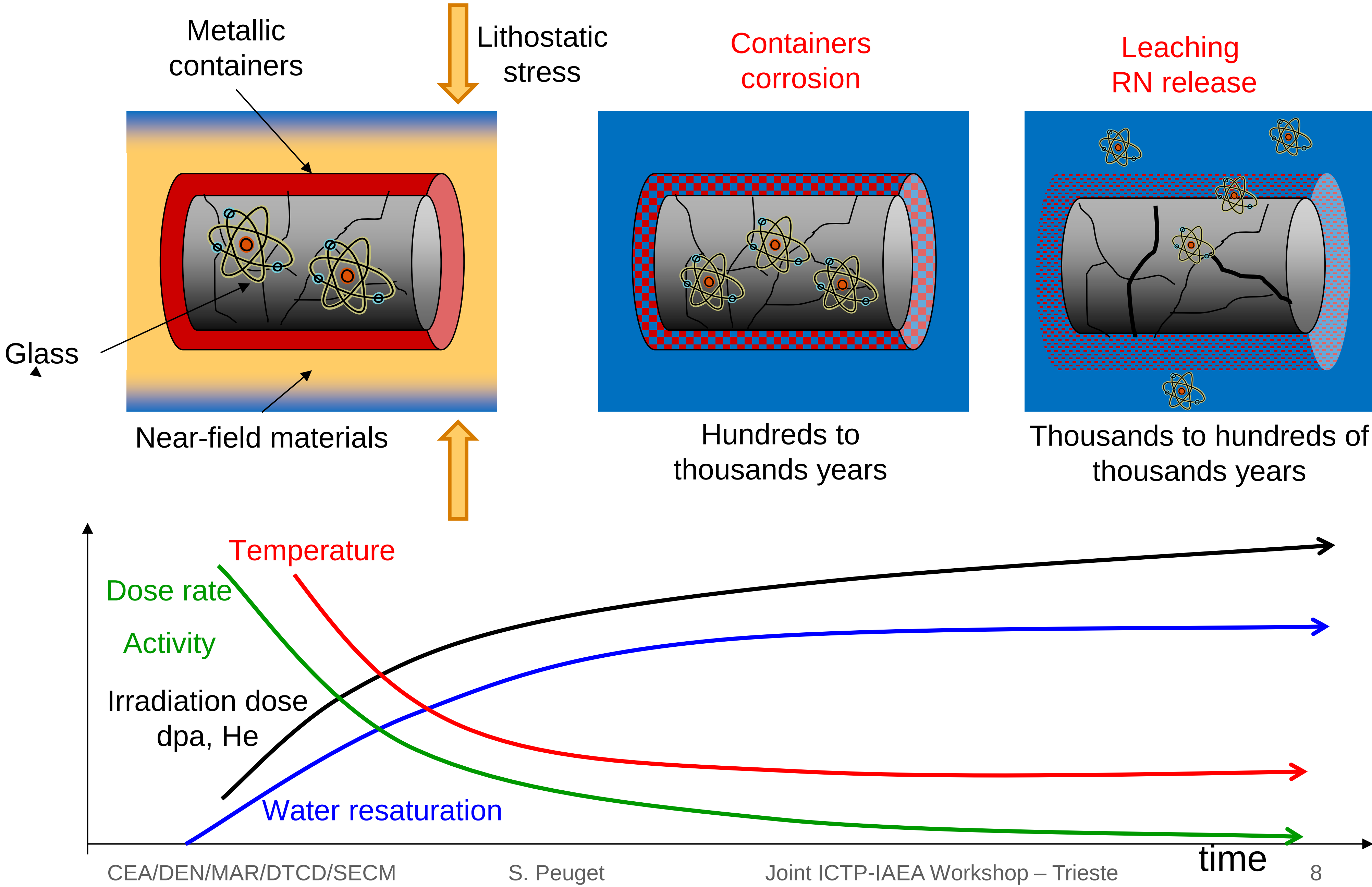


Fission product / Actinide in an octahedric site

Domaine de composition chimique des verres R7T7 produits dans les ateliers industriels par AREVA - La Hague			
Oxydes	Intervalle spécifié pour l'industriel (% massique)		Composition moyenne des verres industriels (% massique)
	min	max	
SiO_2	42,4	51,7	45,6
B_2O_3	12,4	16,5	14,1
Al_2O_3	3,6	6,6	4,7
Na_2O	8,1	11,0	9,9
CaO	3,5	4,8	4,0
Fe_2O_3		< 4,5	1,1
NiO		< 0,5	0,1
Cr_2O_3		< 0,6	0,1
P_2O5		< 1,0	0,2
Li_2O	1,6	2,4	2.0
ZnO	2,2	2,8	2,5
Ox (PF+Zr+ actinides)+ Suspension de fines	7,5	18,5	17,0
Oxydes d'actinides			0,6
SiO_2+B_2O_3+Al_2O_3	> 60		64,4

Monographie DEN : Le conditionnement des déchets nucléaires

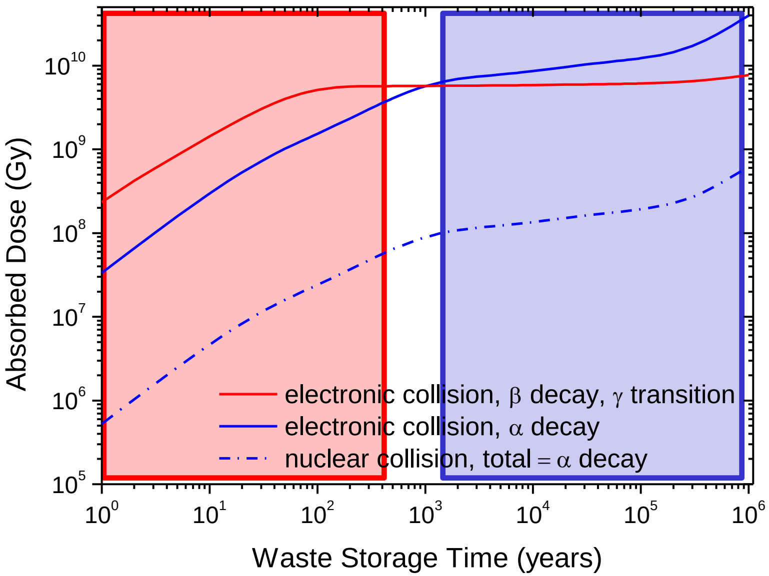
Glass Long Term Behavior



Glass Long Term Behavior: What type of radiation?

Figures for commercial French nuclear glass

Irradiation source	path	Deposited energy (Gy) After 10^4 years	Deposited energy (Gy) After 10^6 years	Number of atomic displacements per event	Number of event per gram of glass after 10^4 years	Number of atomic displacements after 10^6 years
α decay						
He (4 to 6 Mev)	**~ 20 µm**	**~ 3×10^9**	**~ 10^{10}**	**~ 200**	~ 3×10^{18}	~ 6×10^{20}
Recoil Nuclei (0.1Mev)	**~ 30 nm**	~ 6×10^7	~ 3×10^8	**~ 2000**		**~ 6×10^{21}**
β decay	**1mm**	~ 3×10^9	~ 4×10^9	**~1**	7×10^{19}	7×10^{19}
γ transition	**qqs cm**	~ 2×10^9	~ 2×10^9	**<< 1**	2×10^{19}	<<2×10^{19}
(α, n) reactions	1m	~ 2×10^2	~ 9×10^3	200 à 2000	3×10^{12}	6×10^{14} à 6×10^{15}
Spontaneous Fissions	Ffs : 10µm neutron: 1m	~ 2×10^4	~ 4×10^4	10^5 200 à 2000	10^{11} à 10^{12}	10^{16} à 10^{17} 2×10^{13} à 10^{15}



- Stage 1 : mainly β,γ effects, ionization effects
- Stage 2 : mainly α decays effects, electronic and nuclear collisions

Most of the studies have addressed the effects separately !

Outline

1- Effects of beta decays

2- Effects of α decays

3- Effects of α decays on the glass structure

4- Roles of Recoil Nuclei and Alpha Particle?

Outline

1- Effects of beta decays

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Effects of beta decays

Simulation of beta decays: Dose (@1000 years) ~ 5GGy (~ 10^3 Gy/hr)

	Dose Rate Gy/hr	Irradiation time (Equivalent of 1000 years of aging)	Irradiation volume	Remarks
Intense ^{60}Co Gamma Source	3×10^4	~20 years	bulk	Studies in MGy dose range
Electron Accelerators	10^7 to 10^8	Several days to weeks	Up to 1 mm thick	Most of the studies performed
Electron microscopes (SEM & TEM)	10^{11} to 10^{13}	Several minutes	Irradiation Technique	Be careful …

Effects of beta decays

Main effects observed:

- ✓ **Transmutations – Charge imbalance; minor chemistry changes**
- ✓ **Covalent and ionic bond rupture**
- ✓ **Localized electronic excitations, production of electrons and holes**
- ✓ **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)**
- ✓ **Phase separation and/or oxygen bubbles (TEM studies only)**
- ✓ **Only few macrosocpic measurements were performed**
- ✓ **Complex glasses are less modified than simple glasses**

Do these processes may affect pathways for phase separation or evolution of microstructure (bubbles, precipitates, etc.) of nuclear glasses?

Why complex glasses are less modified than simple glasses?

No significant evolution of R7T7 glass properties after irradiation with electrons
[Jacquet-Francillon, Radiochimica Acta 25 (1978), Ollier, J.Non-Cryst. Solids 323 (2003)]

But: Evolution of the structure and properties of simple oxide glasses

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)

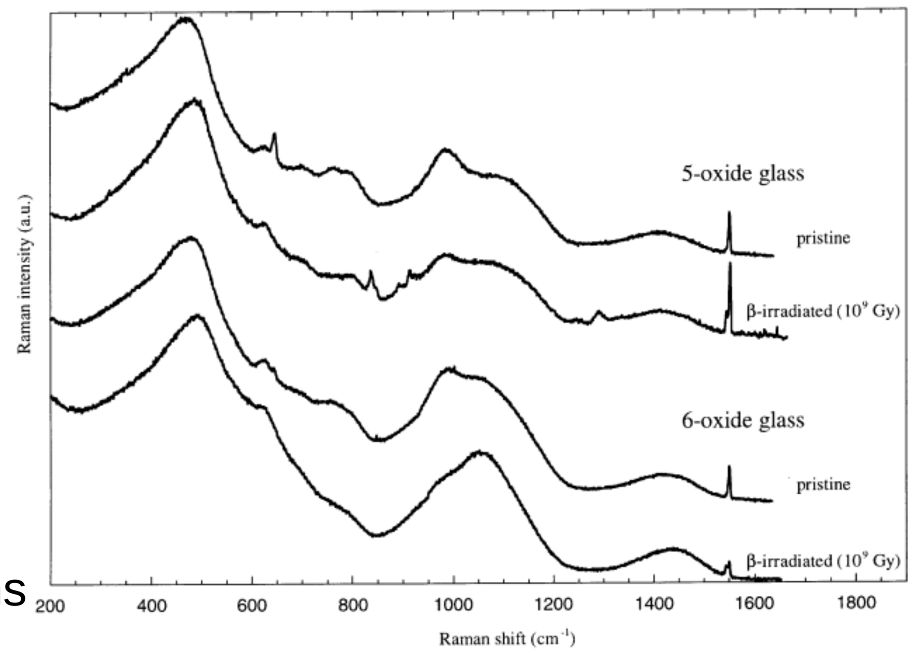
↓

Ponctual defects, alkali migration

Accumulation with dose…

↓

Structural and macroscopic properties evolutions



Effect of alpha decay

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

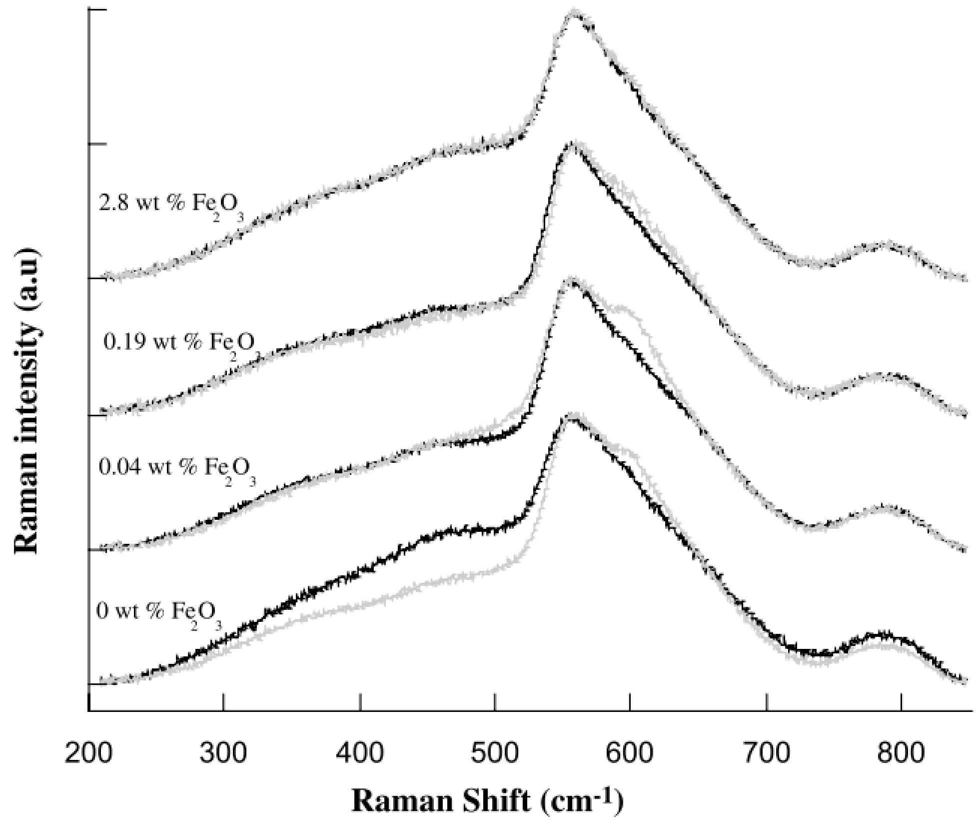
Equilibrium:

$Fe^{3+} + e^-$ → Fe^{2+}

$Fe^{2+} + h^o$ → Fe^{3+}

↓

Decrease or vanishing of structural evolution



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

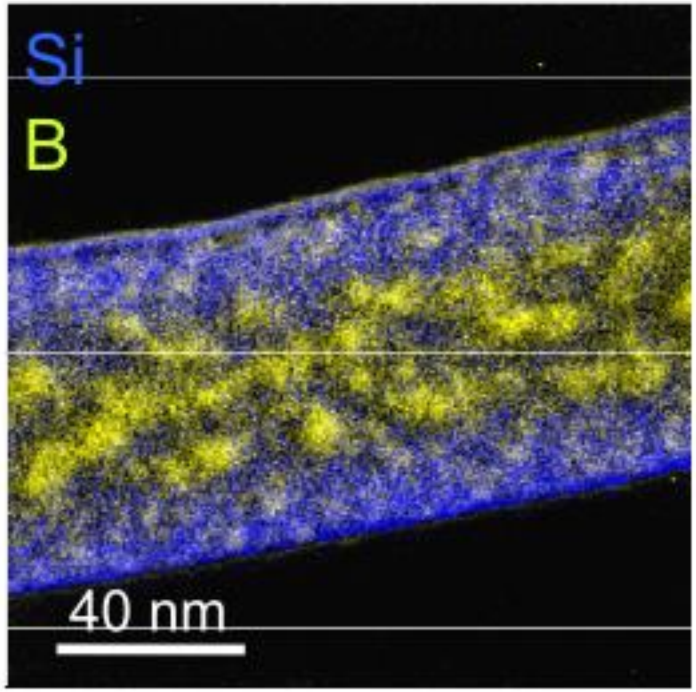
Glass LTB: Effect of beta decay

Phase separation, bubble formation in nuclear glasses?

- **Only observed under TEM studies**

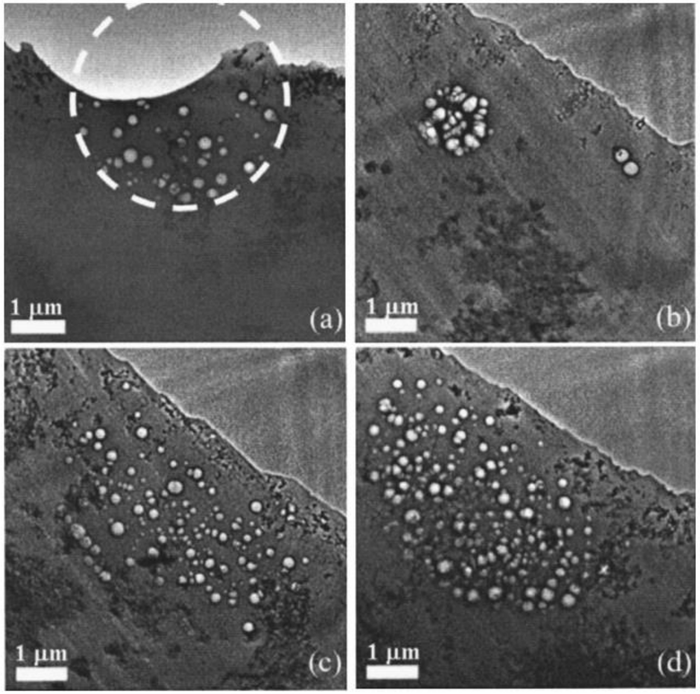
10 orders of magnitude higher dose rate than expected in HLW glass

Na-Borosilicate Glass



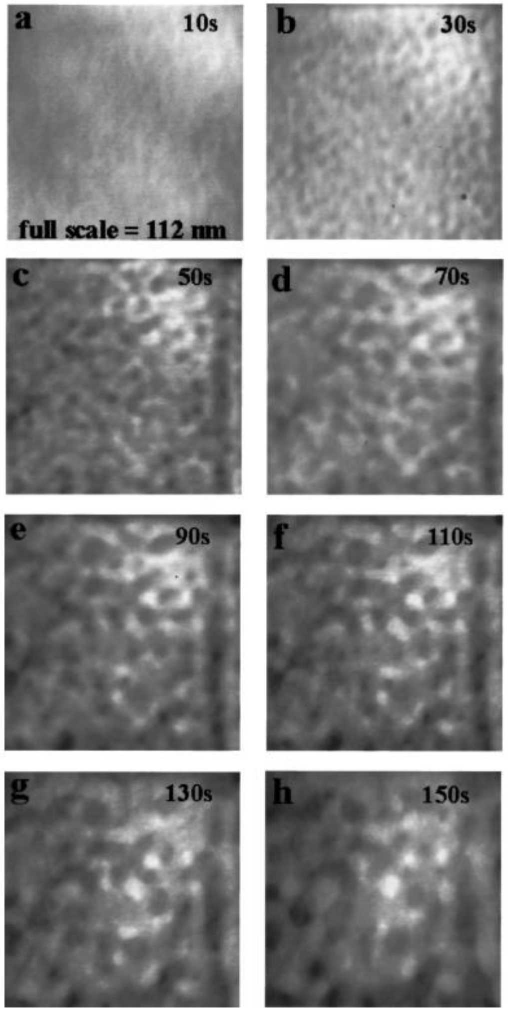
Sun, Microscopy and Analysis 106 (2005)

Li-Borosilicate Glass



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

Ca-Borosilicate Glass

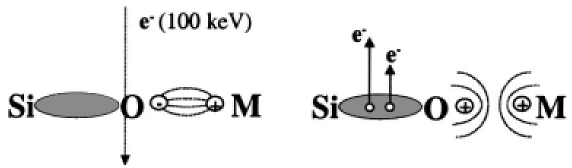


*Jiang, JAP **92** (2002)*

Glass LTB: Effect of beta decay

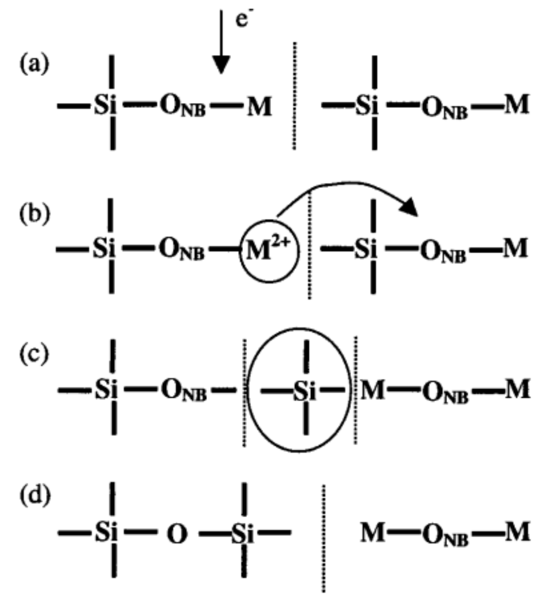
Phase separation, bubble formation in nuclear glasses: several contributions?

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



- **Modification of the glass chemical composition**

(Electron Stimulated Desorption) *Sun, NIMB 218 (2004)*

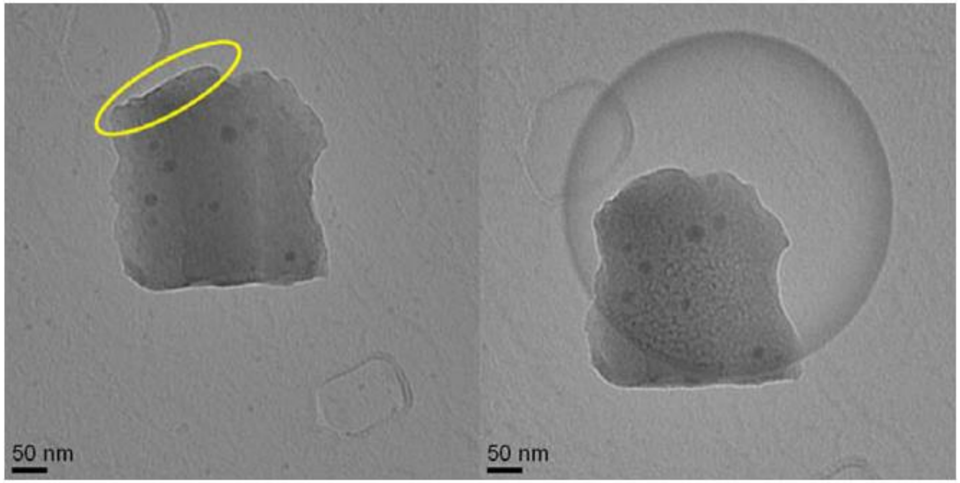


Favors oxygen bubbles and phase separation

Glass LTB: Effect of beta decay

Phase separation, bubble formation in nuclear glasses: several contributions?

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



Favors oxygen bubbles and phase separation

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

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)

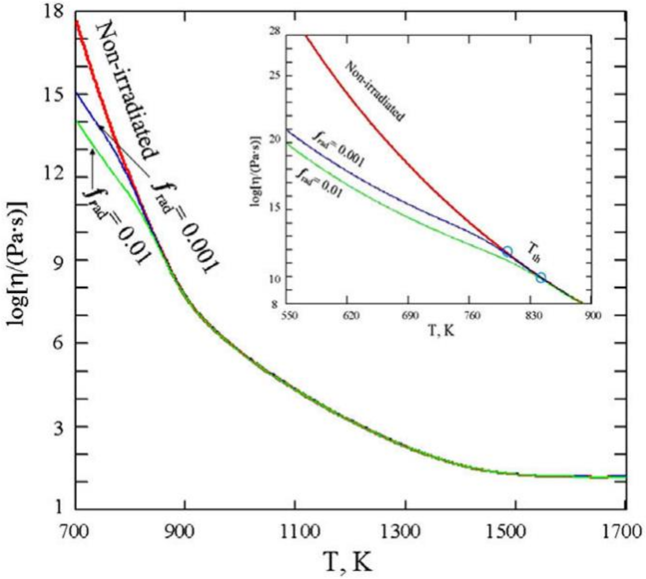


Fig. 9. Viscosities of non-irradiated and electron beam irradiated amorphous soda lime silicate system $70SiO_2 21CaO 9Na_2O$ as a function of temperature for two dimensionless electron flux densities ($f_{rad} = \alpha_e I_e$) 0.01 and 0.001.

Glass LTB: Effect of beta decay

Phase separation, bubble formation in nuclear glasses?

- **Only observed under TEM studies**

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

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

Mir et al, JNCS

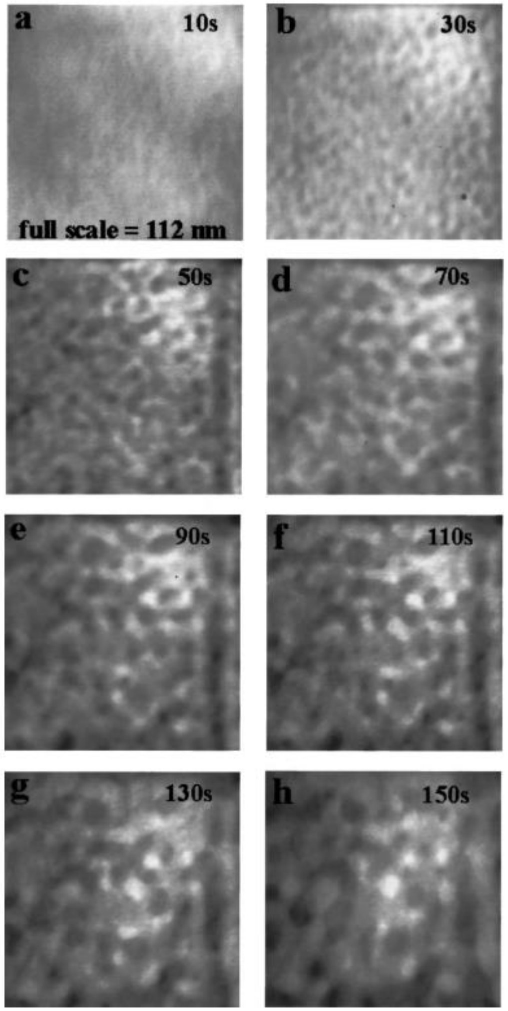
Relevance for long term aging of nuclear glass?

Need to characterize old radioactive glasses

30 years of storage time:

~3GGy ~half of the dose @ 1000years

Ca-Borosilicate Glass



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

Glass LTB: Effect of beta decay

Back

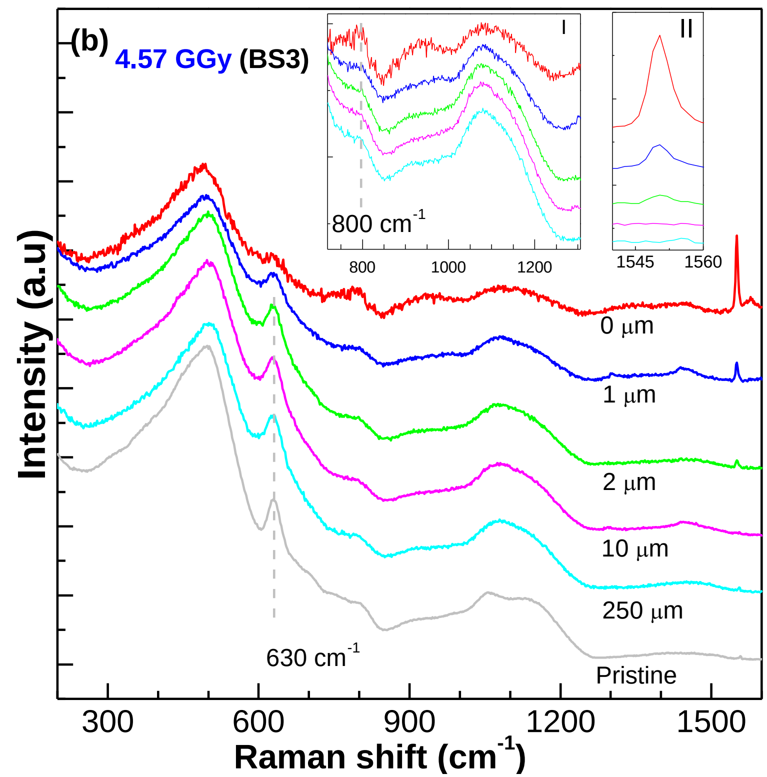
Phase separation, bubble formation in nuclear glasses?

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

Irradiation of a Na, B, Si glass at 4,5GGy

Raman, NMR, TOF-SIMS, AFM

Recent work of Mir, submitted to JNCS



<u>Surface phenomena</u> :

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

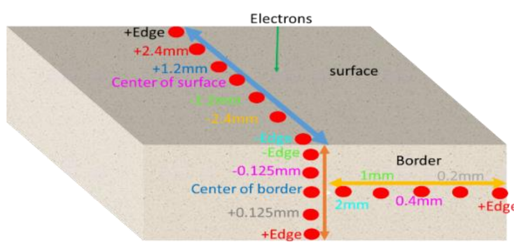
Modification of the glass composition

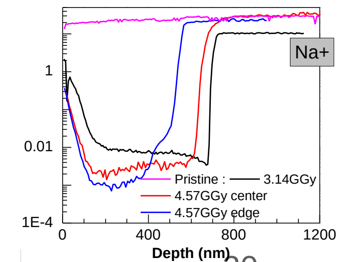
Tendancy of phase separation in the first µm

<u>Bulk</u> :

No phase separation in the bulk,

Slight depolymerization of the network (decrease of boron coordination and increase of NBO around Si and B atoms)





Glass LTB: Effect of beta decay

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Mir et al, JNCS

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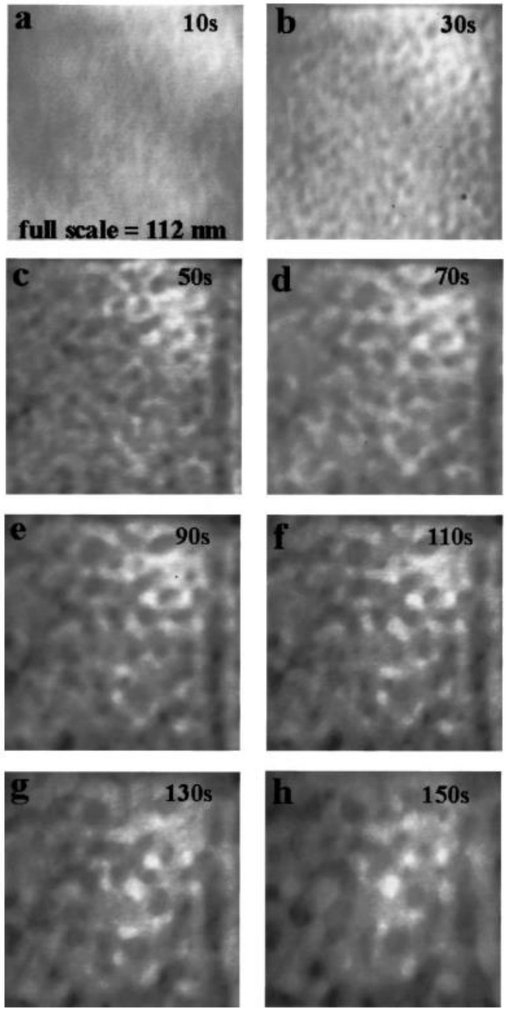
Ca-Borosilicate Glass



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

Outline

1- Effects of beta decays

2- Effects of α decays

3- Effects of α decays on the glass structure

4- Roles of Recoil Nuclei and Alpha Particle?

Glass Long Term Behavior: main past studies?

Main laboratory studies of alpha decay impact

USA (NLs)	70's-90's	<3 x 10^{18} α/g
UK (AERE)	70's-80's	<3 x 10^{18} α/g
France (CEA)	70's- 80's	<3 x 10^{18} α/g
EU (ITU)	70's-90's	<5 x 10^{18} α/g
JAPAN (JAERI)	90's	<10^{19} α/g

Macrosocpic behavior in a limited level of dose

but no data on the glass structure !

Need to improve the understanding of alpha decays effects

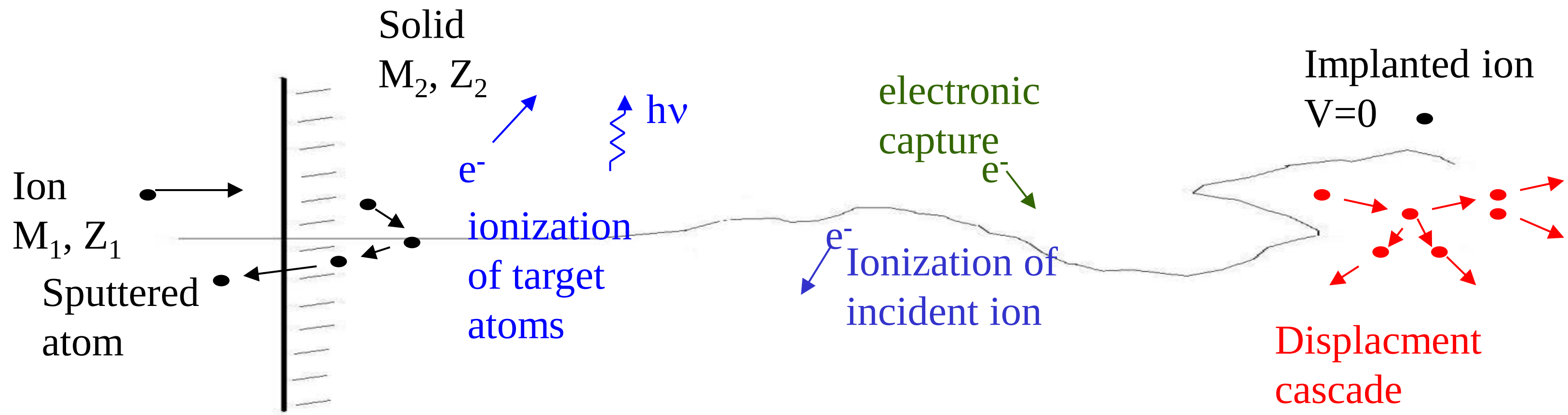
To predict long term behavior

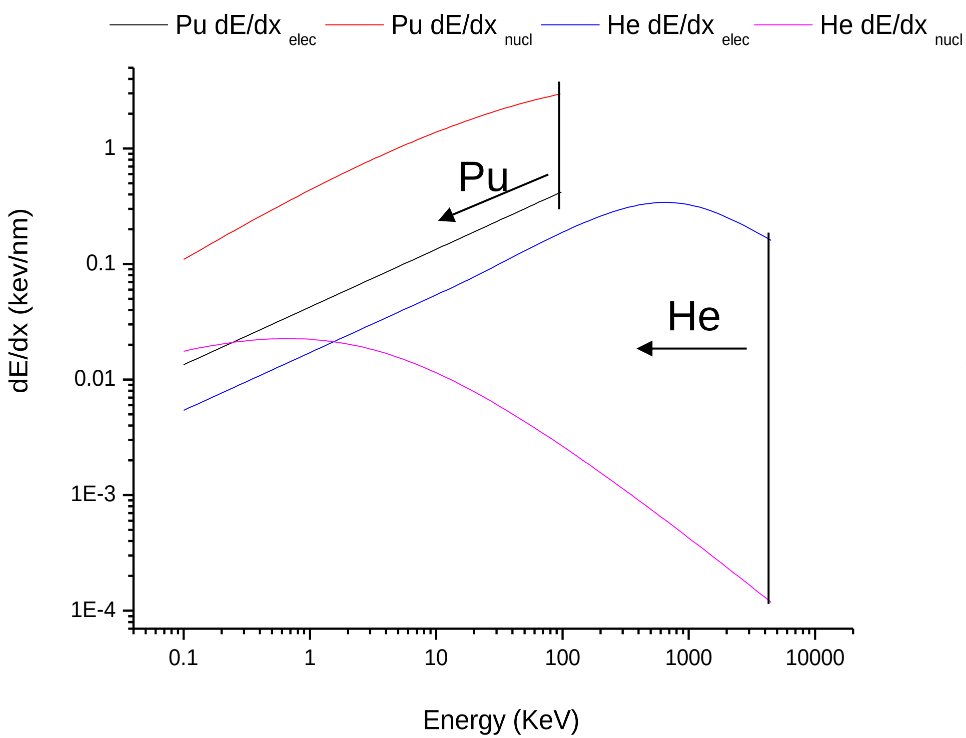
To explore nuclear glass limits

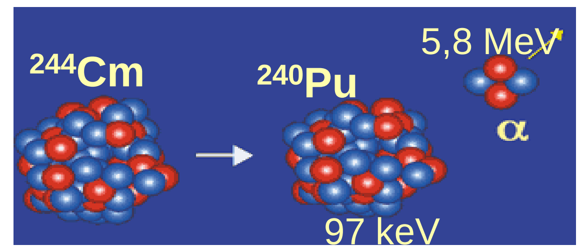
To optimize the future glass or glass ceramics composition

Focus on the results of the research program started in 2001 at CEA

1- Methodology to simulate alpha decays effects







Recoil nuclei (~100 keV) 30-40nm

Mainly nuclear collisions
Ballistic damage
Displacement cascade

α (4-5 MeV) 20-30 μm

Mainly electronic collisions

1- Methodology to simulate alpha decays effects

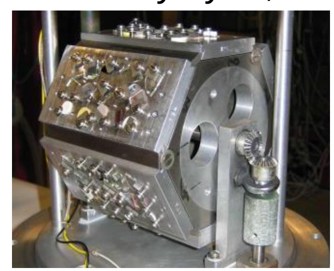
- **Accelerate the time scale**
- **Dissociate the effects of self-irradiation (*electronic / nuclear*) and *helium generation***
- **Evaluate the effects on the confinement properties**
- **Evaluate the effects on the glass structure**

Propose some models to explain the glass behavior under alpha self-irradiation

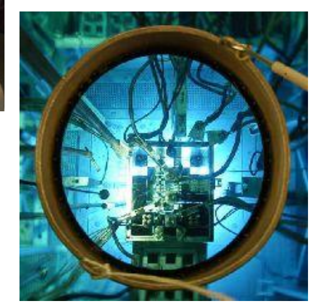
1. **Curium doped glasses**
2. **External irradiation with light and heavy ions**
3. **In pile irradiation : $^{10}B(n,\alpha)^{7}Li$**
4. **Molecular dynamic modeling of ballistic effects**

Atalante DHA, CEA

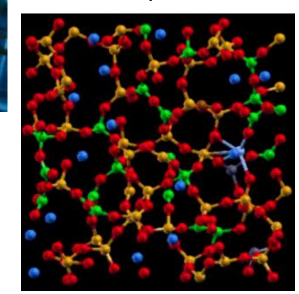
IPN Orsay Lyon, Ganil



OSIRIS, CEA

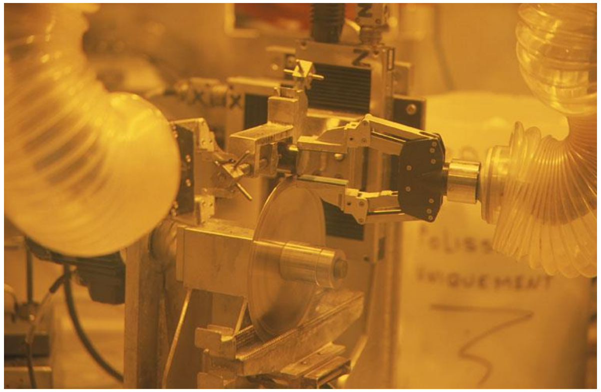


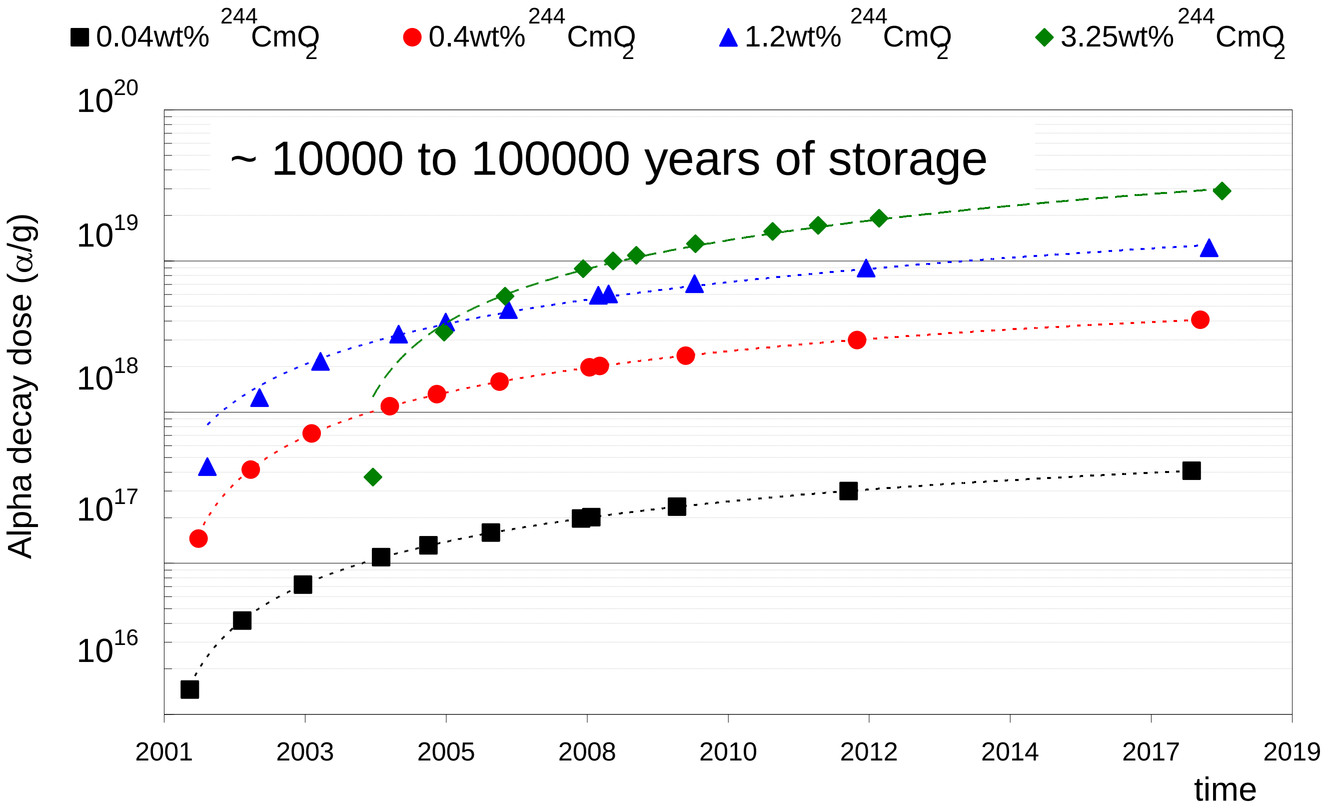
DM, CEA



1- Methodology: Cm doping

- SON68 glasses doped with 0.04, 0.4, 1.2, 3.25wt% of $^{244}CmO_2$
- International Standard Glass (ISG) doped with 0.7wt% of $^{244}CmO_2$



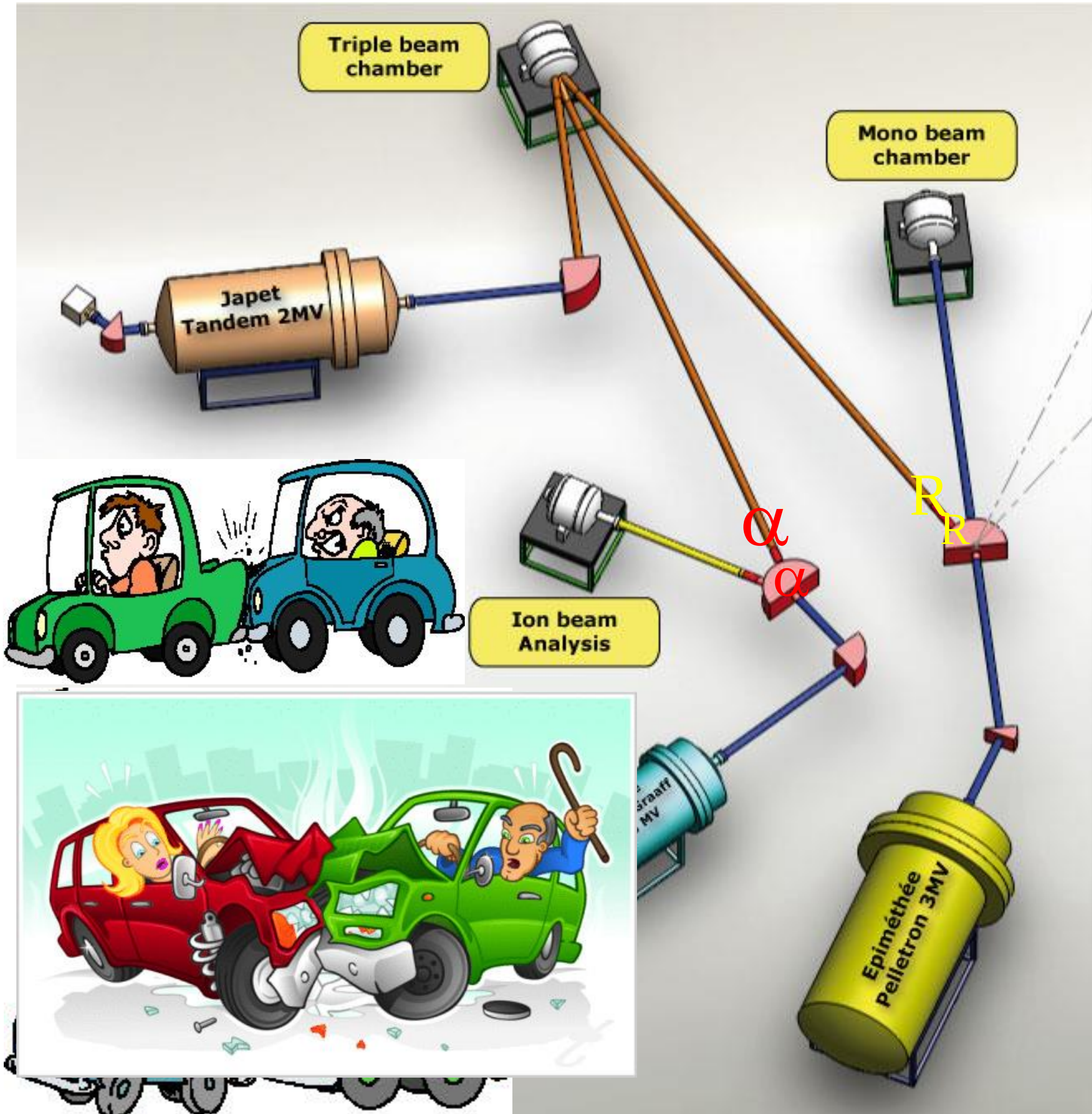


Mol%	SiO_2	Na_2O	B_2O_3	Al_2O_3	CaO	ZrO_2	Other oxides
ISG/CJ4	60.1	12.6	16.0	3.8	5.7	1.7	
R7T7	52.8	11.3	14.1	3.4	5.0	1.6	11.8

- Initial characterizations of the glasses (homogeneity, chemical composition)
- Periodical characterizations of the glass properties

1- Methodology: Ion beam irradiation experiment

Jannus Saclay,Orsay, Ganil



Alpha particles

Heavy ions (RN)

Mono beam

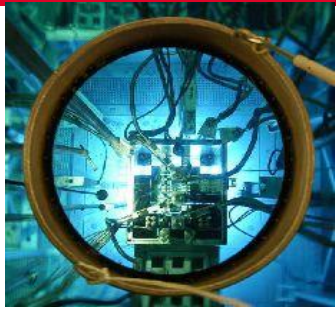
Alpha + Heavy ion

Heavy ion + Alpha

Simultaneous

Double beam

1- Methodology: In pile irradiation : $^{10}B(n,\alpha)^{7}Li$

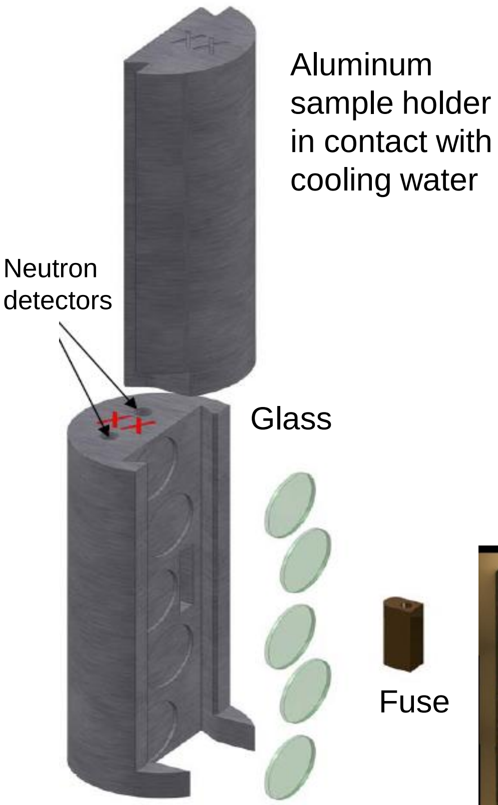


OSIRIS reactor, CEA SACLAY

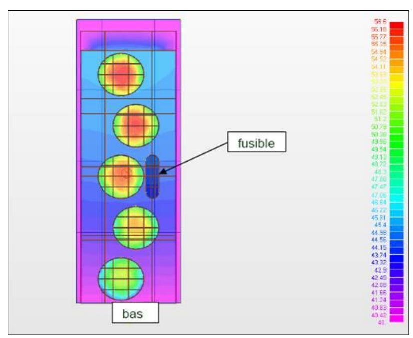
Glass samples : polished disks thickness 0.5 mm

	D1	D2	D3	D4
E (MeV)	He(1.47) + Li(0.84)			
Fluence (neutron cm^{-2})	5.9 ×10^{18}	1.2 ×10^{19}	3.5 ×10^{19}	5.2 ×10^{19}
Number of events (ion cm^{-3})	3.5 ×10^{19}	7.0 ×10^{19}	2.1 ×10^{20}	3.1 ×10^{20}
dE/dx_{nucl} (keV nm^{-1})	dE/dx(He) <0.03 dE/dx(Li) <0.06			
dE/dx_{elec} (keV nm^{-1})	dE/dx(He) <0.33 dE/dx(Li) <0.56			
E_{nucl} (GGy)	**0.06**	**0.13**	**0.39**	**0.57**
E_{elec} (GGy)	**5.16**	**10.45**	**30.69**	**45.71**
Dpa	**0.27**	**0.54**	**1.6**	**2.38**

~ 1 million years of disposal



Mol%	SiO_2	Na_2O	B_2O_3	Al_2O_3	CaO	ZrO_2	Other oxides
CJ1	67.7	14.2	18.1				
SON68	52.8	11.3	14.1	3.4	5.0	1.6	11.8



Thermal modeling and fuses observations after irradiation:

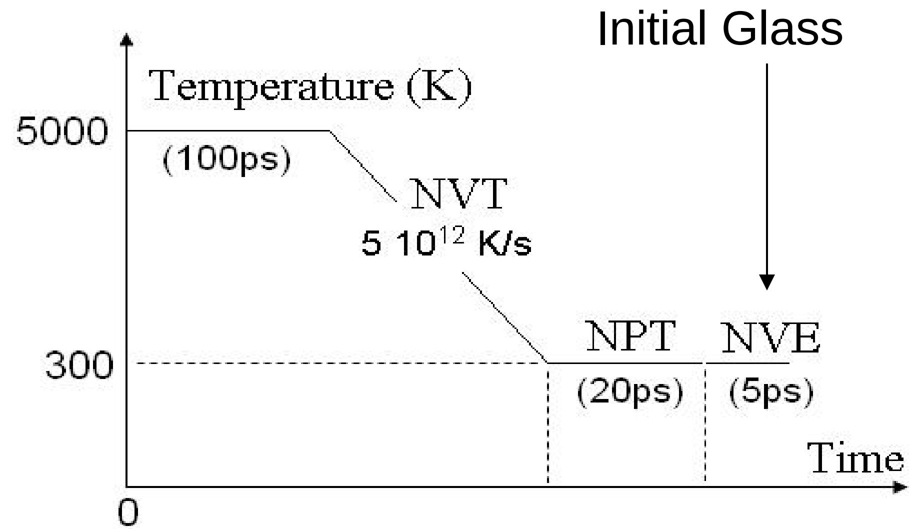
T<70°C

- Simplified borosilicate glasses (CJ1, CJ7)

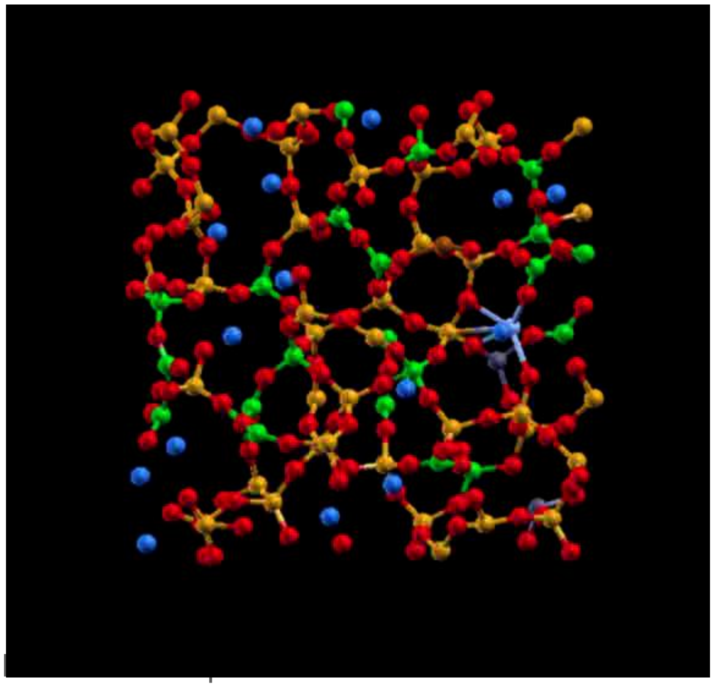
$$\phi(r_{ij}) = \frac{q_i q_j}{r_{ij}} + B_{ij} \exp\left(-\frac{r_{ij}}{\rho_{ij}}\right) - \frac{C_{ij}}{r_{ij}^6}$$

- Accumulation of displacement cascades caused by uranium atoms of energies from 700ev to 70keV

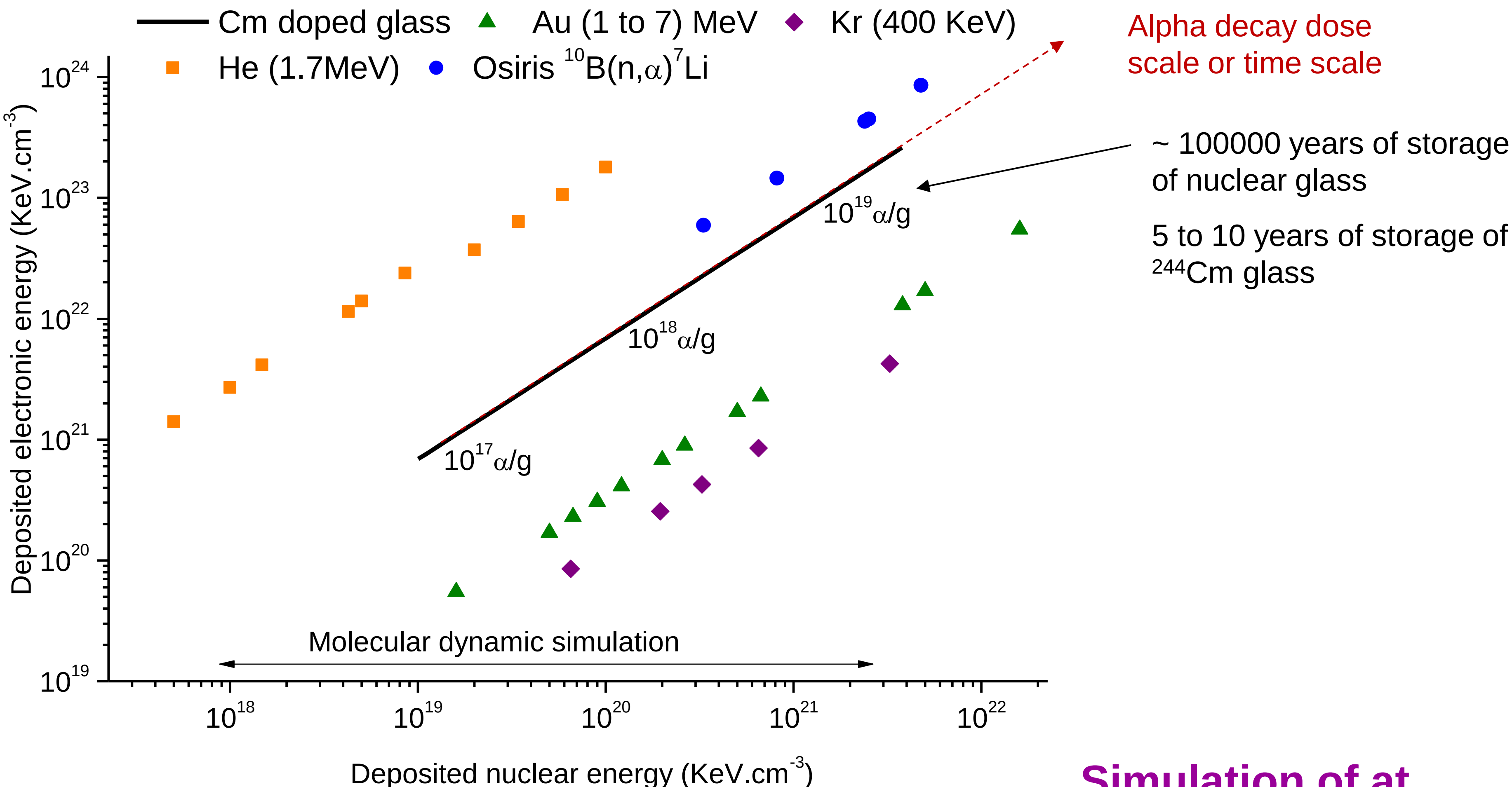
- Characterization of the structural modifications induced by displacement cascades (SRO and MRO)



Mol%	SiO_2	Na_2O	B_2O_3	Al_2O_3	CaO	ZrO_2	Other oxides
CJ1	**67.7**	**14.2**	**18.1**				
CJ7	**63.8**	**13.4**	**17.0**	**4.1**		**1.8**	
SON68	*52.8*	*11.3*	*14.1*	*3.4*	*5.0*	*1.6*	*11.8*



1- Methodology: Materials and irradiation conditions



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

Light ions irradiations (He) : mainly electronic interactions

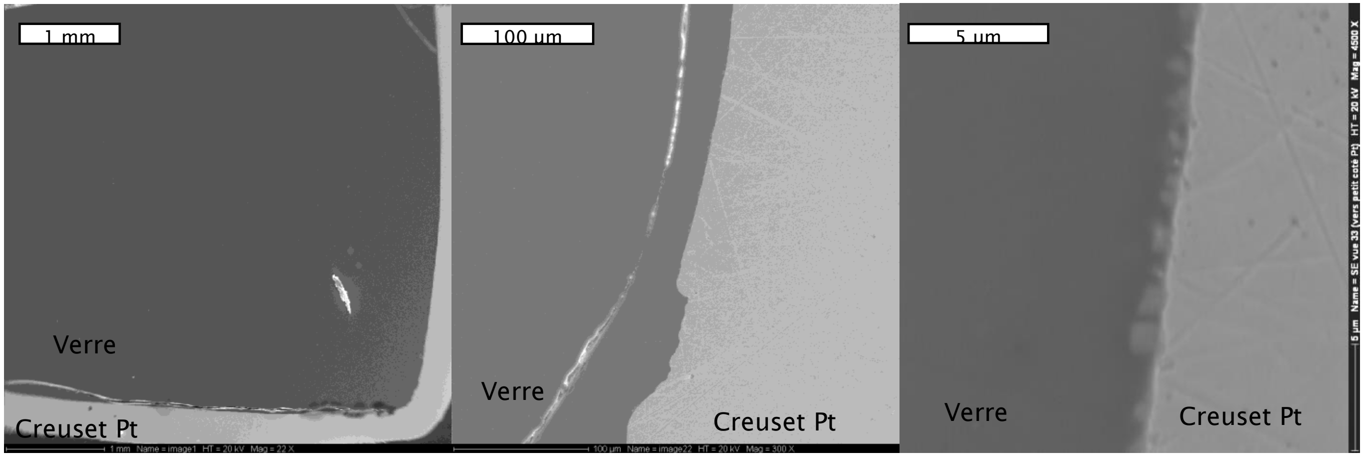
Heavy ions irradiations (Kr, Au) : mainly nuclear interactions

Doped glasses and OSIRIS irradiation : electronic and nuclear interactions

Molecular Dynamics : only nuclear interactions

2- Effect on the glass microstructure

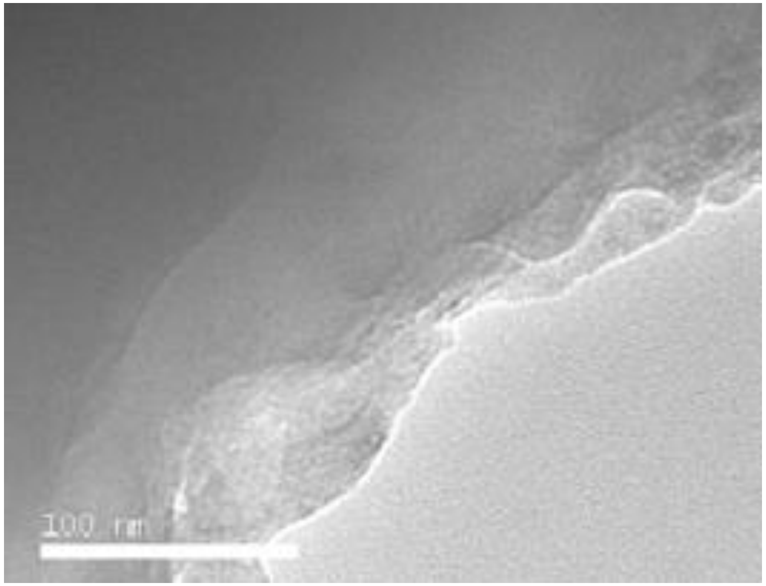
^{244}Cm SON 68 glass : SEM (CEA Marcoule), alpha decay dose $2x10^{19}\alpha/g$



(Around 100000 years of storage)

S. Peuget et al. JNM 44 (2014)

^{244}Cm SON 68 glass : TEM (ITU Karlsruhe), alpha decay dose $8x10^{18}\alpha/g$



Homogeneous microstructure, without bubbles, phase separation or crystallization

Stability of the glassy state

ACTINET i3 Integrated Infrastructure Initiative for Actinide Science

2- Effect on the macroscopic properties? Density

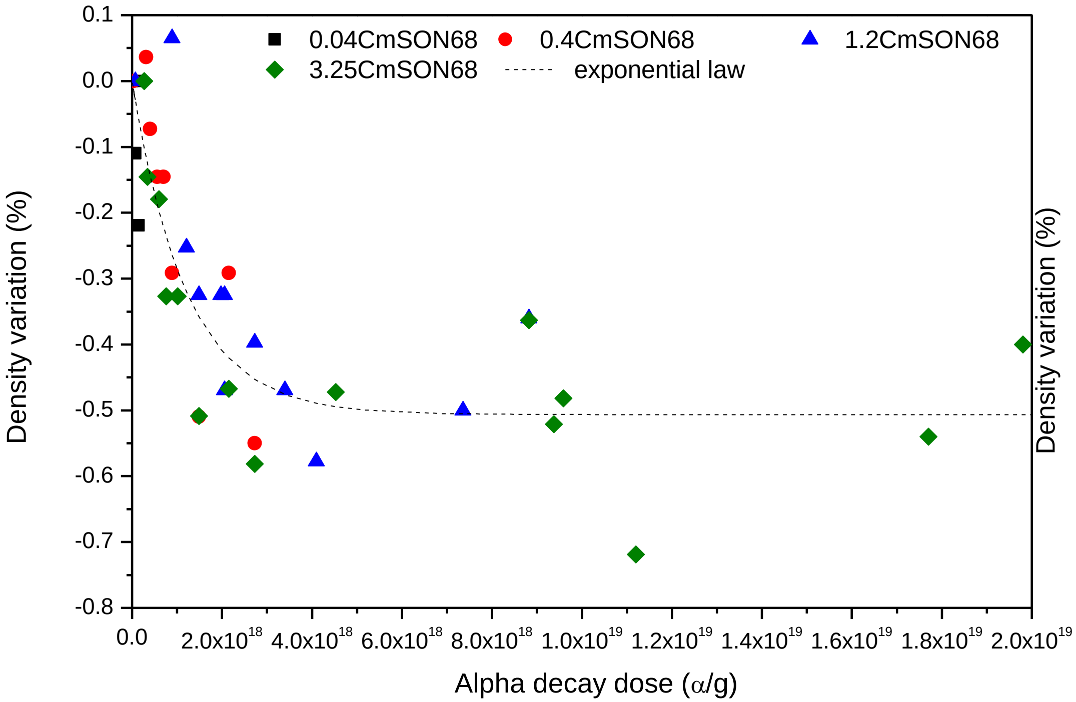
Slight decrease of the glass density (0.5%)

No effect of the dose rate

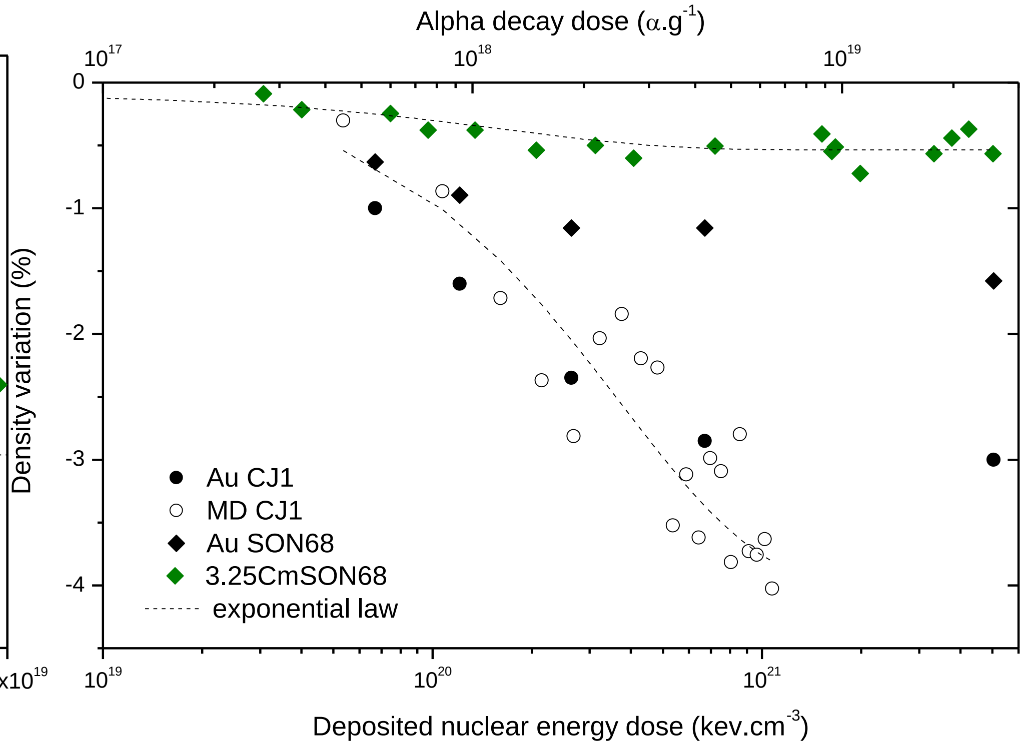
Stabilization of the evolution at around 4x10^{18} α/g

Evolution according to an exponential law (direct impact model)

- ✓**Variations correclty simulated by external irradiations wih heavy ions and MD simulation**
- ✓**Swelling level is lower under α decays irradiation (0,5% compared to 1,2% Au irradiation)**



S. Peuget et al. J. Nucl. Mat. 354 (2006) 1



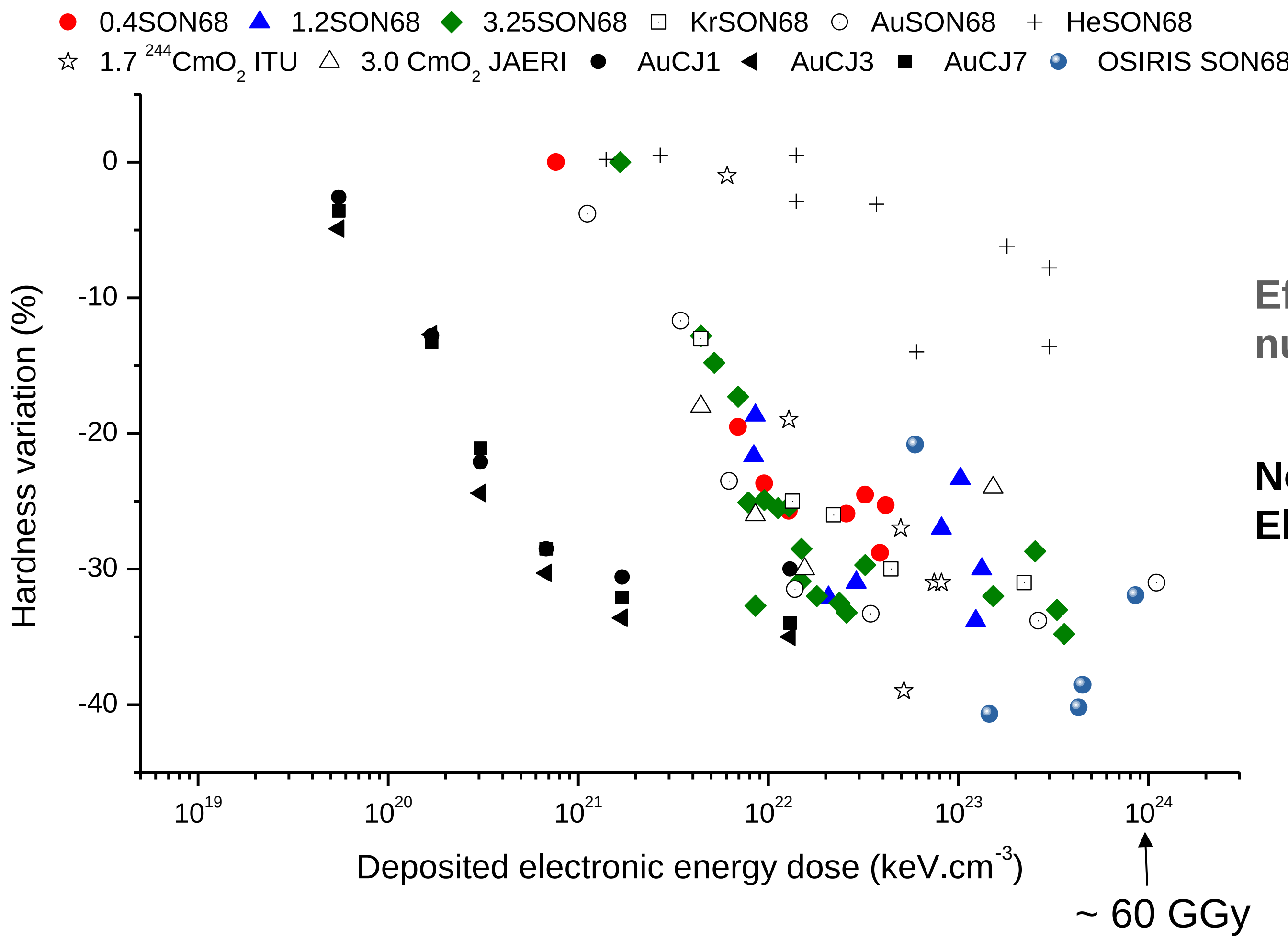
S. Peuget et al. J. Nucl. Mat. 354 (2014) 1

2- Effect on the macroscopic properties?

Mechanical properties: example of hardness

Decrease of hardness on curium doped glasses and ions irradiated glasses

He induced lower changes



Effect of electronic or nuclear interactions?

No agreement versus Electronic dose

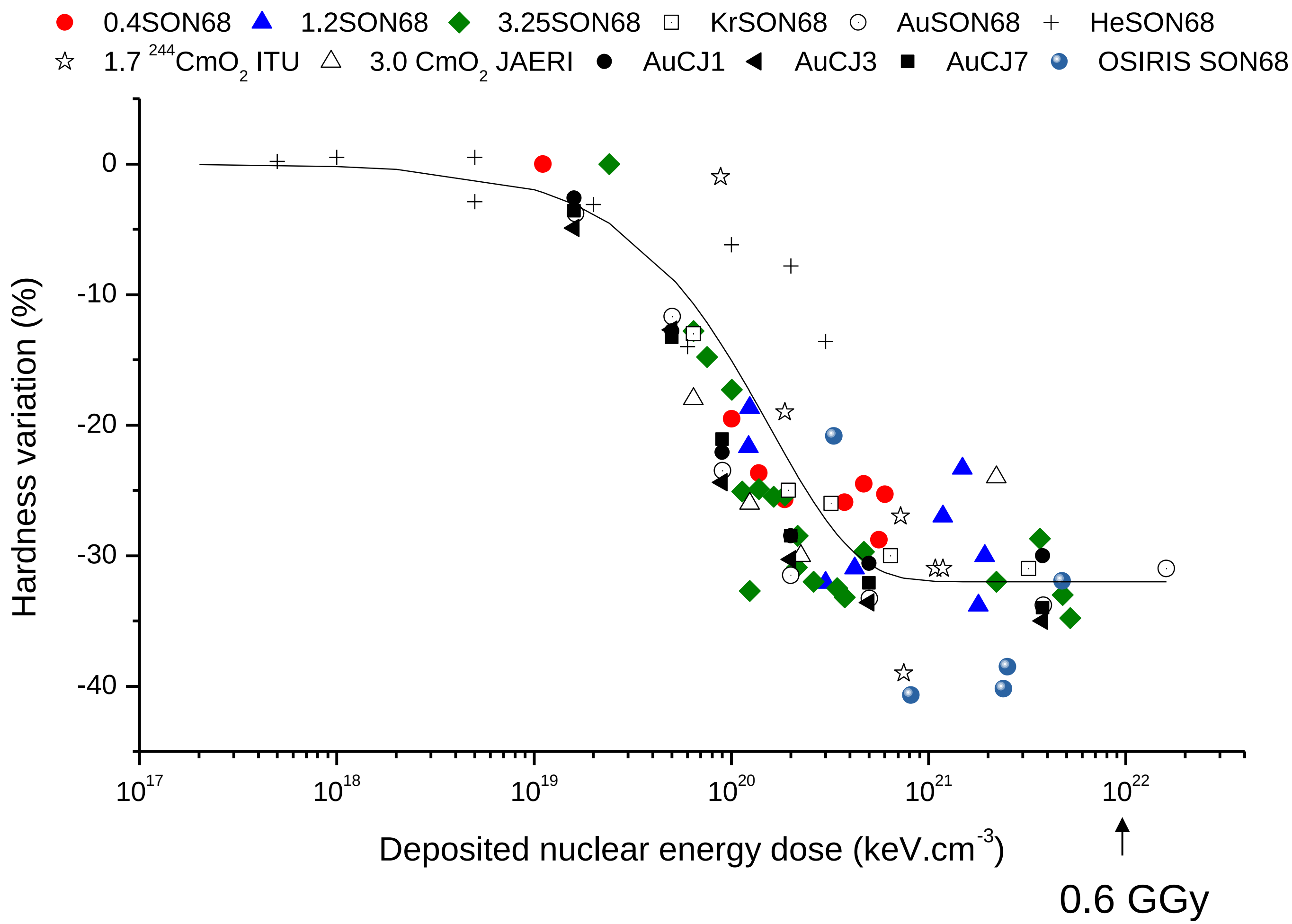
S. Peuget et al, NIMB 246 (2006) 379 — S. Peuget et al. JNM 354 (2014) 1 — Mir et al. . JNM 469 (2016) 244

2- Effect on the macroscopic properties?

Mechanical properties: example of hardness

Decrease of hardness on curium doped glasses and heavy ions irradiated glasses

He induced lower changes



Effect of electronic or nuclear interactions?

Quite good agreement between doped glasses and heavy ions irradiated glasses

⇩

Effect induced by nuclear interactions, rôle of RN !

S. Peuget et al, NIMB 246 (2006) 379 S. Peuget et al. JNM 354 (2014) 1 Mir et al. . JNM 469 (2016) 244

Outline

1- Effects of beta decays

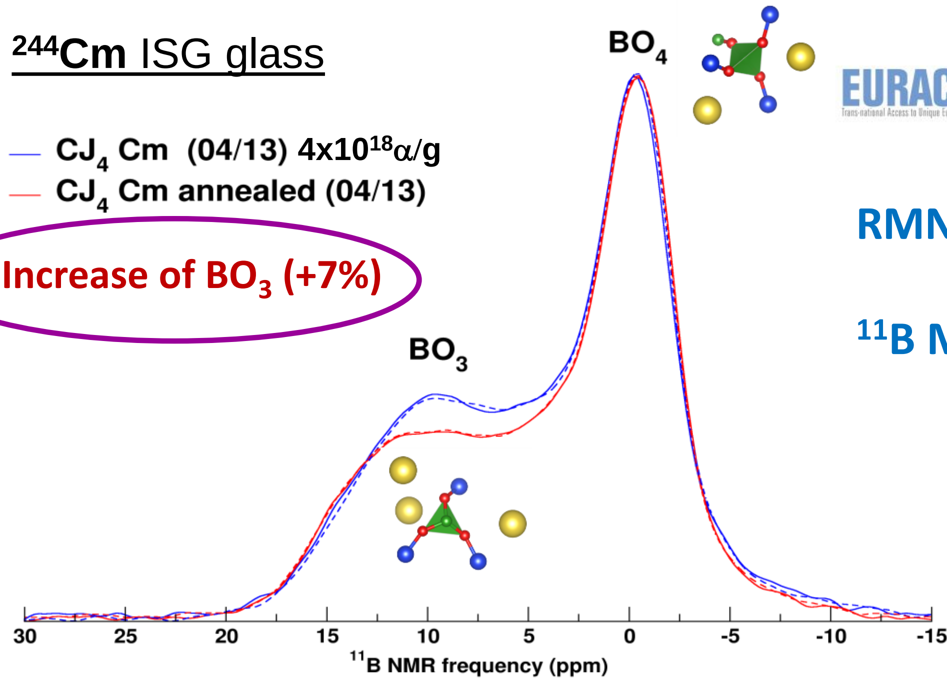
2- Effects of α decays

3- Effects of α decays on the glass structure

4- Roles of Recoil Nuclei and Alpha Particle?

3- Effect on glass structure : SRO around B

^{244}**Cm** ISG glass



Increase of BO_3 (+7%)

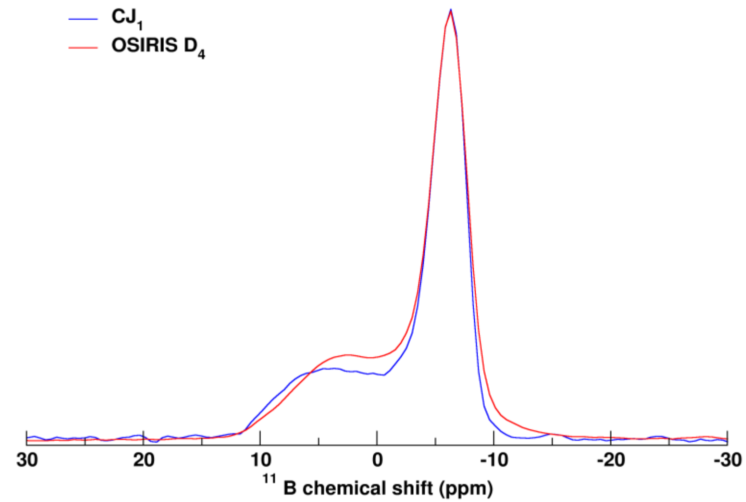
T. Charpentier et al. Scientific Reports 6:25499 (2016)

RMN at ITU

^{11}B MQMAS

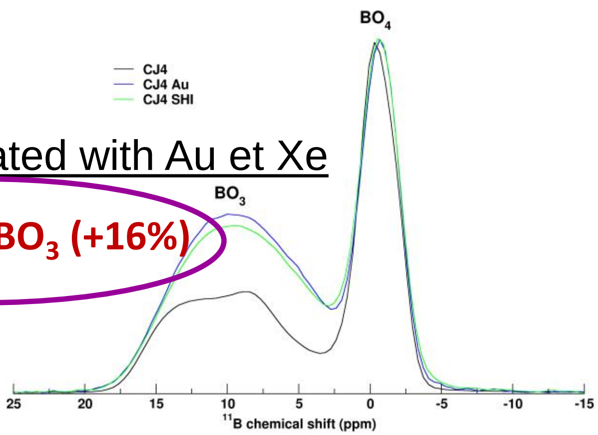
CJ1 irradiated in OSIRIS reactor

S. Peuget et al, NIMB 327 (2014) 22-28



ISG irradiated with Au et Xe

Increase of BO_3 (+16%)



C. Mendoza et al. NIMB 325 (2014) 54-65

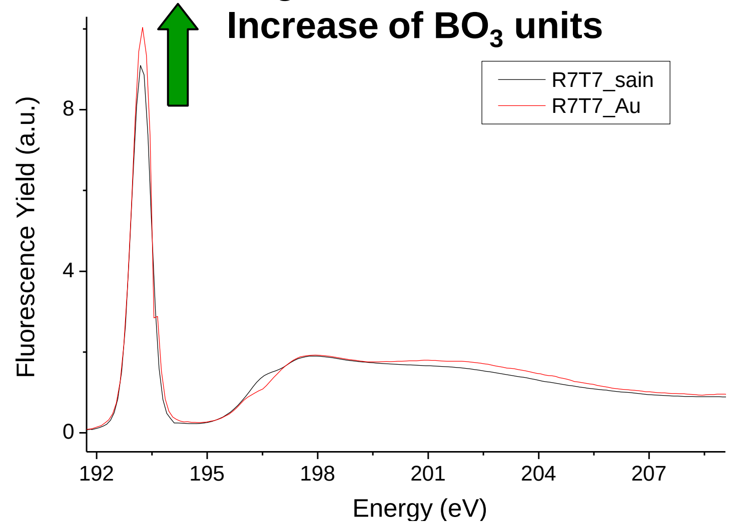
Conclusion :

Partial conversion BO_4 into BO_3 Complex and simplified glasses

BO_3 increase is lower under α decays irradiation

Xanes B K edge: R7T7 irradiated with Au

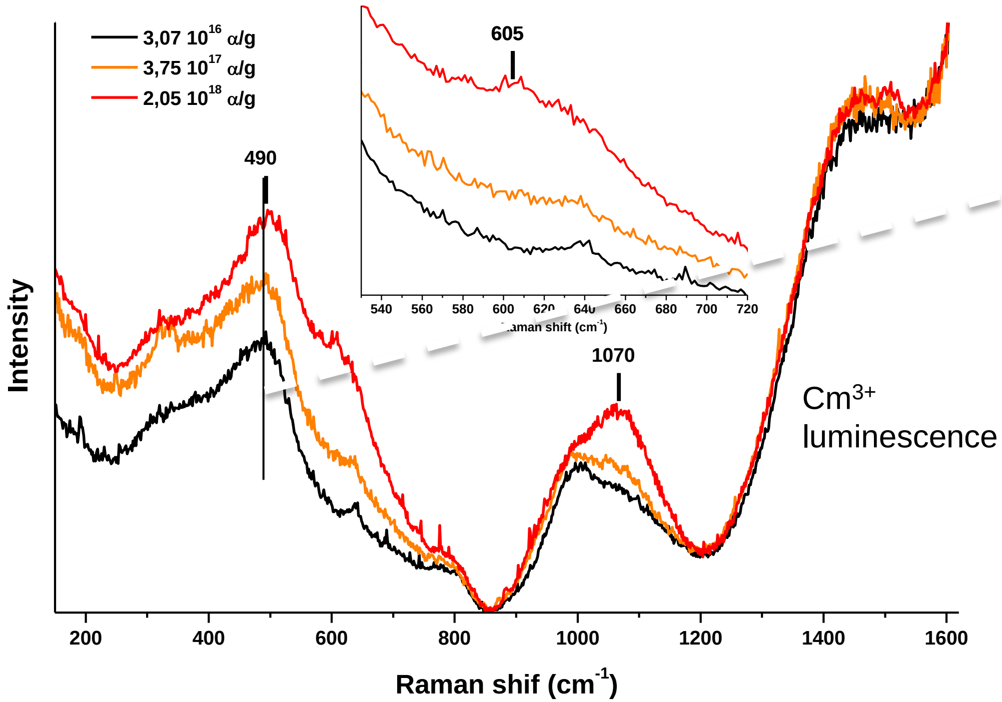
Increase of BO_3 units



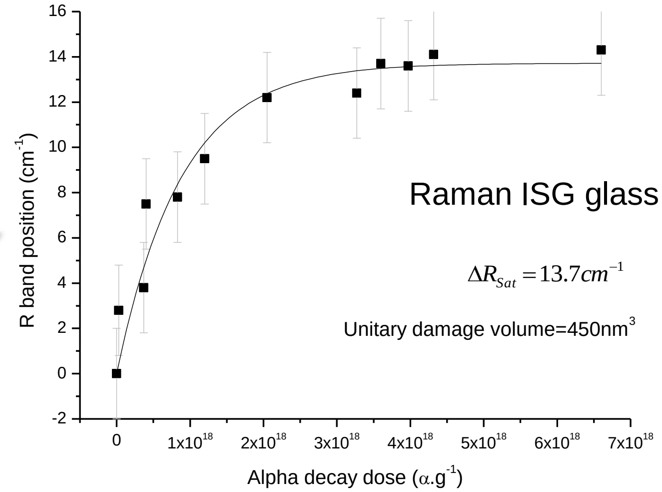
G. Bureau, thesis, (2008)

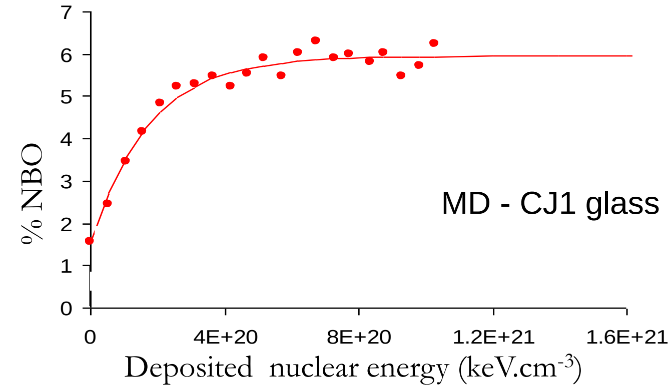
Effect on glass structure : SRO around Si and MRO

Raman spectroscopy on Cm doped ISG (Atalante, DHA)



C. Mendoza et al. Proc. Chem. 7 (2012) 581





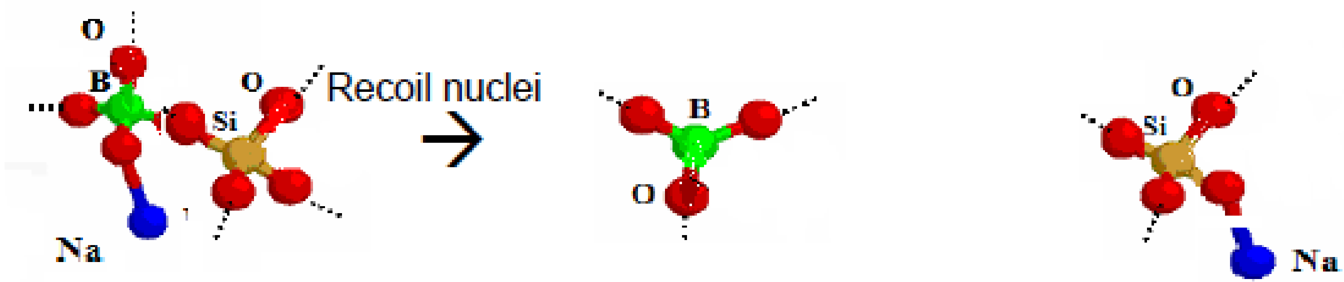
J.-M. Delaye et al, J. Non-Cryst. Solids 357 (2011) 2763

- **Increase of Q3 contribution in ISG glass : more NBO**
- **Slight shift of the vibration band around 500cm^{-1}**

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

Modification of the Short Range Order

Increase of trigonal boron, increase of NBO



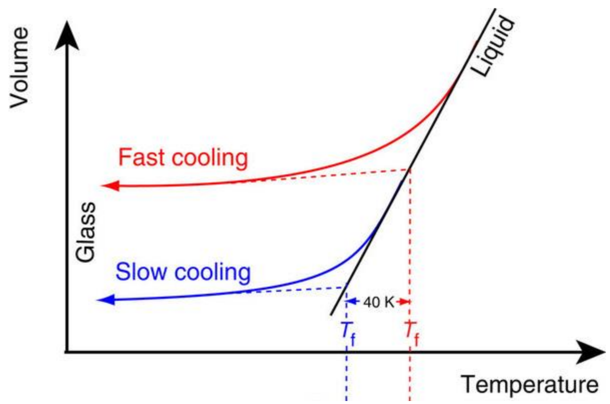
Modification of the Medium Range Order

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

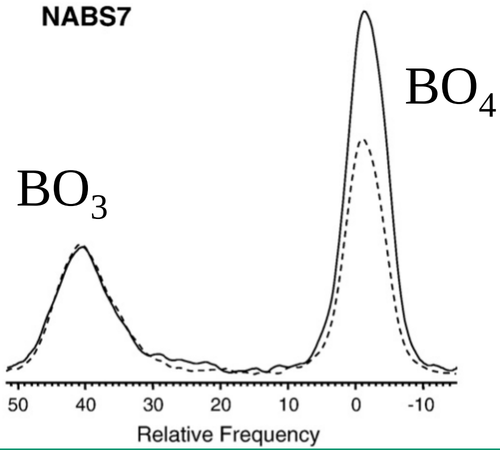
Effects similar to those induced by thermal quenching of a molten glass

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

Decrease of the glass density



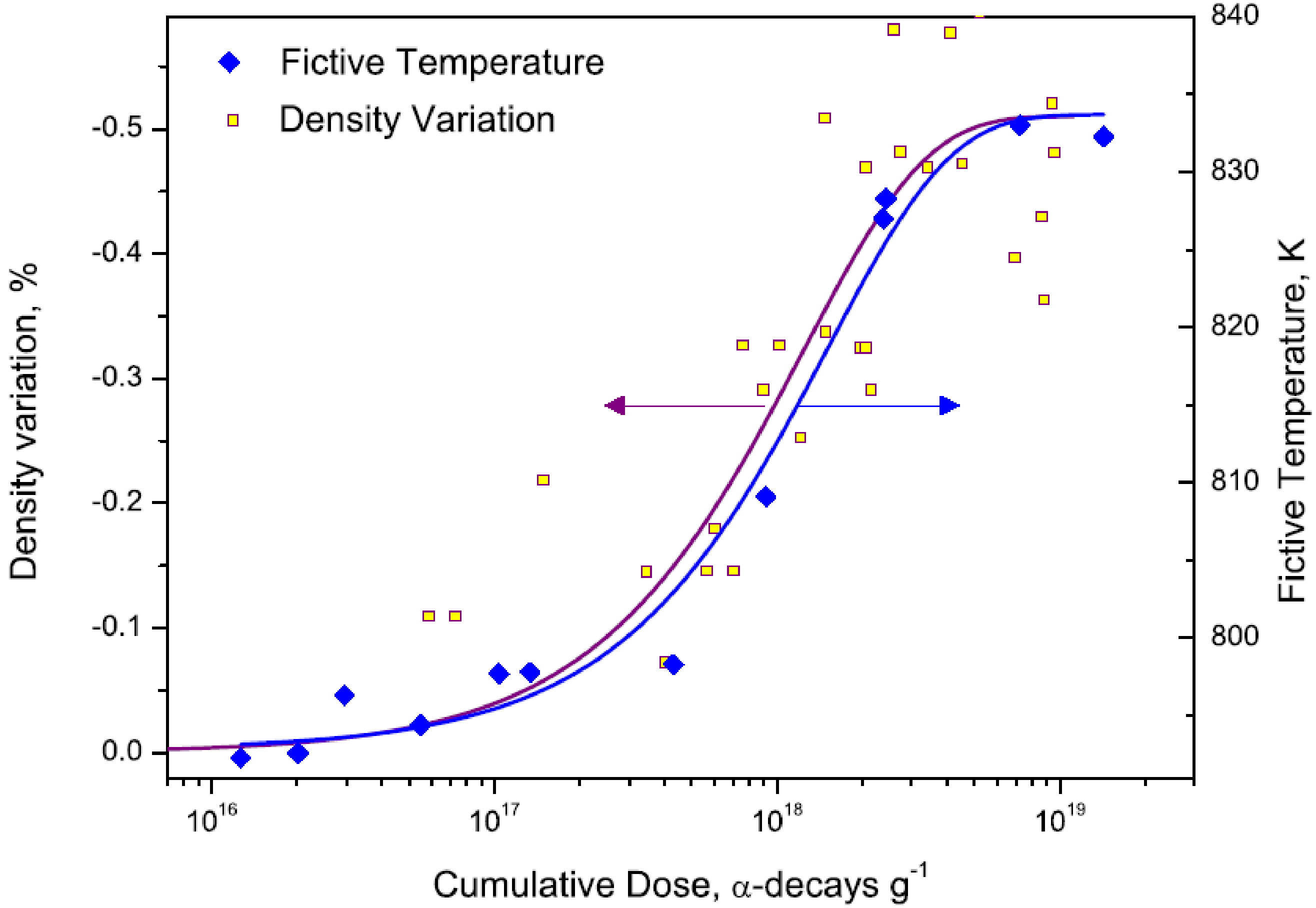
[11]B NMR on quenched and annealed glass



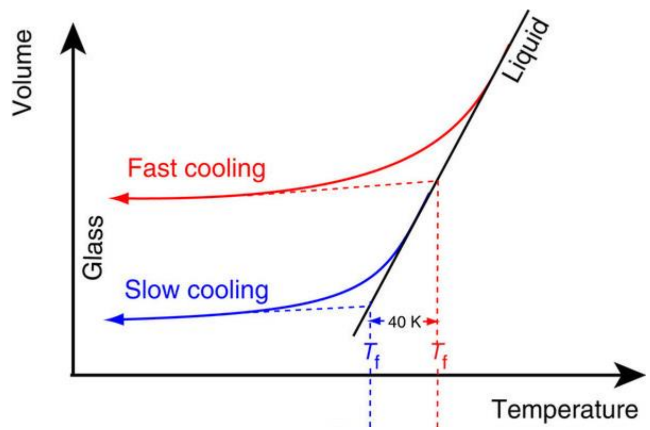
Wu and Stebbins JNCS 356 (2010)

3- Effects on the glass structure? Fictive temperature

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



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



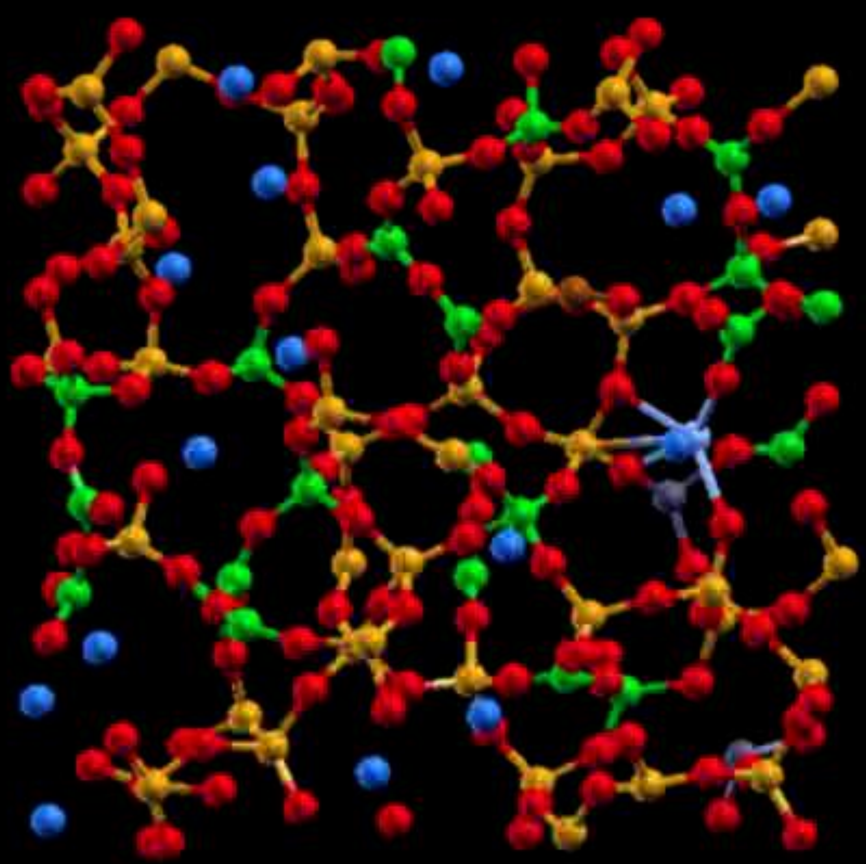
Increase of the glass fictive temperature with alpha decay dose

Formation a new structure similar to a fast quenched glass
New metastable phase induced by irradiation

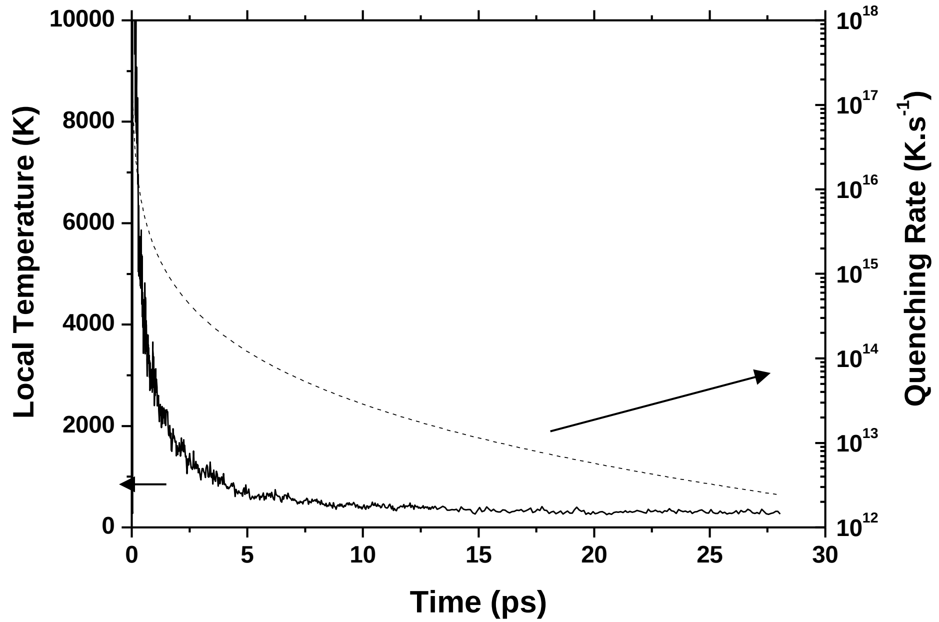
3- Effects on the glass structure? Ballistic damage

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

JM Delaye, PRB 61 (2000) 14481



1. Balistic phase
2. Thermal phase



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

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

Supervitrification

Outline

1- Effects of beta decays

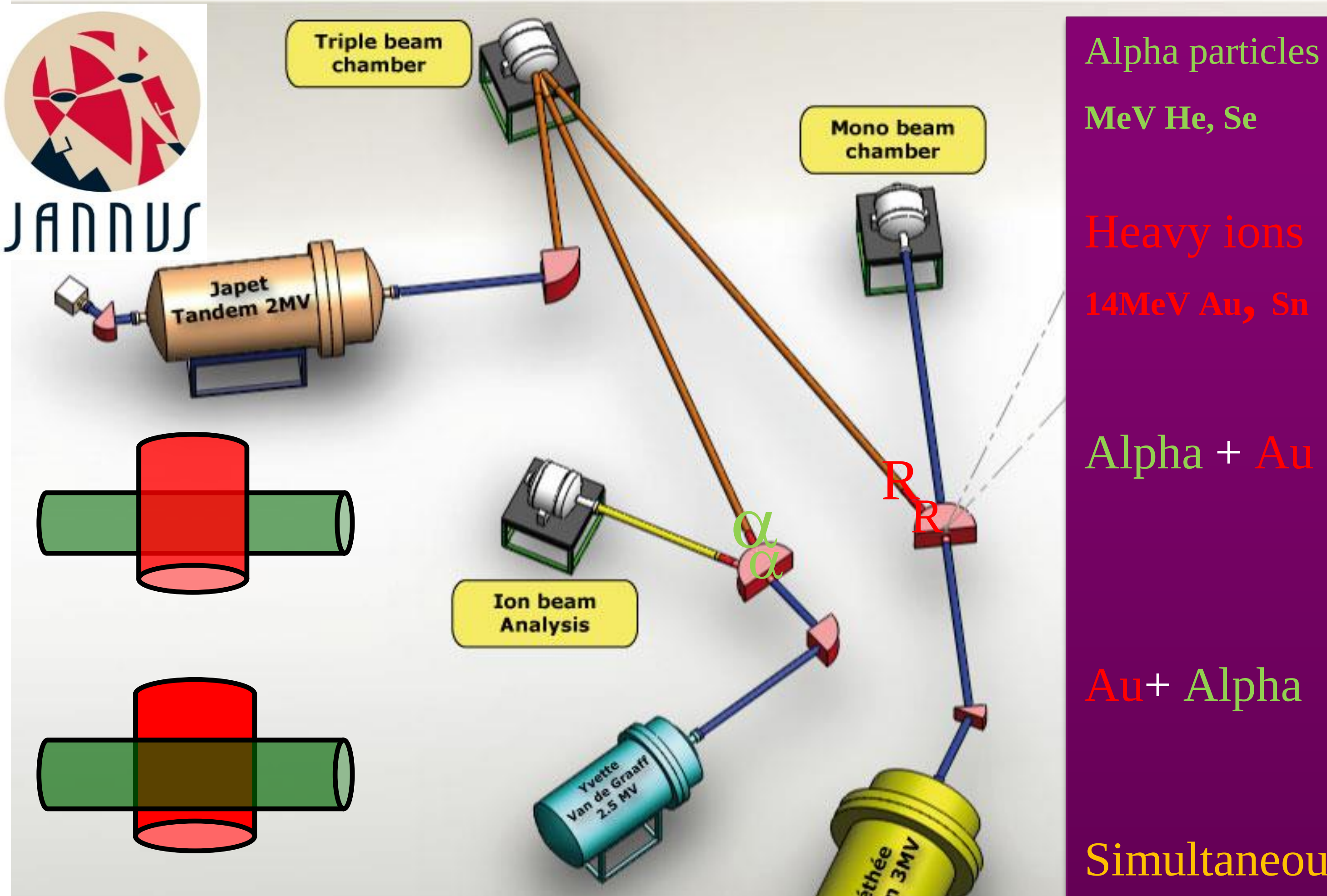
2- Effects of α decays

3- Effects of α decays on the glass structure

4- Roles of Recoil Nuclei and Alpha Particle?

4- Roles of Recoil Nuclei (RN) and Alpha Particle (AP)?

Studied glass: a-SiO_2, CJ1, ISG, SON68

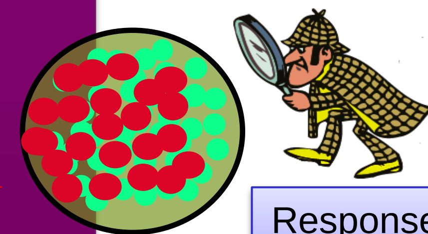


Alpha particles 2 MeV He, Se

Heavy ions 14MeV Au, Sn

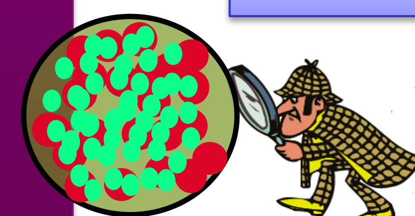
Single beam irradiation
Response of the Pristine Glass

Alpha + Au

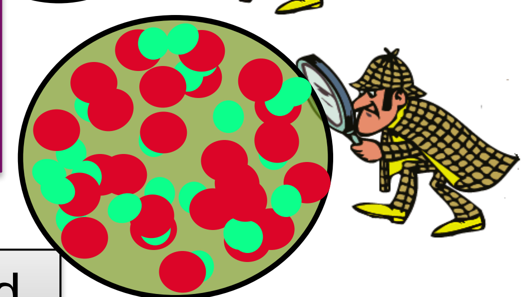


Response of the Pre-damaged glass

Au+ Alpha



Simultaneous

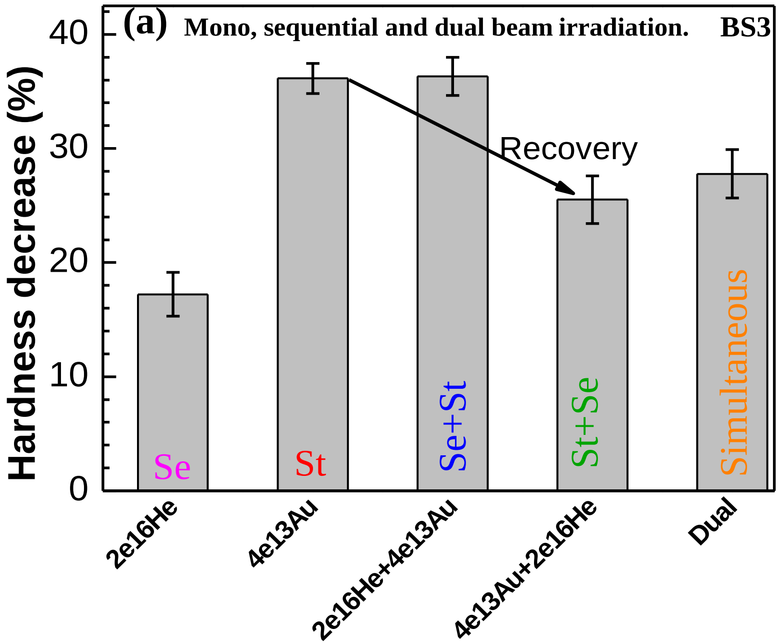


Mutual Interaction and slow buildup with time

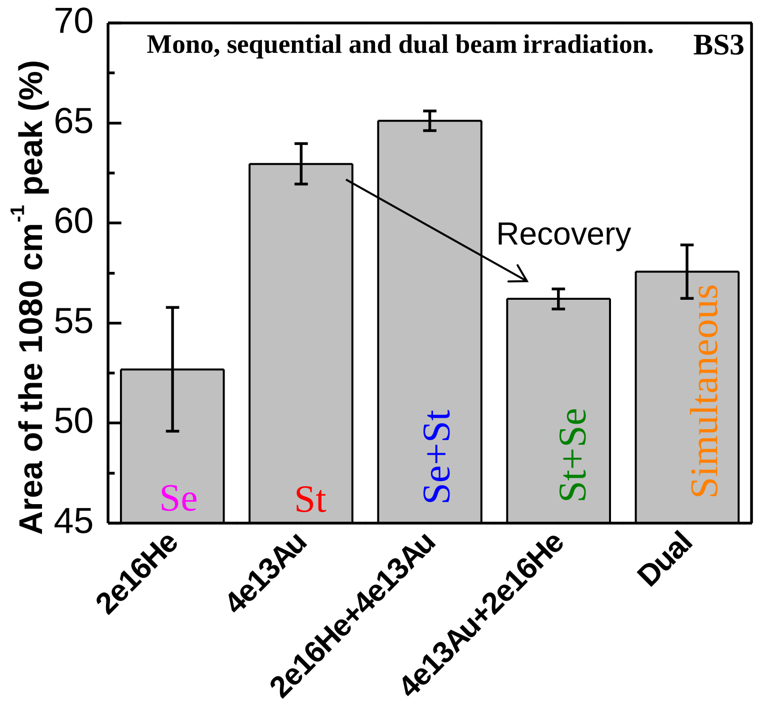
Ion	Se (keV/nm)	Sn (keV/nm)	Flux(cm²/s)
2 MeV Alpha	0.4	10^{-4}	$2x10^{13}$
14 MeV Au	2.5	2	$1x10^{10}$

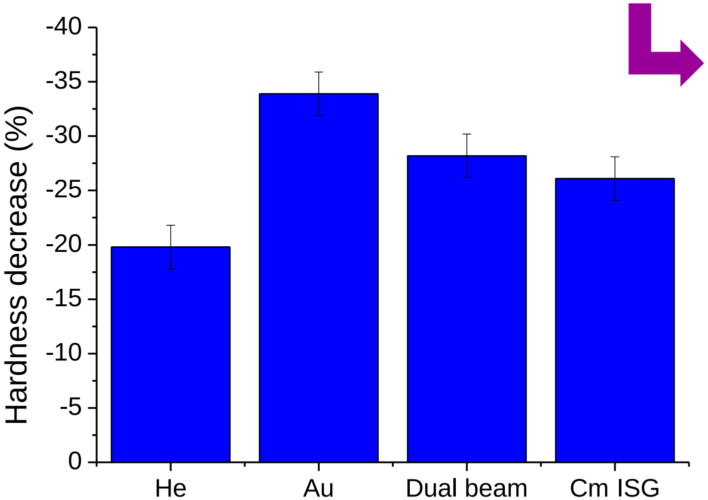
4- Roles of Recoil Nuclei (RN) and Alpha Particle (AP)?

A.H. Mir et al, Eur. Phys. Lett. 112 (2015) 36002



A.H. Mir et al, Scientific Reports 6:30191 (2016)

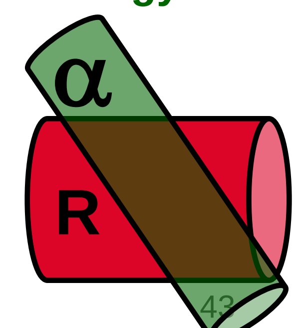




Role of both nuclear and electronic stopping powers

- **Heavy ions: main damage (supervitrification)**
- **Alpha particle: recovery effect due to electronic energy loss**

Explain the lower property variation observed on ^{244}Cm doped ISG glass compared to heavy ions irradiated ISG glass



4- Roles of Recoil Nuclei (RN) and Alpha Particle (AP)?

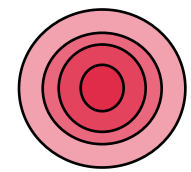
Mechanism of α particle induced recovery? Thermal spike?

Se: Electronic energy loss (heating of electrons)

→ Transfer of energy from electrons to atoms (e-phonon coupling)

→ Lattice heating (Melting at high energy input)
Thermal spike

Thermal history around the ion track



→ Cooling of lattice (10^{12} to 10^{13} K/s)

→ **High Se:** Defect freezing, phase transformation…

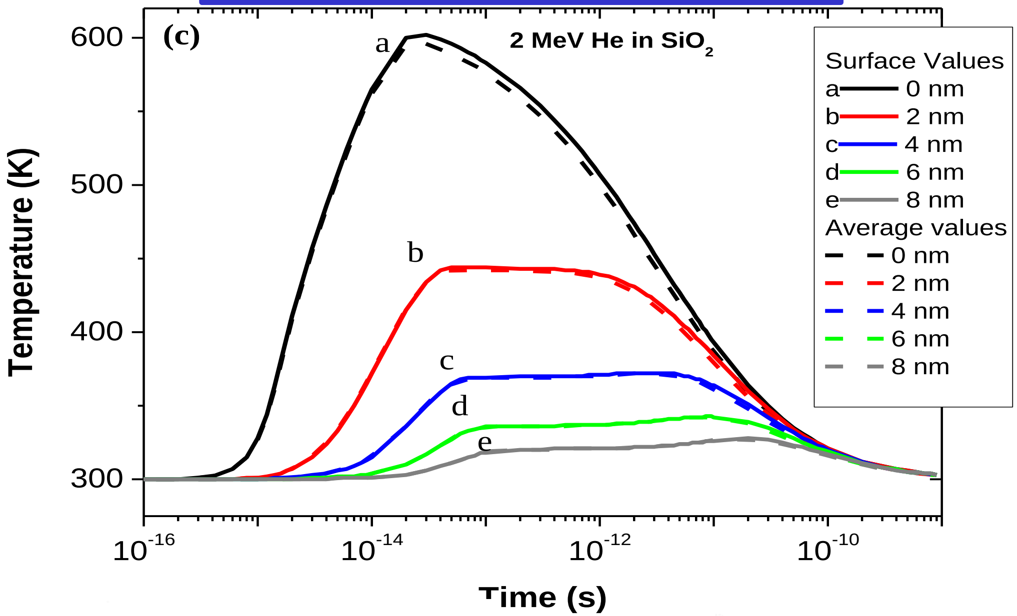
Low Se: Defect annealing for temperatures below melting temperature in a glass ?

Thermal spike model
(Applied on many materials).

Kobetich et al, Phys. Rev. 170, 391 ; Gervais et al, NIMB-88, 355 ; Meftah et al., Phys. Rev B-49,12457 ; Toulemonde et al, Acta. Phys. Pol. A-109, 311.

Mechanism of alpha induced recovery? Ionization induced annealing?

Amorphous SiO2



Temperature of 450 to 600 K in the center of the track

O diffusion activated near 450 K.

diffusion coefficient~10 A^2ps^{-1}

Alpha induced thermal spike could activate oxygen diffusion leading to recombination with the pre-existing defects and partial damage recovery.

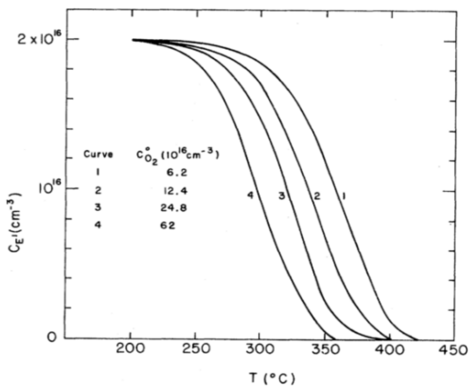


FIG. 10. Plot of E' concentration $C_{E'}$, vs temperature for several different initial concentrations of $O_2(C^0_{O_2})$, from Eq. (4). Other parameters are as given in text.

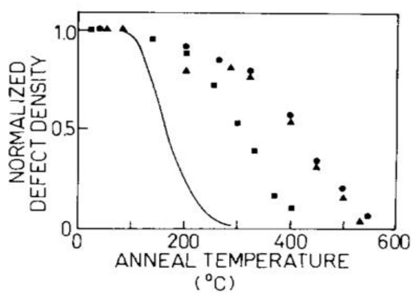


Fig. 7. 10 min isochronal annealing curves for (●) 16.5% densified Suprasil. 1, 76 Mrad irradiation, (▲) 16% densified Suprasil W1 76 Mrad irradiated and (■) PECVD oxide, 10% densified, 20 Mrad irradiated. The solid line shows the annealing curve for E'_1 defects in undensified a-SiO_2.

A.H. Mir et al, Scientific Reports 6:30191 (2016)

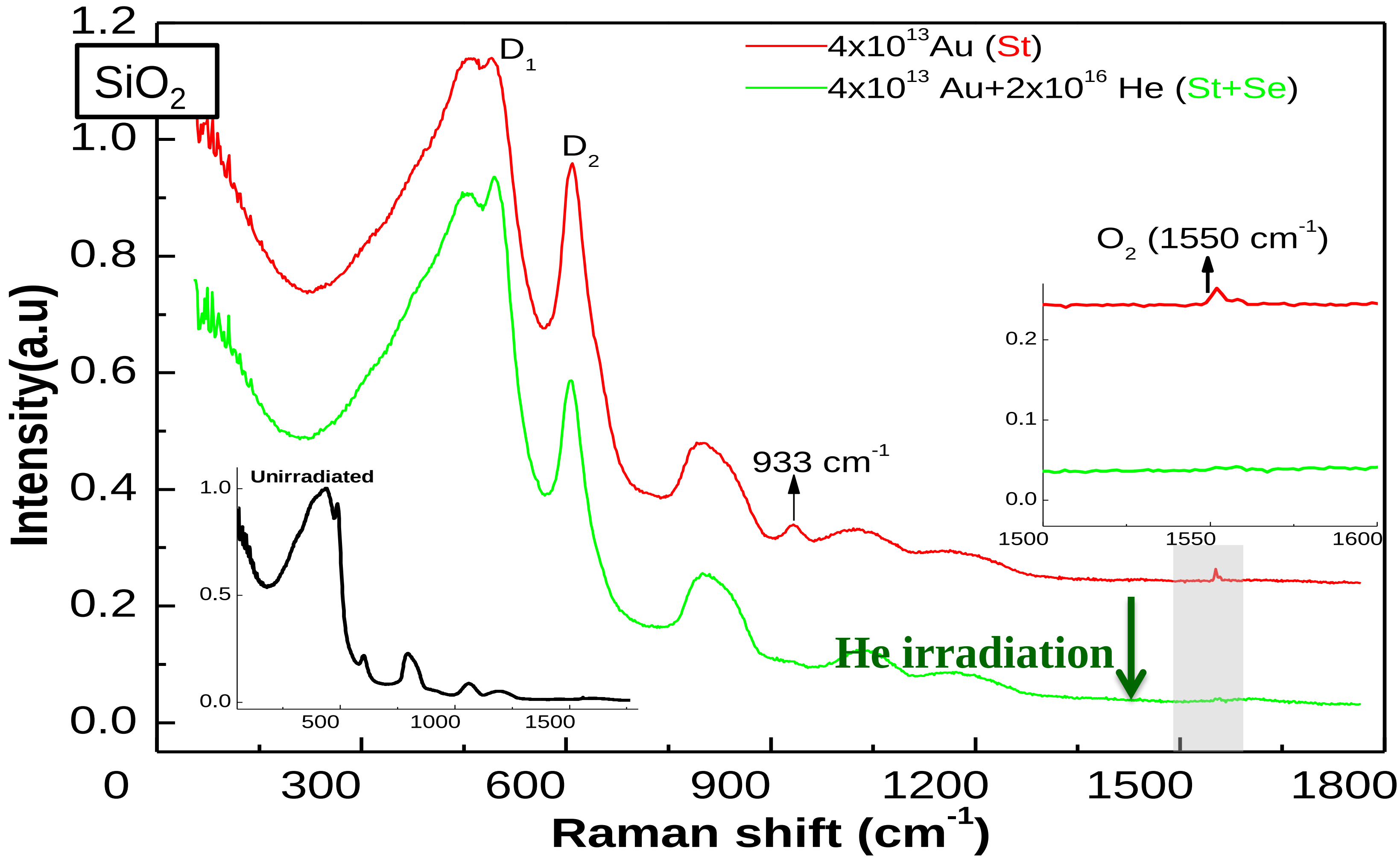
Edwards et al 1982; PRB-26

Devine 1990; NIMB-46

4- Roles of Recoil Nuclei (RN) and Alpha Particle (AP)?

Ionization induced annealing is possible and involves O atoms

A.H. Mir et al, Scientific Reports 6:30191 (2016)



Ionization by He irradiation

O_2 ⟶ 2 O

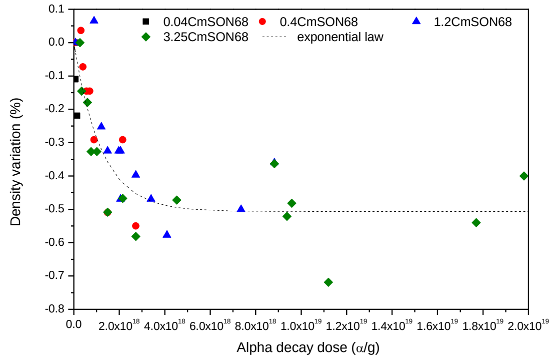
Activated by the thermal spike

Reaction of O° with defects ⟶ Structural relaxation

D.L. Griscom, Nucl. Instrum. Methods Phys. Res. B1 (1984) 481–488

Conclusion on alpha decays effects

- Slight modification of density and mechanical properties
- Glass is still homogeneous (SEM and TEM scale)
- No effect on initial alteration rate
- Modification of glass Short Range Order (boron coordination, NBO …)
- Modification of Medium Range Order (ring statistic, angle distribution)
- No effect of accelerating the time scale



Modifications observed in the first $4x10^{18}\alpha/g$ according to a direct impact model … (1st approx)

Saturation when all the glass has been damaged by recoil nuclei events and alpha particles

Recoil nuclei : supervitrification of the glass (1&2)

Alpha particles : partial repair of the damage (3&4)

Double ion beam irradiation is necessary to accurately simulate α decays

Prospects :

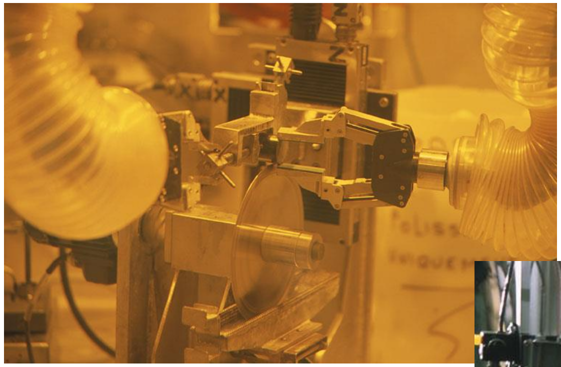
Coupling alpha and beta decays and thermal history

Is helium generation a problem?

Thank you for your attention !!!

Special Thanks to

DHA - Atalante



DE LA RECHERCHE À L'INDUSTRIE

THANKS TO

Funded by **CEA** and **AREVA NC**

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