DE LA RECHERCHE À L'INDUSTRIE

FRACTURE IN OXIDE GLASSES

Financement

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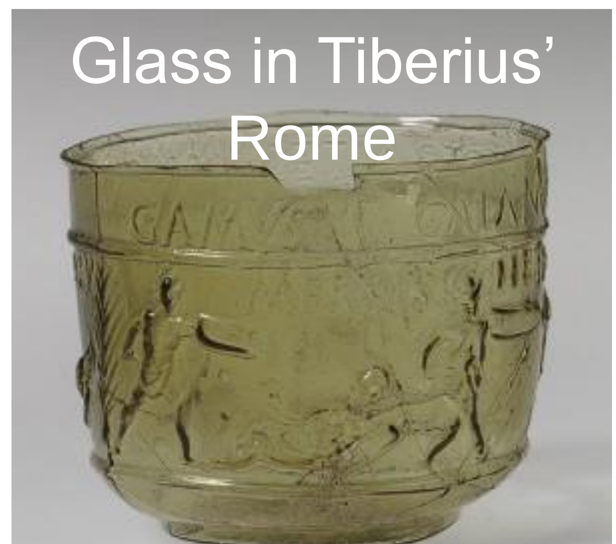
Gif-sur-Yvette, France

MechGlass2024

DE LA RECHERCHE À L'INDUSTRIE

DAMAGE PROPAGATION IN OXIDE GLASSES



http://www.strangehistory.net/2011/02/20/flexible-glass-in-tiberius-rome/

DELAYED FRACTURE

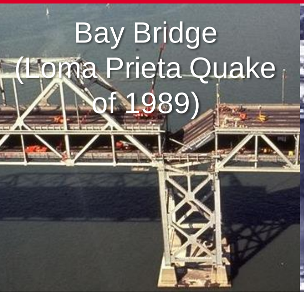
~8 minutes to failure

Polariscope
Visible
stresses

Polariscope
Non-visible
stresses

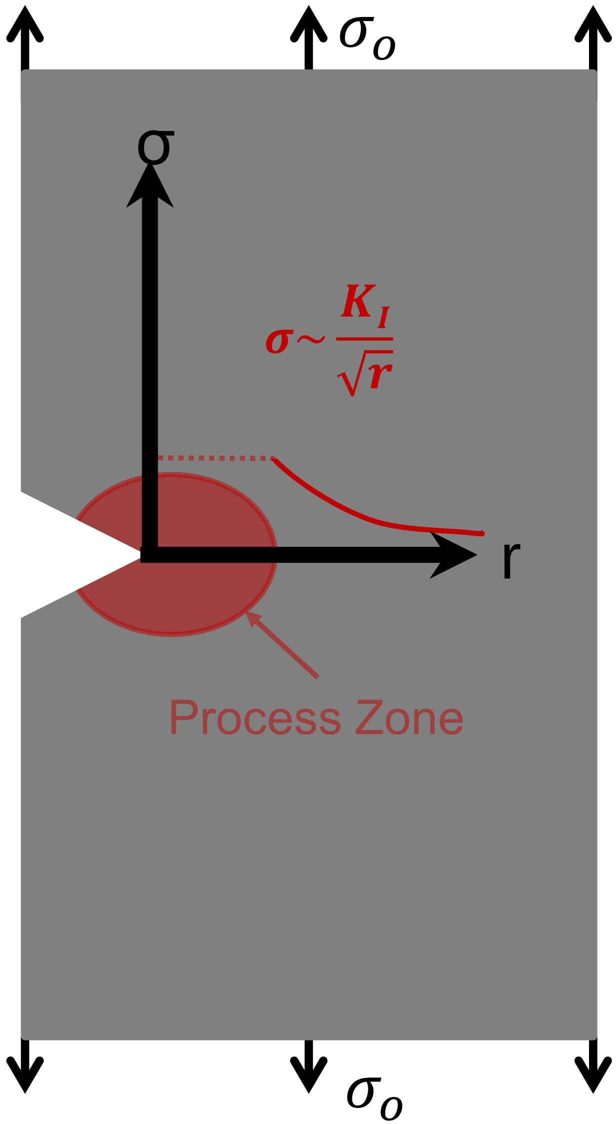
Corning Museum of Glass
www.cmog.org
YouTube

OUTLINE



- Introduction: Stress corrosion cracking
- Measuring the process zone size
  - What is the process zone
  - Results || and ⊥ to crack front
- SBN Glasses
  - Understanding mesoscale structural fluctuations
  - Mesoscale fluctuations to physical properties
  - Mesoscale fluctuations to fracture properties
- Conclusion

CRACK FRONT STABILITY …
IN LIEU OF A CORROSIVE ENVIRONMENT



Lawn, *Fracture of Brittle Solids*,1993

Irwin (1955), Orrowan (1957)

K_I: stress intensity factor

Stress intensity factor

stress near the crack tip due to σ_o
or residual stresses

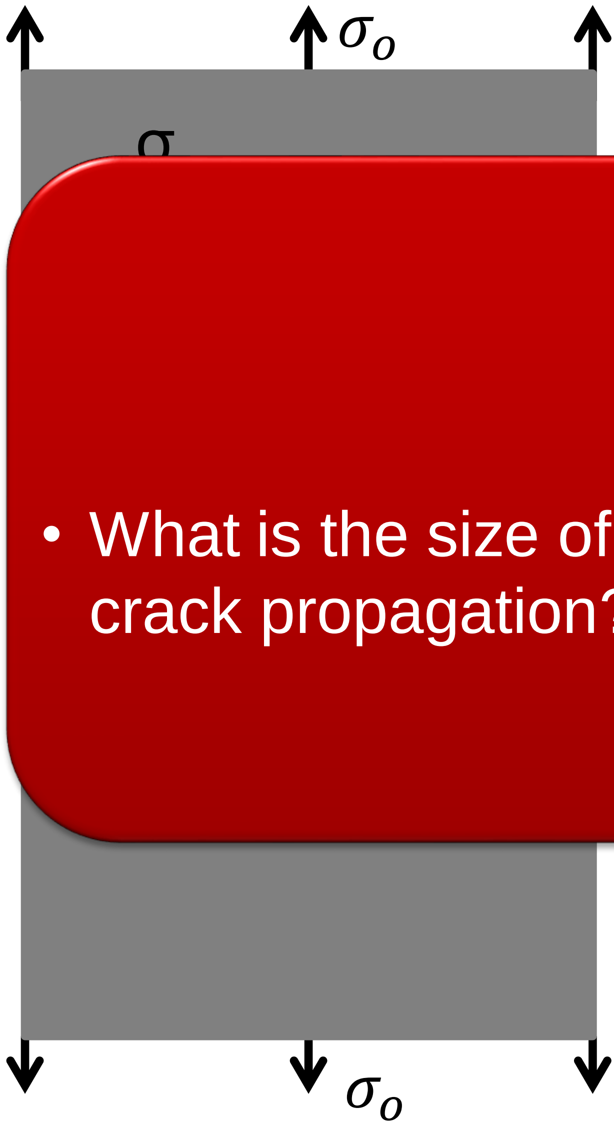
$$K_I = \sigma_o \sqrt{crack} f(geometry)$$

… Assessed by finite elements

Propagation of the crack front

$K_I > K_{Ic}$ Unstable (μs)

K_{Ic} : **fracture toughness**



Irwin (1955), Orrowan (1957)

1^{st} Question?

- What is the size of the zone controlling the dynamics of crack propagation?

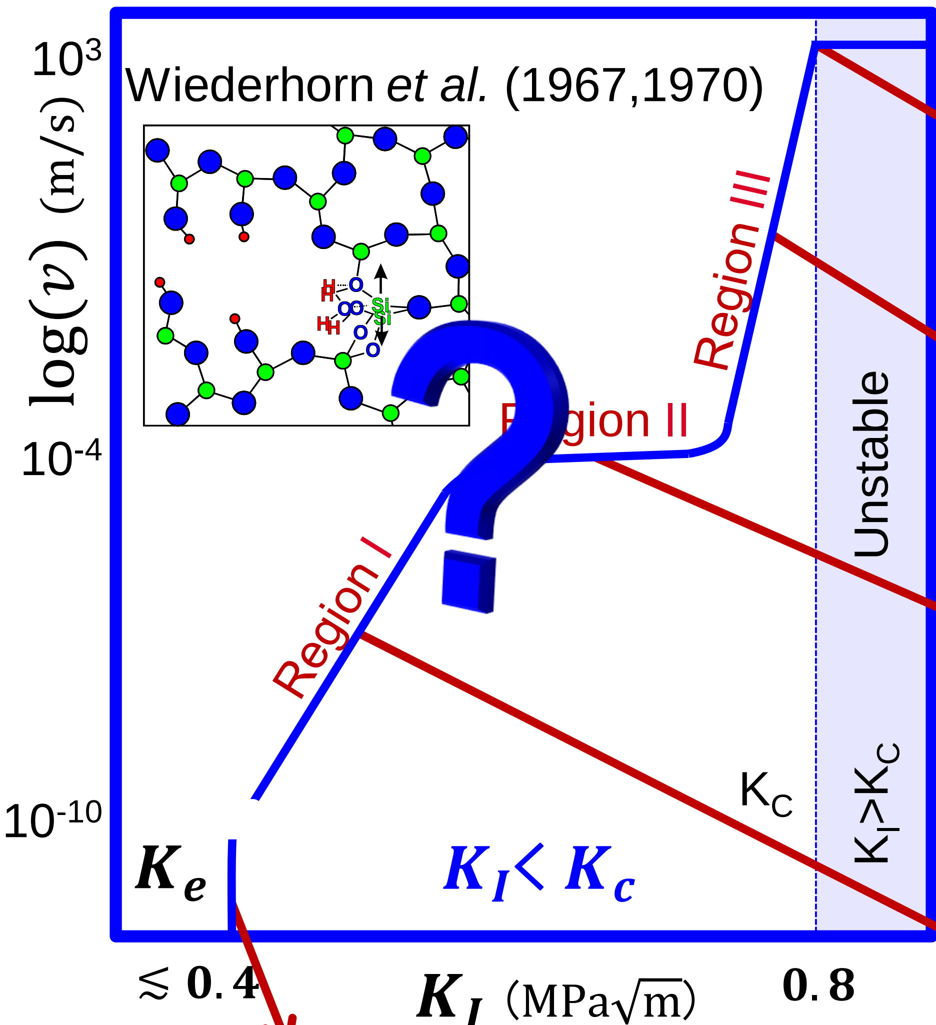
Propagation of the crack front

$K_I > K_{Ic}$ Unstable (μs)

K_{Ic} : **fracture toughness**

Lawn, *Fracture of Brittle Solids*,1993

SUB-CRITICAL CRACKING OF MATERIALS



$K_c = 0.8\text{MPa}\sqrt{\text{m}}$
(Limit in Vacuum)

Region III: The crack front propagates too fast for water molecules

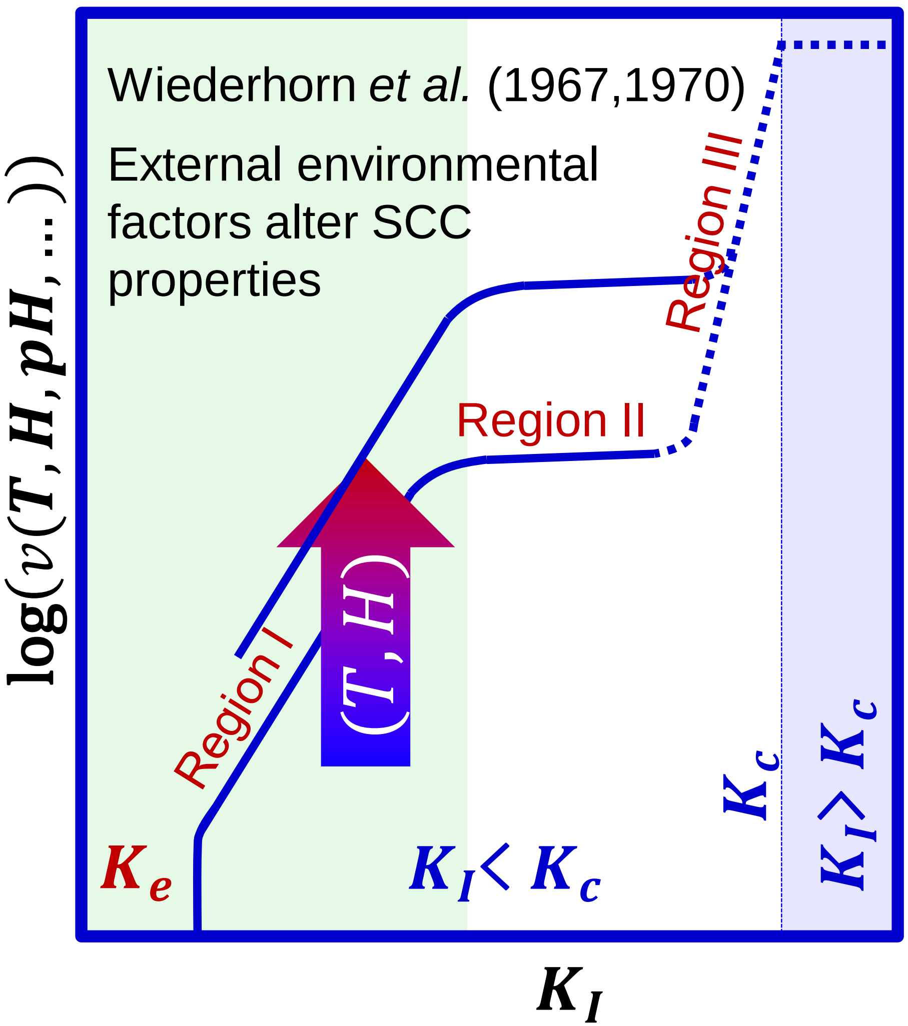
Region II: Limited by the time for the water to reach the crack tip

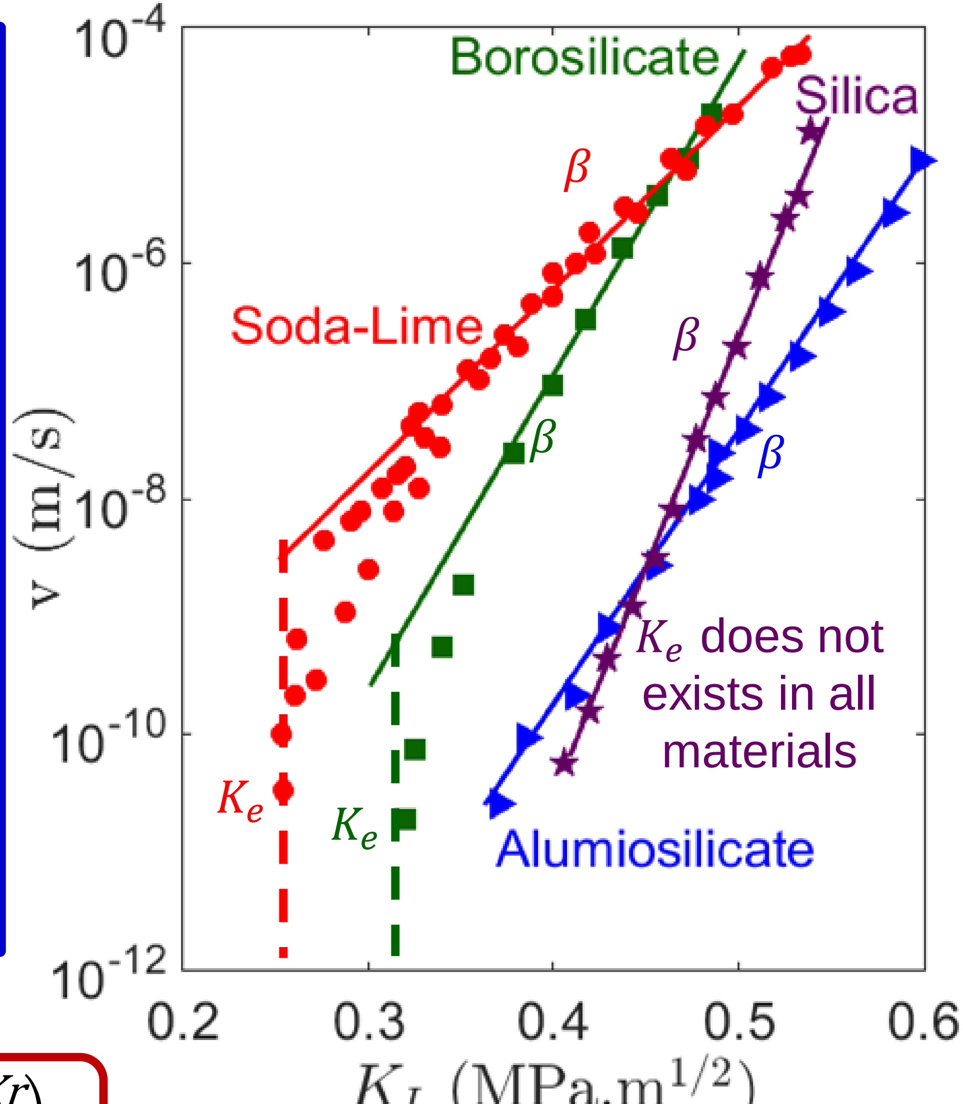
Region I: Controlled by the chemical reaction at the crack tip

$K_I > K_e \quad v = v_o(P^{H_2O})e^{\left(\frac{-E*+bKr}{RT}\right)}$

$K_e = 0.3 - 0.4\text{MPa}\sqrt{\text{m}}$
(static fatigue limit)

KNOWNS ABOUT STRESS CORROSION PROPERTIES

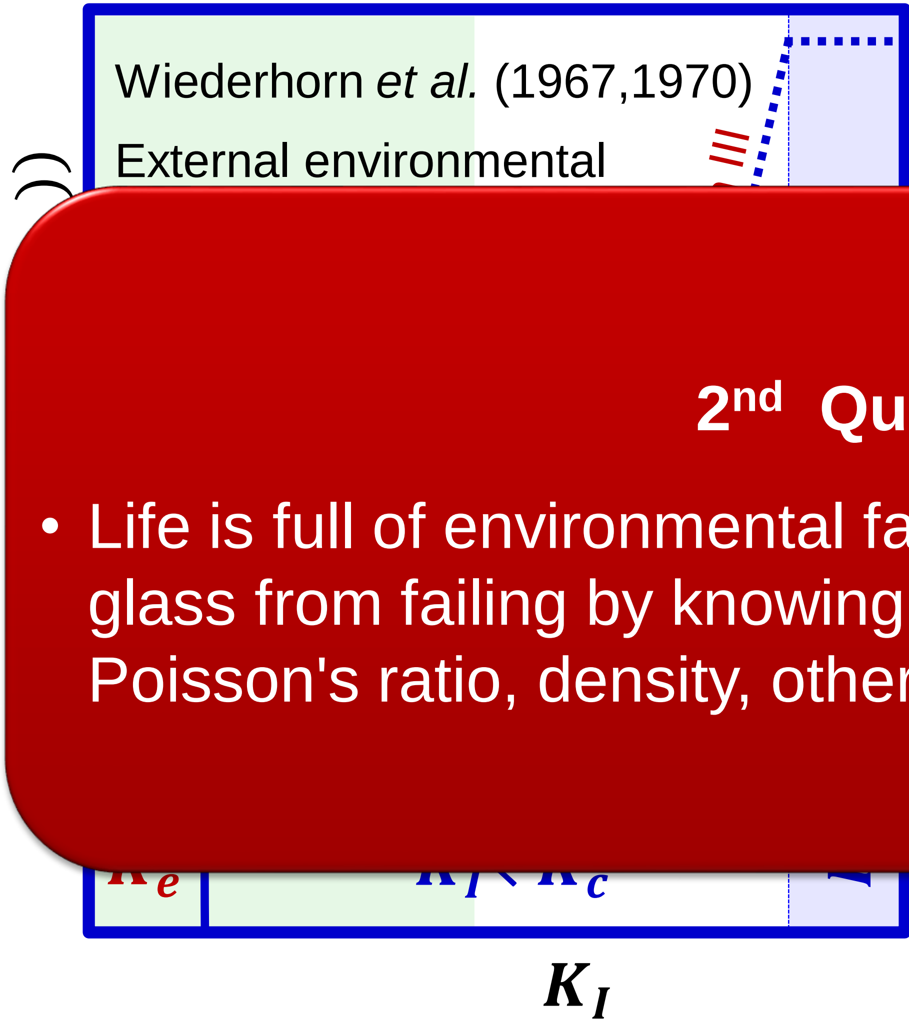


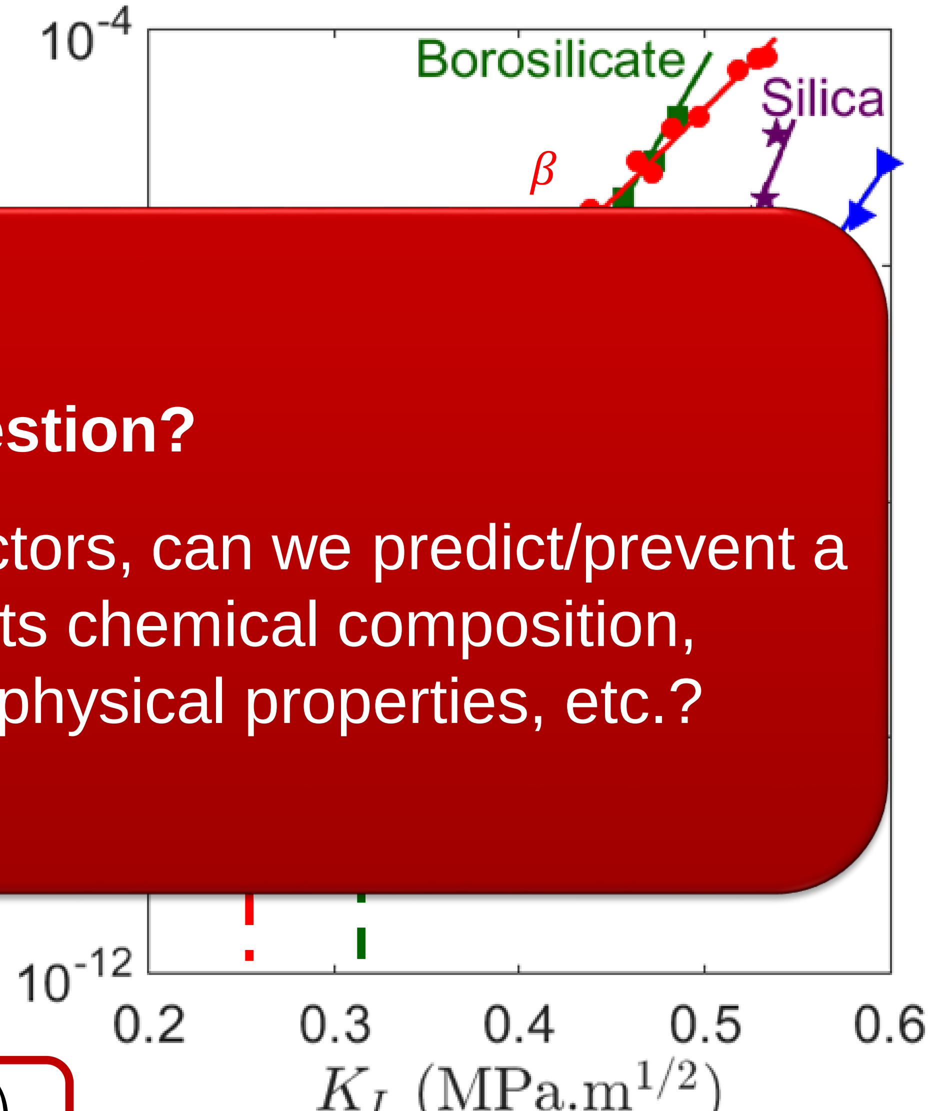


$$K_I > K_e \quad v = v_o(P^{H_2O})e^{\left(\frac{-E^* + bKr}{RT}\right)}$$

The Chemical Composition alters the SCC properties

KNOWNS ABOUT STRESS CORROSION PROPERTIES





2nd Question?

- Life is full of environmental factors, can we predict/prevent a glass from failing by knowing its chemical composition, Poisson's ratio, density, other physical properties, etc.?

$$K_I > K_e \quad v = v_o(P^{H_2O})e^{\left(\frac{-E^* + bKr}{RT}\right)}$$

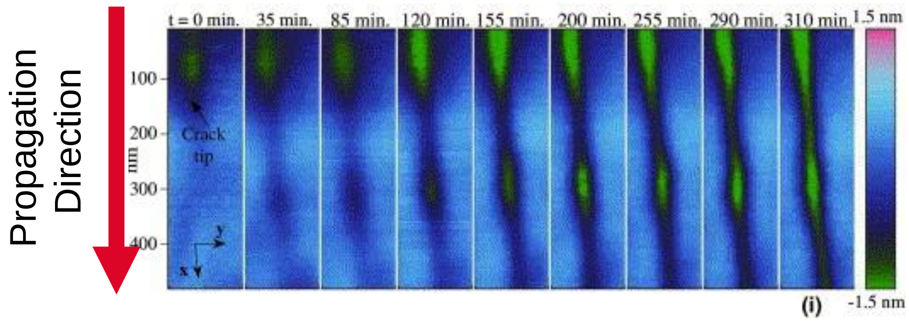
The Chemical Composition alters the SCC properties

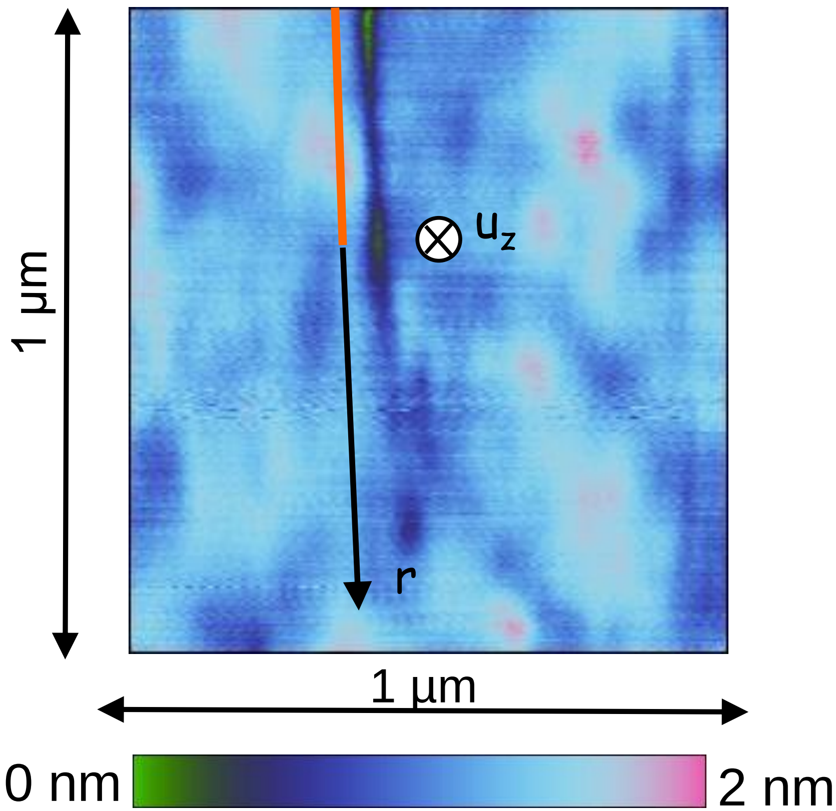
OUTLINE

The Questions?

- What is the size of the zone controlling the dynamics of crack propagation?
- Life is full of environmental factors, can we predict/prevent a glass from failing by knowing its chemical composition, Poisson's ratio, density, other physical properties, etc.?

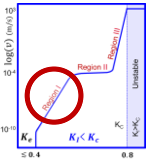
ATOMIC FORCE MICROSCOPIC IMAGING

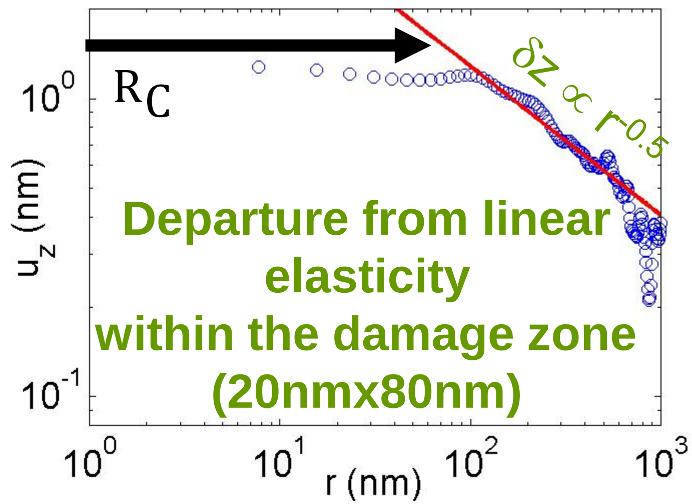


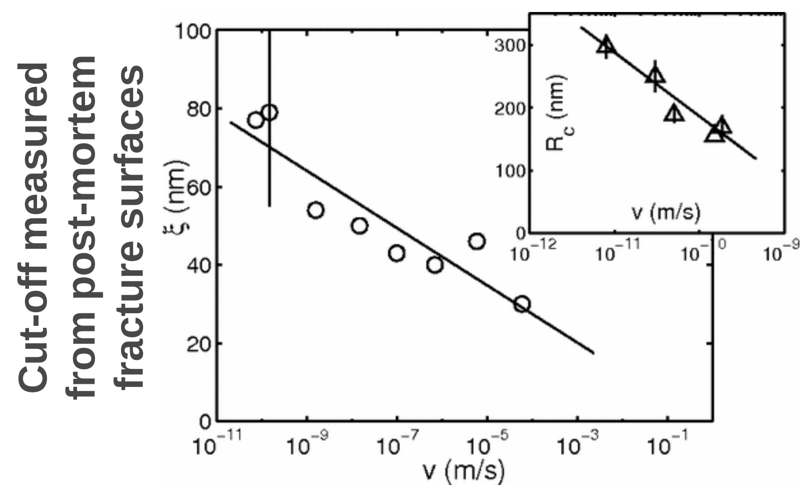


V=3 x 10^{-11} m/s

Linear elasticity :



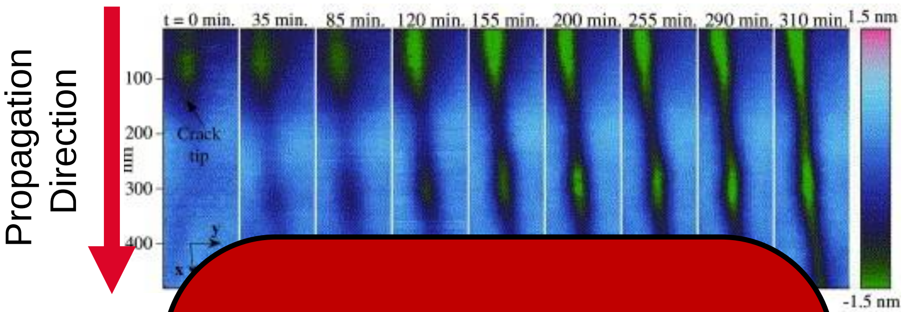




D. Bonamy, et. al. Phys. Rev. Lett. 135504

ATOMIC FORCE MICROSCOPIC IMAGING

DE LA RECHERCHE À L'INDUSTRIE



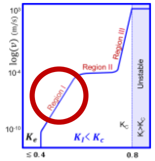
But Han et al pointed out that the 2D plane stress hypothesized is not relevant considering the scale at which the displacements are studied

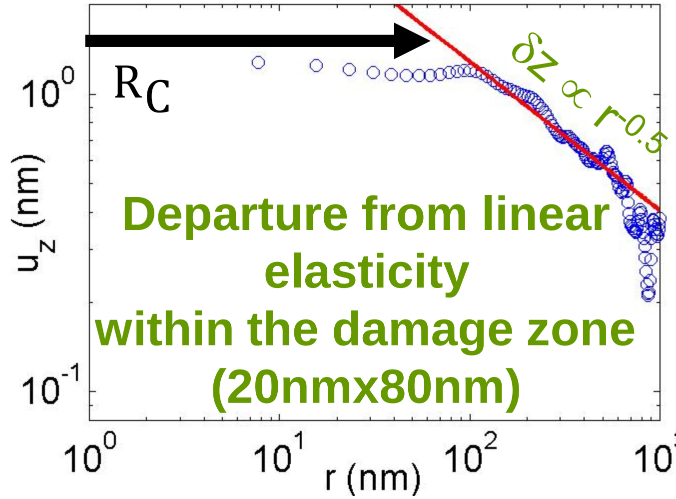
1 µm

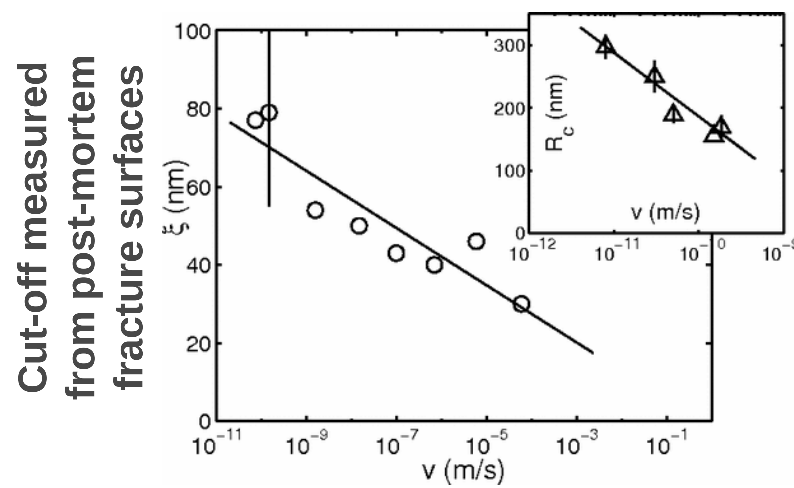
0 nm 2 nm

$V = 3 \times 10^{-11}$ m/s

Linear elasticity :







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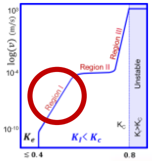
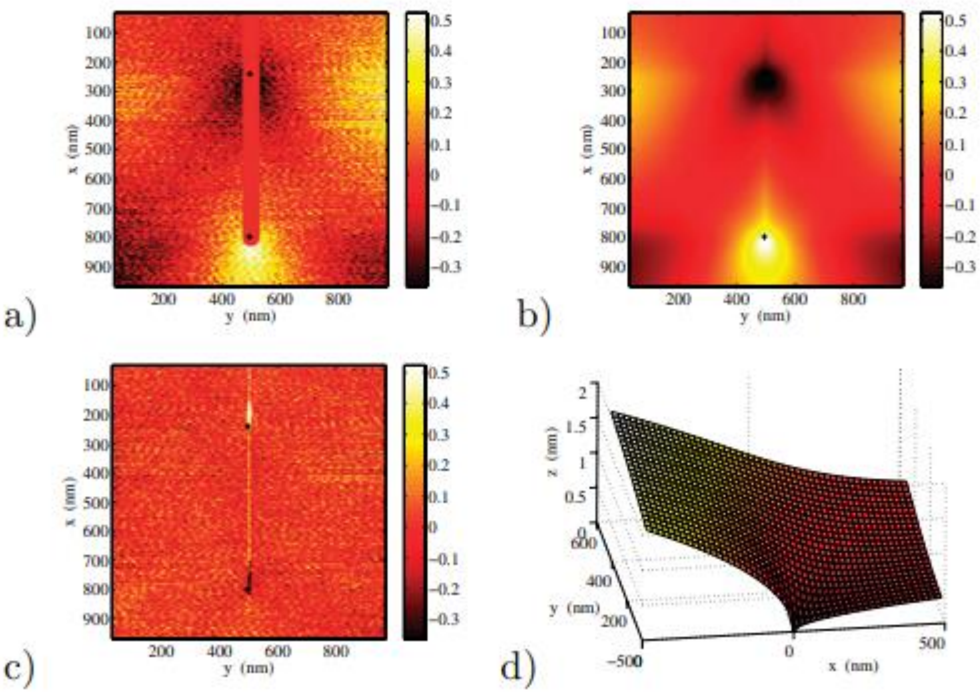


IDIC

Han, et. al. EPL. 66003

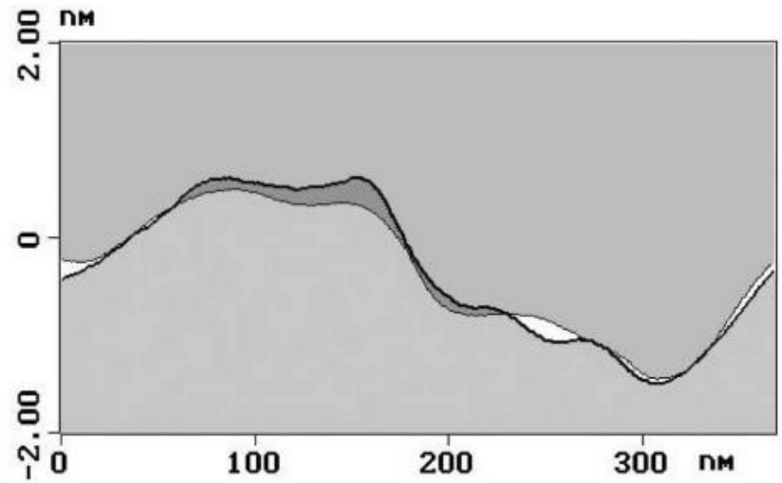


Applied IDIC techniques to AFM images of a moving crack

Process Zone $\lesssim$ 10nm

FRESTA

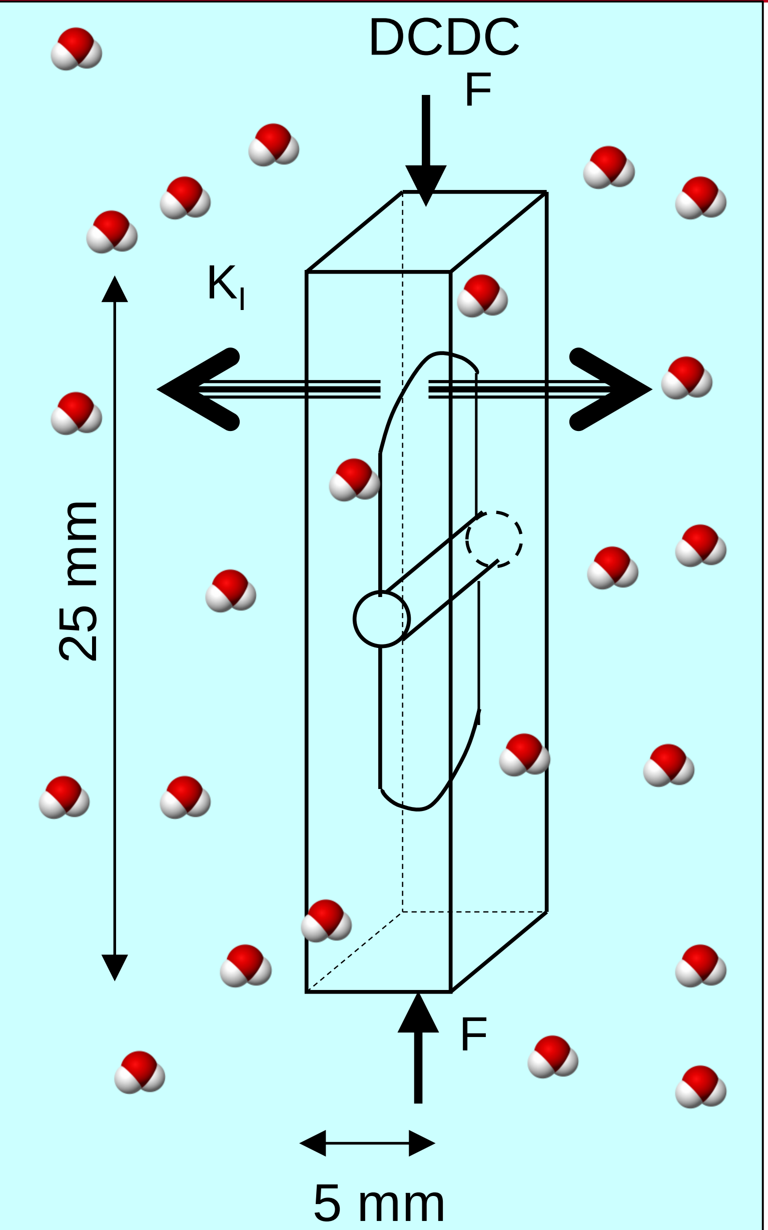
Guin, et. al. PRL. 92, 215502



Applied FRESTA techniques to post-mortem fracture surfaces

Surfaces match up to within 0.3 nm normal to the surface and 5 nm parallel to the surface

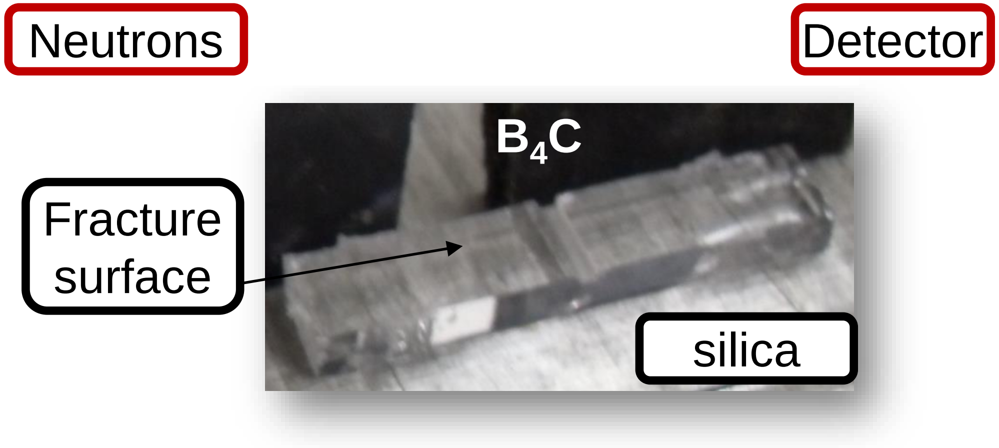
DEEP WATER PENETRATION IN SiO_2 DURING STRESS CORROSION FRACTURE



D_2O: Heavy water

Neutron scattering facility

EROS reflectometer at LLB



F. Lechenault et al. PRL 106:165504 (2011)
F. Lechenault et al. J. Phys.: Conf. Ser. 319:012005 (2011)

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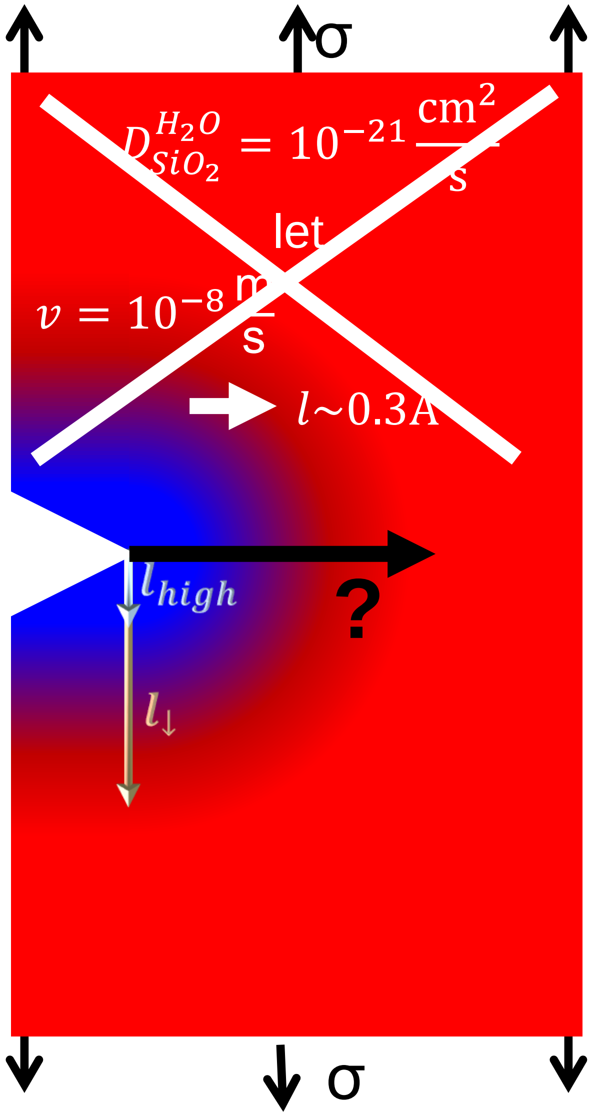
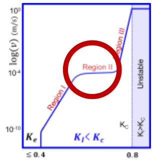




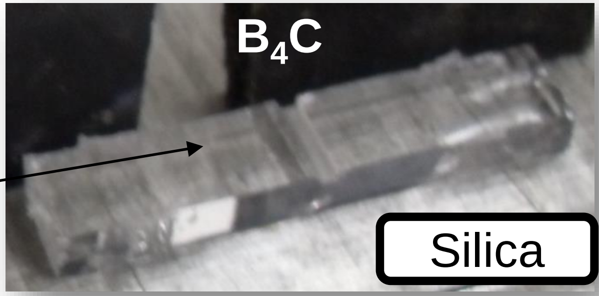
D_2O: Heavy water

Neutron scattering facility

EROS reflectometer at LLB

Neutrons

Detector

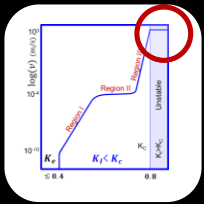


Heavy water was found up to **10 nm** below the fracture surface

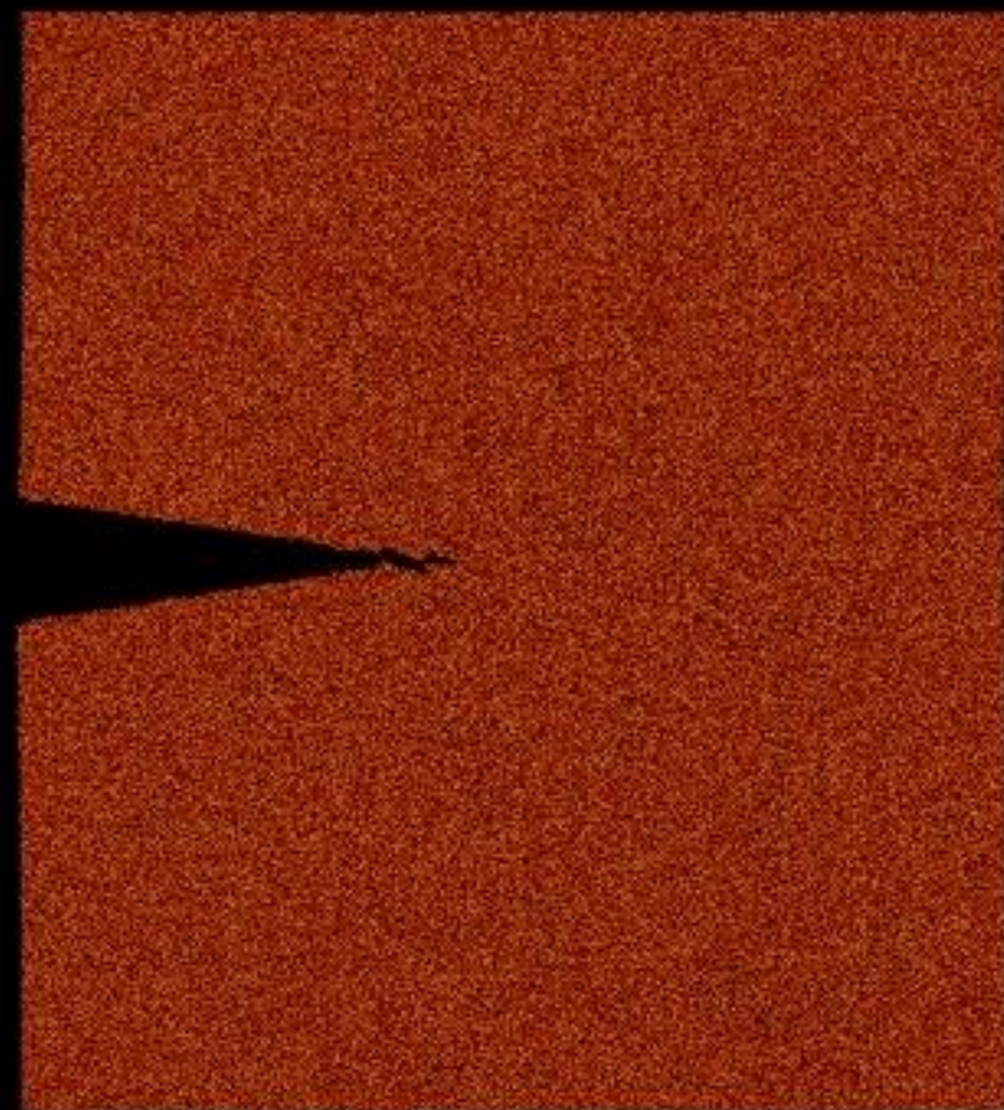
A high concentration was found in the first **4 nm**

F. Lechenault et al. PRL 106:165504 (2011)
F. Lechenault et al. J. Phys.: Conf. Ser. 319:012005 (2011)

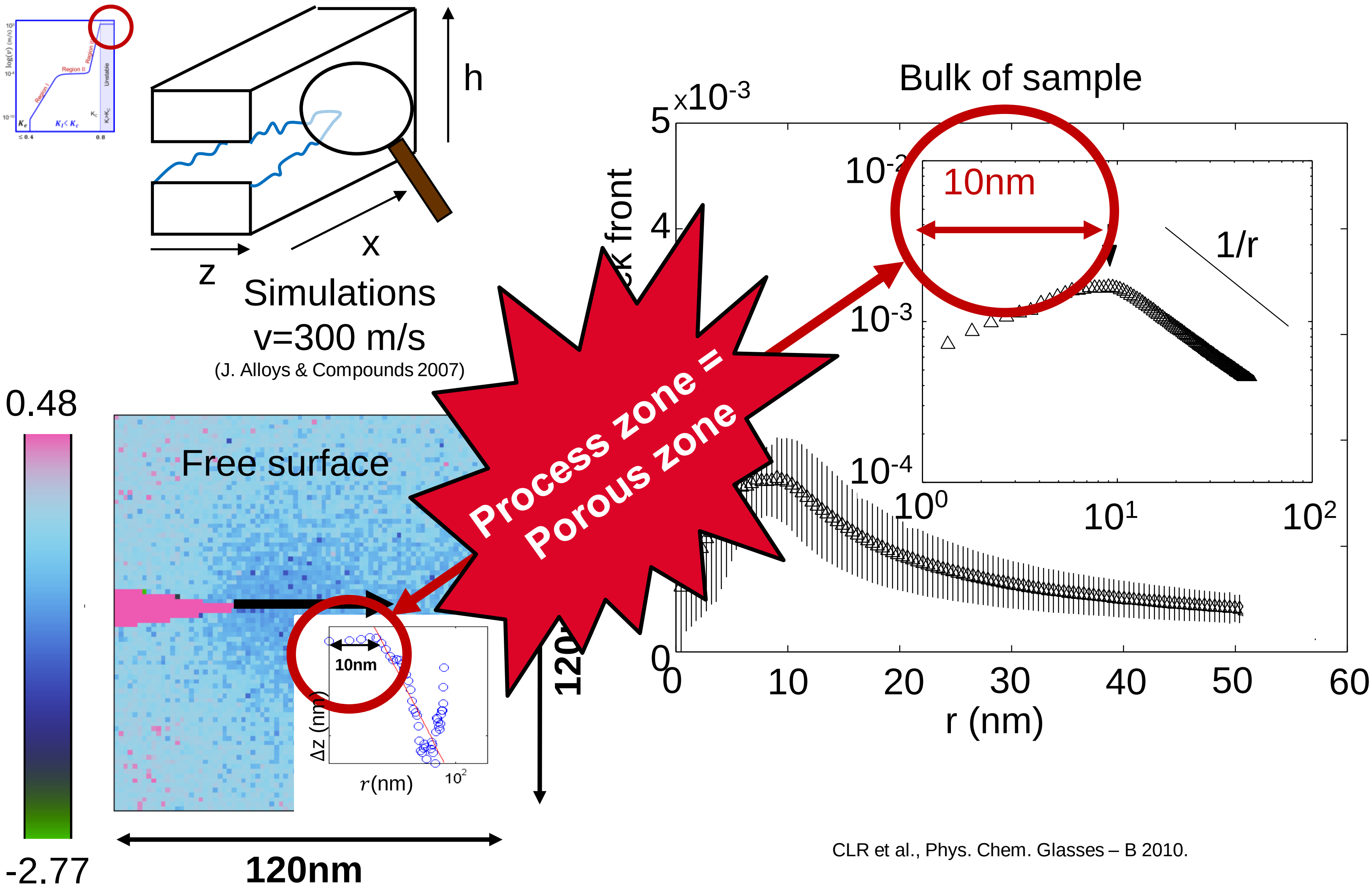
FRACTURE OF SILICA GLASSES: 15 MILLION ATOM SIMULATION



Pure Silica
System size
120nm×120nm×15nm
(CLR et al., J. Alloys & Compounds 2007)

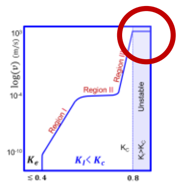


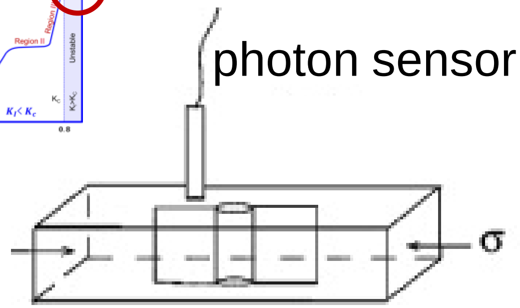
DAMAGE AHEAD OF THE CRACK TIP IN PURE SILICA



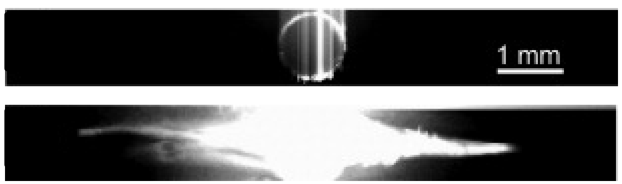
CLR et al., Phys. Chem. Glasses – B 2010.

DYNAMIC FRACTURE : A LUMINOUS VIEWPOINT

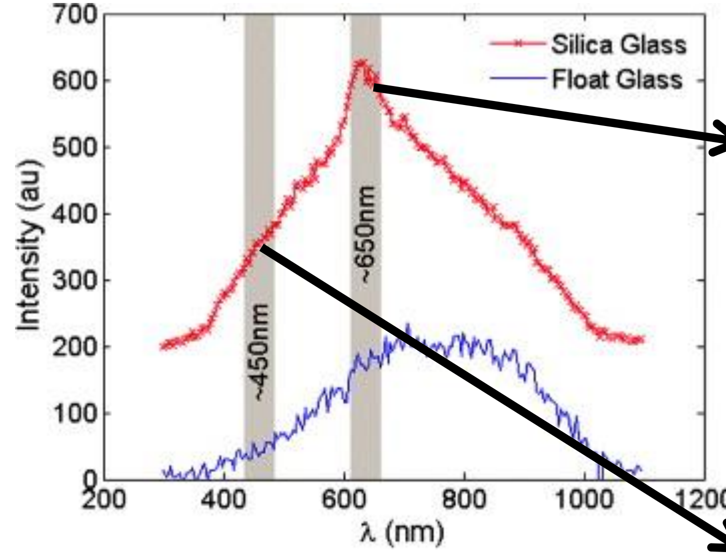




Midplane fracture

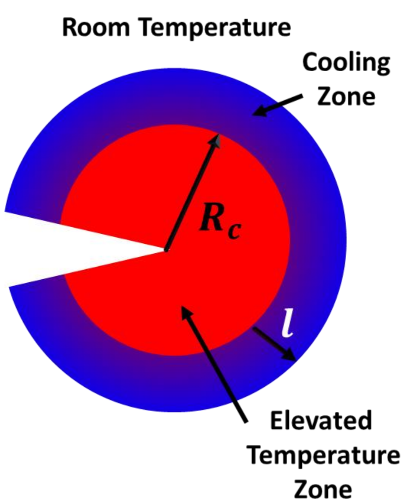


Black-body emission spectrum



Nonbridging oxygen hole center relaxation at 650 nm (~1.9 eV)

oxygen deficient center at 450 nm (~2.7 eV) (OH close to 1000 ppm)



Dynamic Fracture

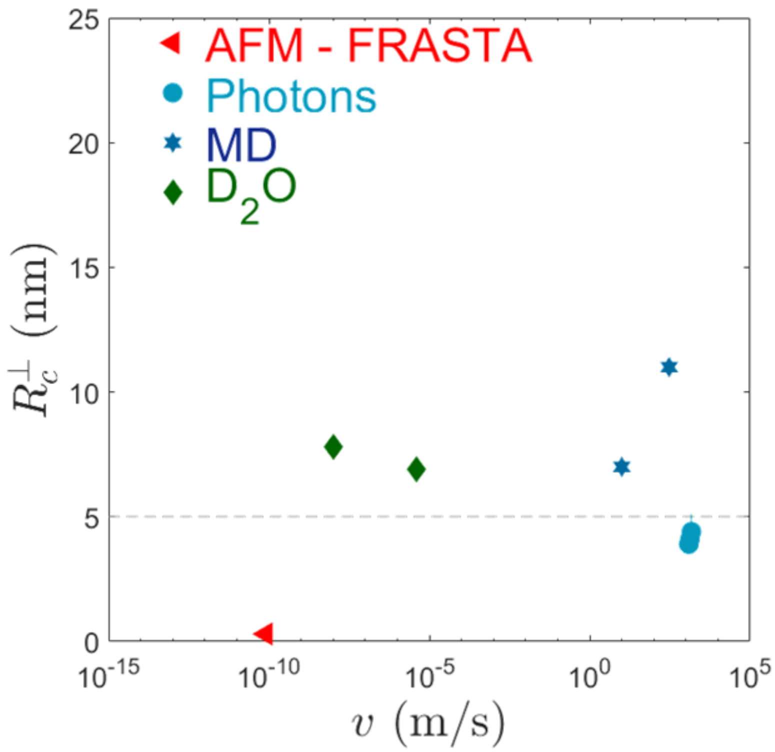
Sample	ΔT (K)	R_c (nm)	l (nm)
Silica 1	$4680_{\pm1[illegible]0}$	$3.9_{\pm0.2}$	$0.68_{\pm0.05}$
Silica 2	$5480_{\pm450}$	$4.4_{\pm0.7}$	$0.56_{\pm0.13}$
Silica 3	$4980_{\pm[illegible]10}$	$4.1_{\pm0.3}$	$0.63_{\pm0.07}$
Float 1	$5180_{\pm8[illegible]0}$	$4.1_{\pm1.3}$	$0.27_{\pm0.13}$
Float 2	$4100_{\pm530}$	$3.4_{\pm0.9}$	$0.36_{\pm0.12}$

Invoking
Planck distribution function
Rice and Levy - ΔT
Dugdale-Barrenblat - PZ

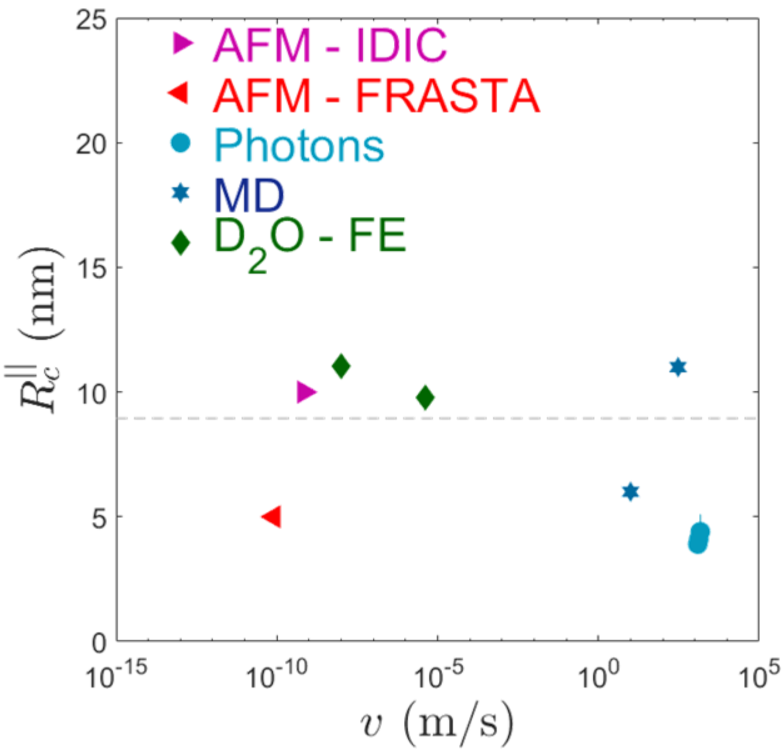
G. Pallares et al. EPL 99 28003. 2012

SUMMARY OF PROCESS ZONE SIZE IN SILICA

⊥ to the direction of propagation



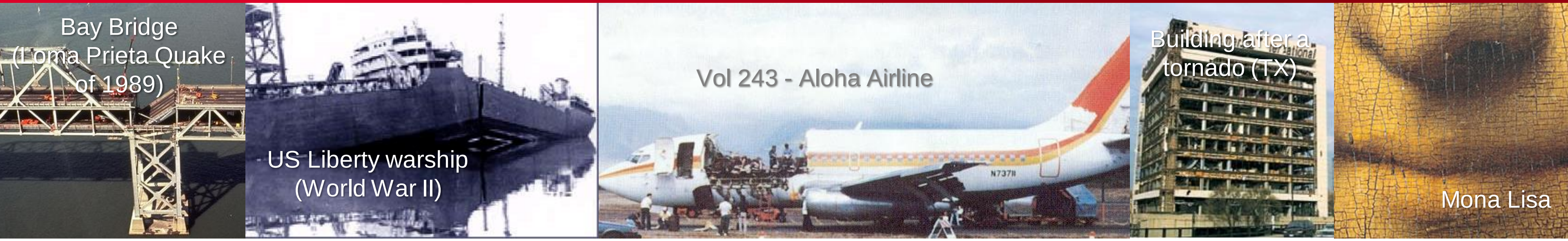
|| to the direction of propagation



Process Zone size 5-10nm in Silica
which is greater than a single bond length

Rountree and Feng. International Journal of Applied Glass. 2022

OUTLINE



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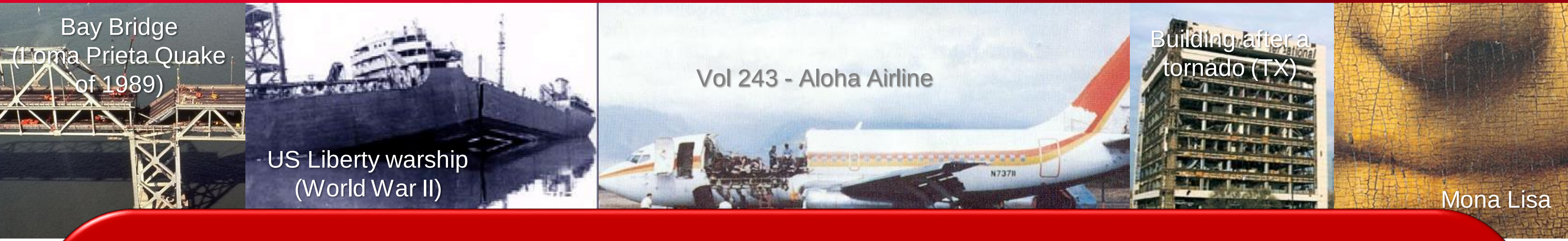
OUTLINE

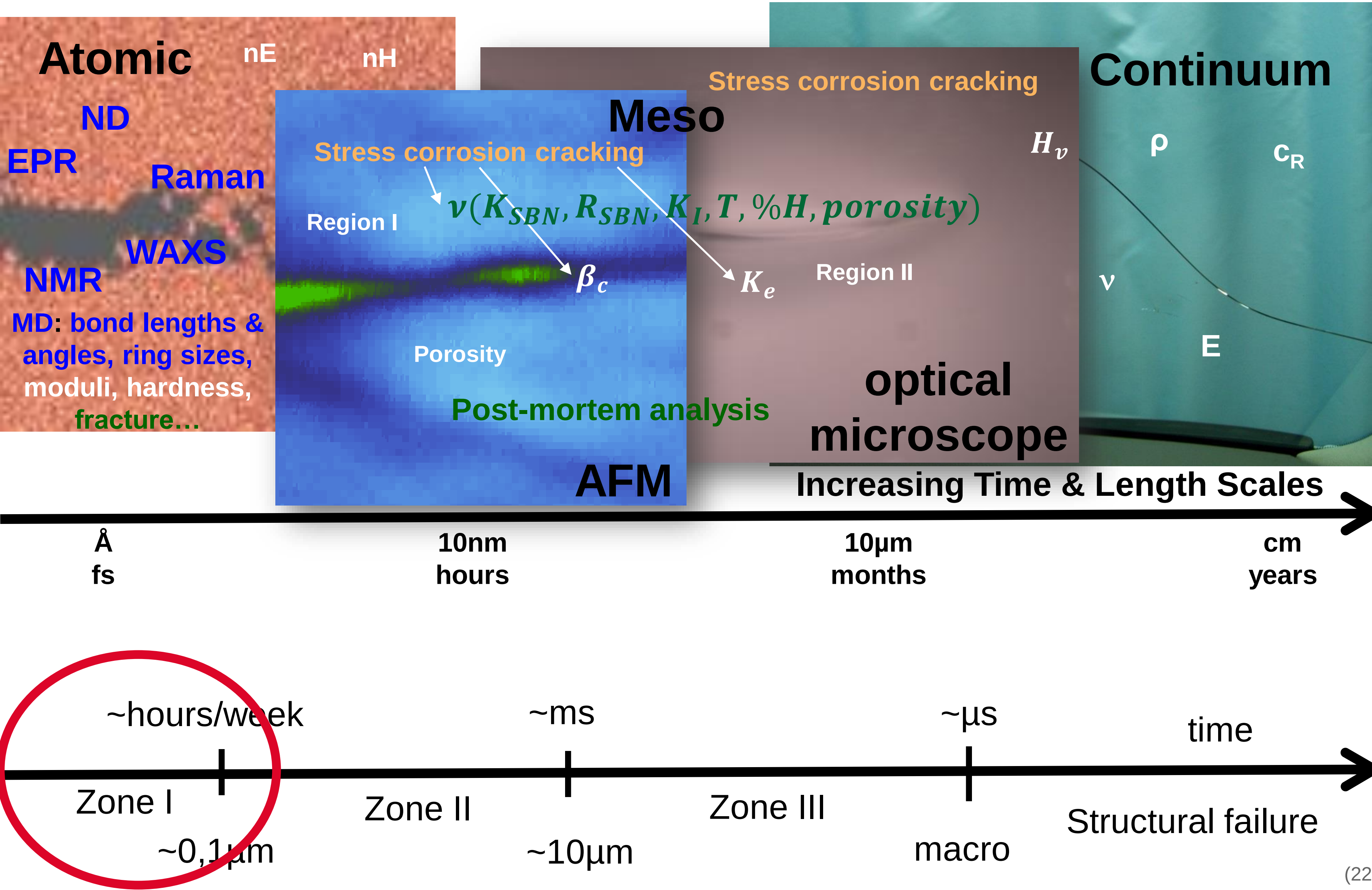


The Questions?

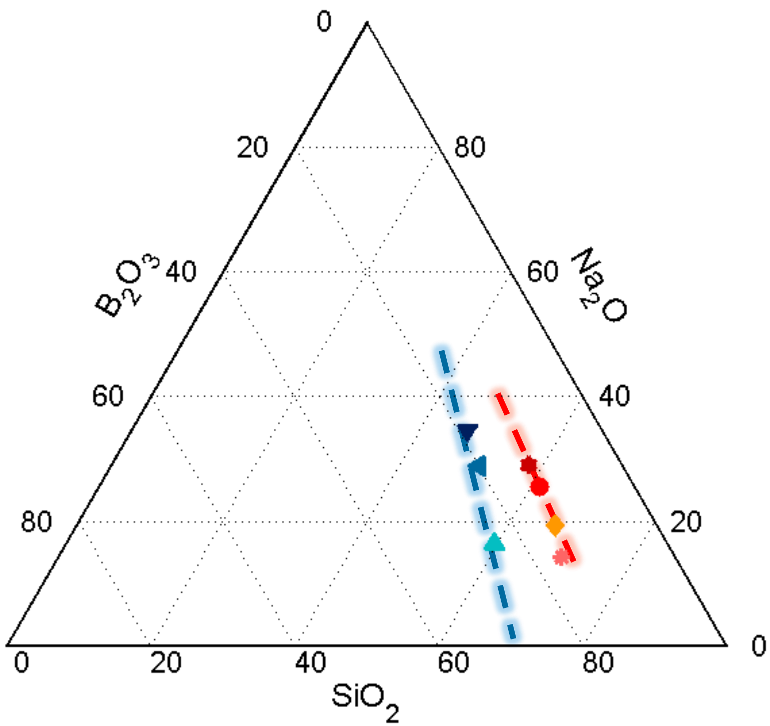
- What is the size of the zone controlling the dynamics of crack propagation?
- Life is full of environmental factors, can we predict/prevent a glass from failing by knowing its chemical composition, Poisson's ratio, density, other physical properties, etc.?

  - Mesoscale fluctuations to physical properties
  - Mesoscale fluctuations to fracture properties
- Conclusion

"STRUCTURAL FLUCTUATIONS" → STRUCTURAL FAILURE



THE 9 SiO_2-B_2O_3-Na_2O (SBN) GLASSES

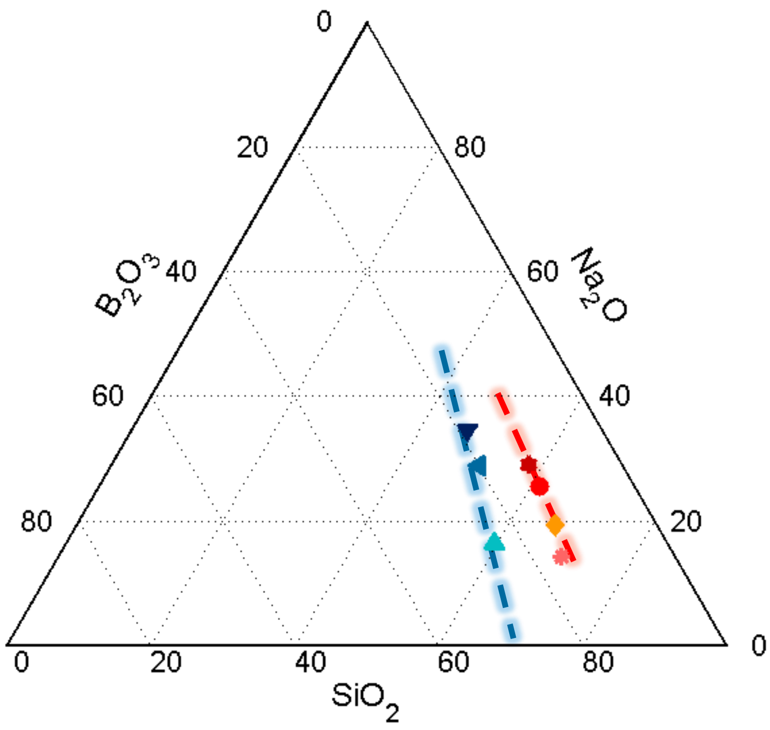


Why SBN Glasses?
Three major components of many industrial glasses!

Why changing the chemical composition?

	SiO_2	B_2O_3	Al_2O_3	NaO_2	K_2O	MgO	CaO
Silica	99.8						
Pyrex	81	13	0.04	2		0.05	0.05
Borosilicate	81	13	2	4			
Fiber wool	64	3	2.92	15.7	0.54	5.22	8.97

THE 9 SiO_2-B_2O_3-Na_2O (SBN) GLASSES



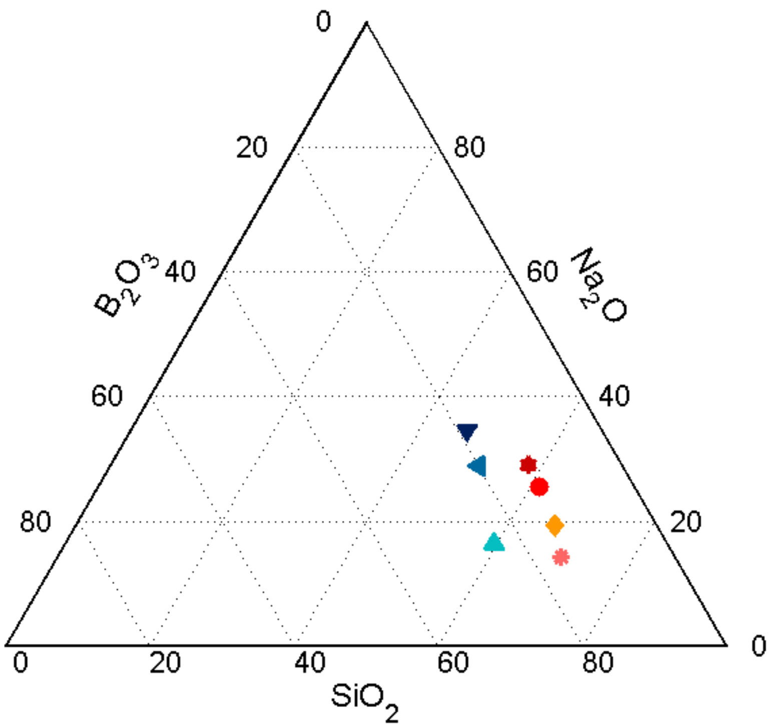
Why SBN Glasses?
Three major components of many industrial glasses!

Why changing the chemical composition?

$$R_{SBN} = \frac{[Na_2O]}{[B_2O_3]}$$

$$K_{SBN} = \frac{[SiO_2]}{[B_2O_3]} \begin{cases} K_{SBN} \sim 2.6 \\ K_{SBN} \sim 4.5 \end{cases}$$

THE 9 GLASSES



Not linear!

… & phase separation possible

JNCS 10.1016/j.jnoncrysol.2013.09.022

PhD candidate
M. Barlet

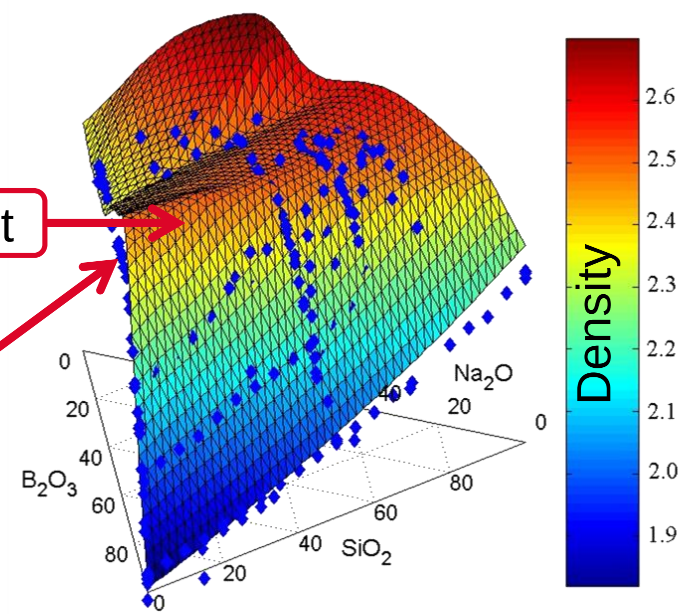
Collaborations:
A. Kerrache, J-M Delaye

Why change the chemical composition?

Δ chemical composition
→ Δ atomic & midrange order
→ Δ physical and mechanical

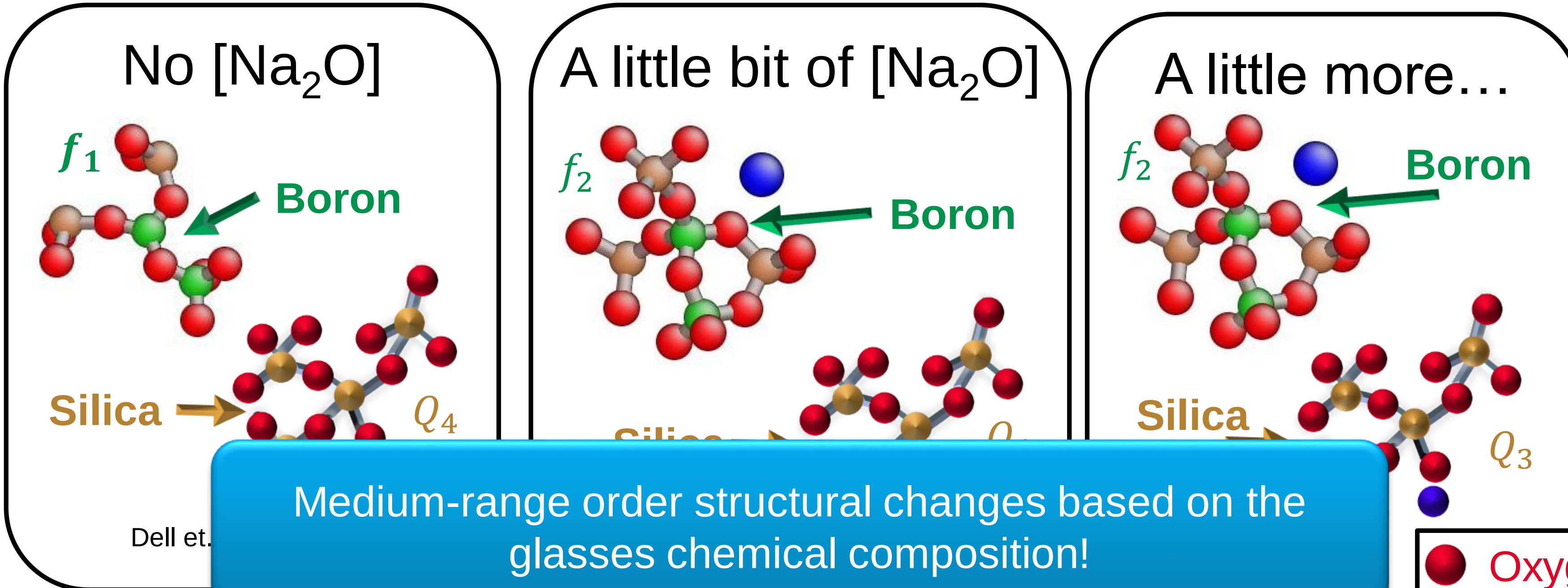
Inoue fit

Data

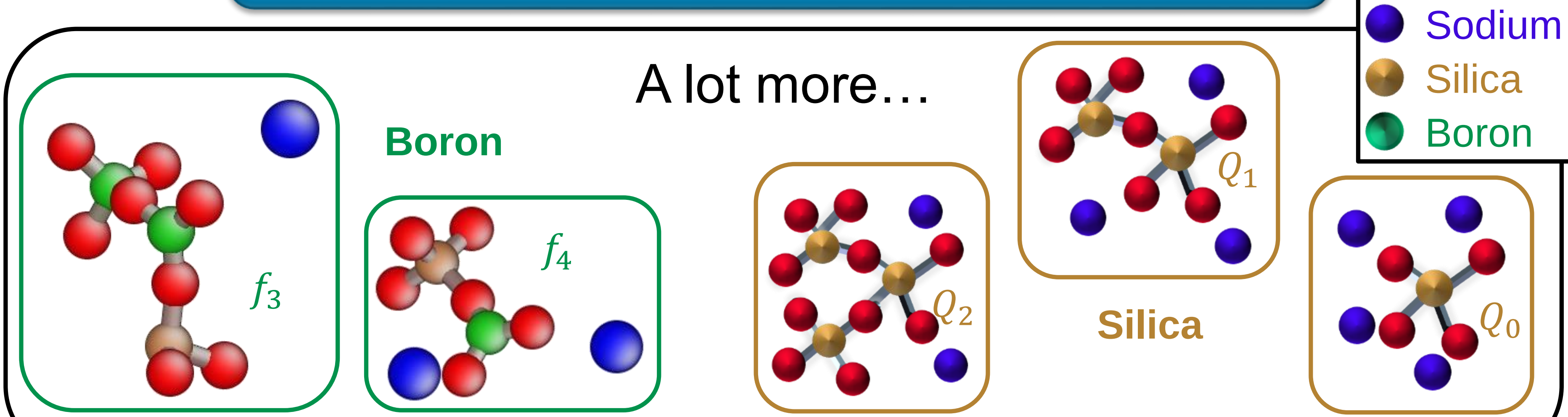


Inoue et. al. JACS. 2012.

GLASS STRUCTURE – ELEMENTARY UNITS



Medium-range order structural changes based on the glasses chemical composition!

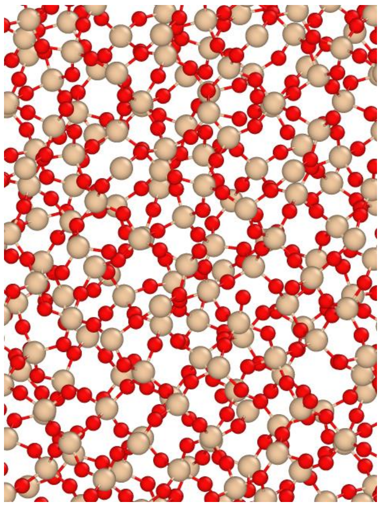


M. Barlet et al, JNCS 10.1016/j.jnoncrysol.2013.09.022

NUMERATION OF THE RETICULATION LEVEL

SiO_2

- 100% SiO_2



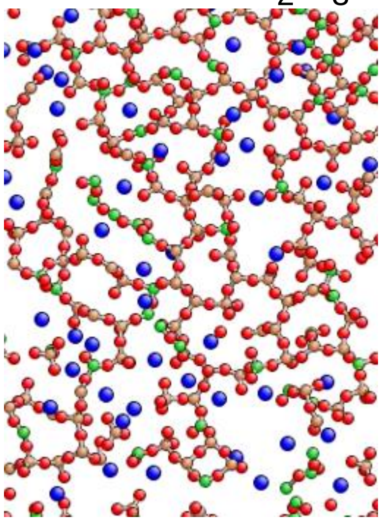
Molecular Dynamics simulations provide a count of species of bonds.

Most scientist count the number of elementary units

Here the oxygen atoms are given more importance

SBN14

- 67.73% SiO_2
- 14.20% Na_2O
- 18.04% B_2O_3



Degree of Polymerization (DP) at the atomic level

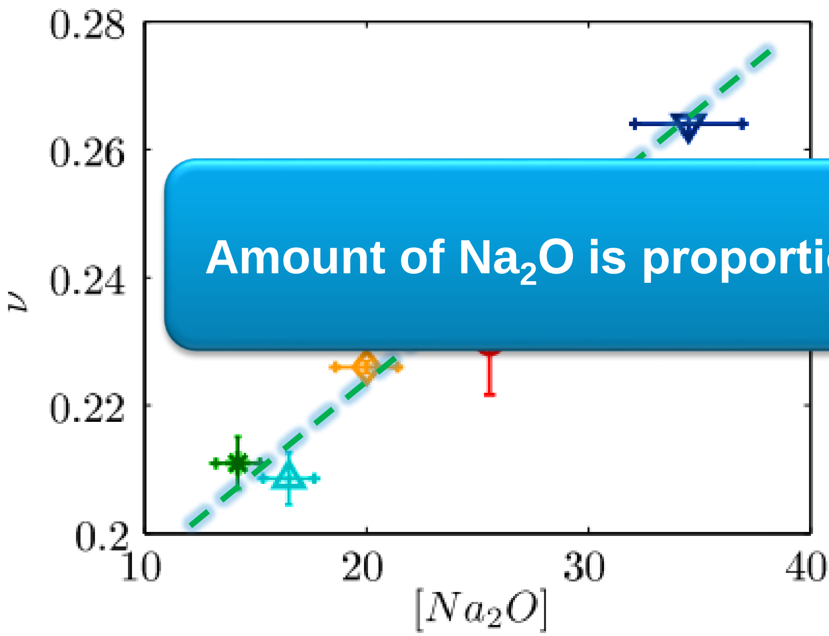
$$DP_{total} = \frac{2\ SiSi + 2\ SiB + 2\ BB + 3\ SiSiSi + 3\ SiSiB + 3\ SiBB + 3\ BBB}{Si + B + 2\ SiSi + 2\ SiB + 2\ BB + 3\ SiSiSi + 3\ SiSiB + 3\ SiBB + 3\ BBB}$$

M. Barlet et al. JNCS
doi: 10.1016/j.jnoncrysol.2016.07.017

PHYSICAL PROPERTIES OF SBN GLASSES

PhD candidate M. Barlet

Correlation with the Poisson's ratio



Amount of Na_2O is proportional to the Poisson's ratio.

Poisson's ratio is proportional to the sodium content

$[Na_2O]$

Compact

D ization

modifier

Free space

Reticulate

Na^+ charge compensator

↑ $[Na_2O]$ ⇒ ↓ Depolymerization

↓ Dimensionality ⇒ ↑ Poisson's ratio

Rouxel, *PRL*, 2008

M. Barlet et al, JNCS 10.1016/j.jnoncrysol.2015.02.005

PHYSICAL PROPERTIES OF SBN GLASSES

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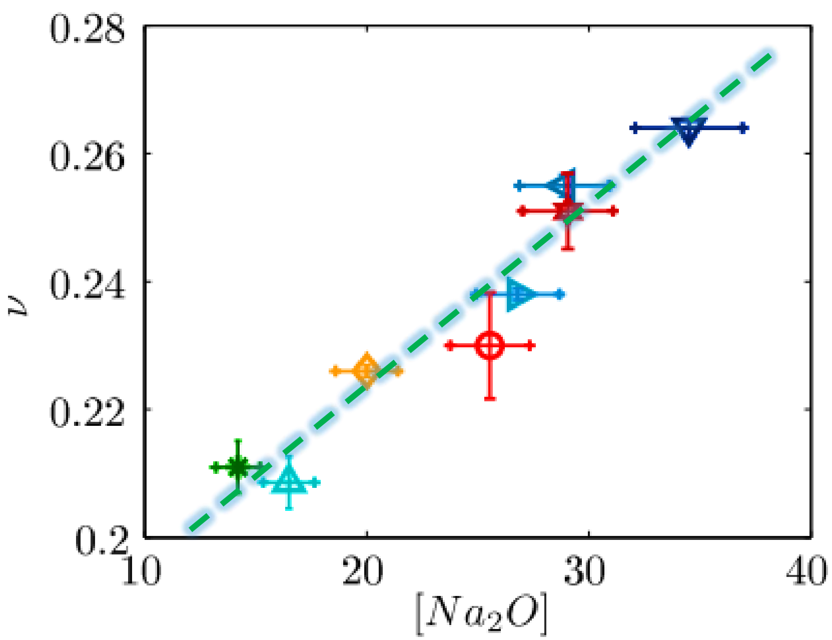


Poisson's ratio is proportional to the sodium content

$[Na_2O]$

Compact

Depolymerization

Na^+ network modifier

Free space

Reticulate

Na^+ charge compensator

↑ $[Na_2O]$ ⇒ ↓ Depolymerization

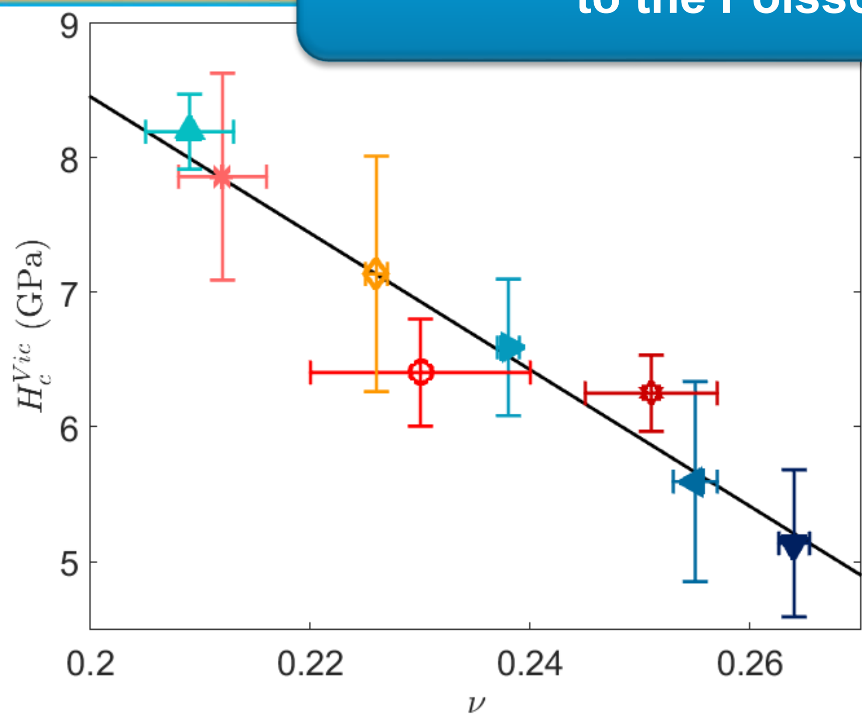
↓ Dimensionality ⇒ ↑ Poisson's ratio

Rouxel, *PRL*, 2008

M. Barlet et al, JNCS 10.1016/j.jnoncrysol.2015.02.005

EVOLUTION OF H_V

Hardness SBN glasses is proportional to the Poisson's ratio (or [Na_2O]).

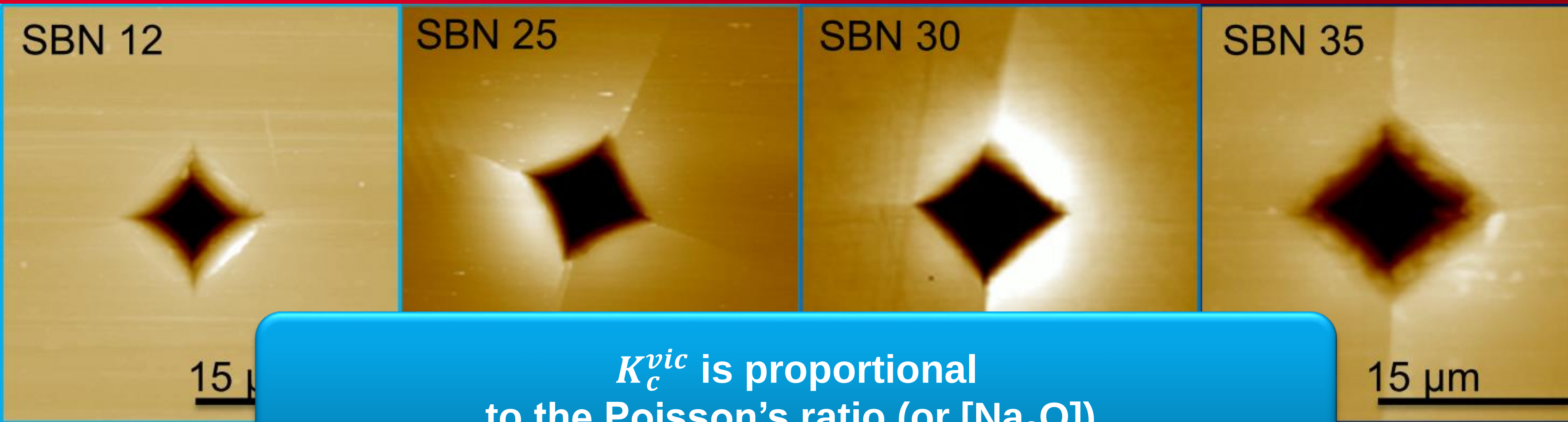


$$H_V^{vic} = \frac{1.8544 * F}{d_i}$$

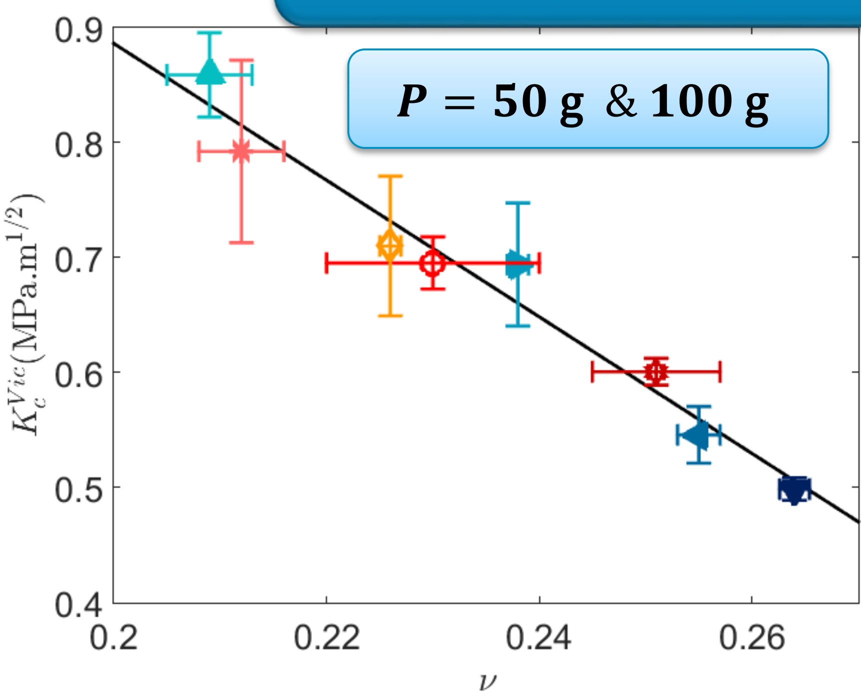
- H_v↓ for ↑ [Na_2O] for constant K_{SBN}
- ↑ [Na_2O] indenter penetrates deeper

Barlet et. al. *J. Non-Crys.* Solids 417-418:66-79 (2015).

EVOLUTION OF K_c^{vic}



K_c^{vic} is proportional to the Poisson's ratio (or $[Na_2O]$).



$P = 50\ \mathrm{g}\ \&\ 100\ \mathrm{g}$

$$K_c^{Vic} = 0.022 \left(\frac{E}{H^{Vic}}\right)^{0.4} \frac{P}{c^{1.5}}$$

$K_c^{vic} \downarrow$ for $\uparrow \nu$

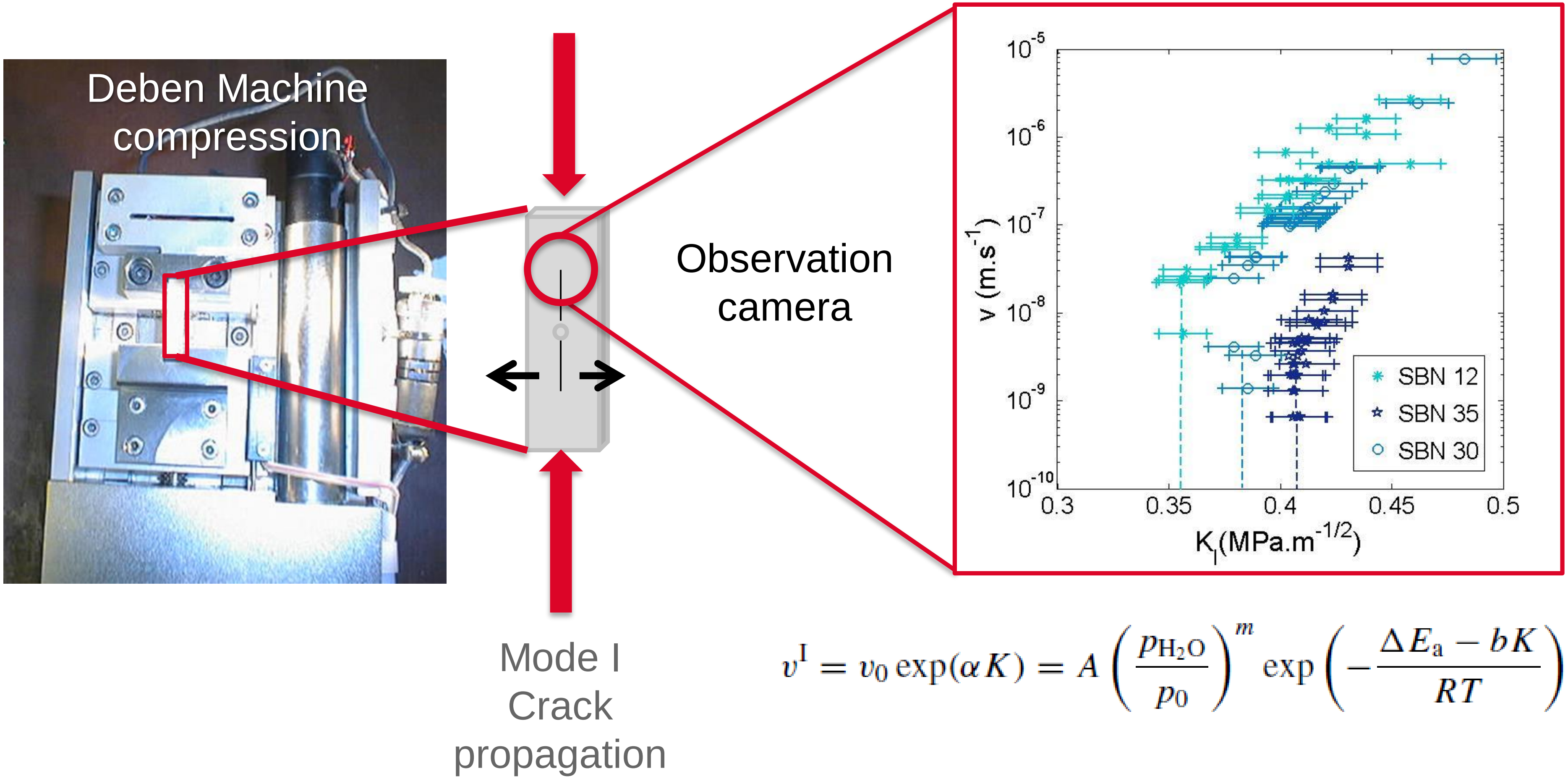
Barlet et. al. *J. Non-Crys.* Solids 417-418:66-79 (2015).

$P = 50\ \mathrm{g} \rightarrow 0.49\ \mathrm{N}$

STRESS CORROSION CRACKING

[Na_2O]

16.5 → 28.9 → 34.5



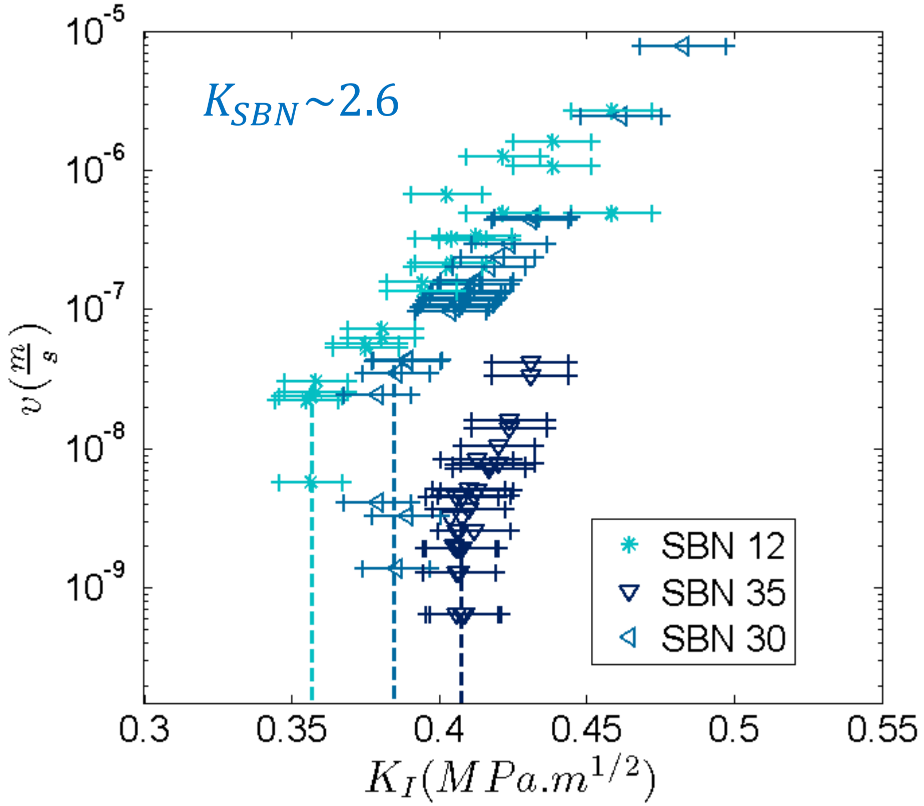
$$v^{\mathrm{I}} = v_0 \exp(\alpha K) = A \left(\frac{p_{H_2O}}{p_0}\right)^m \exp\left(-\frac{\Delta E_a - bK}{RT}\right)$$

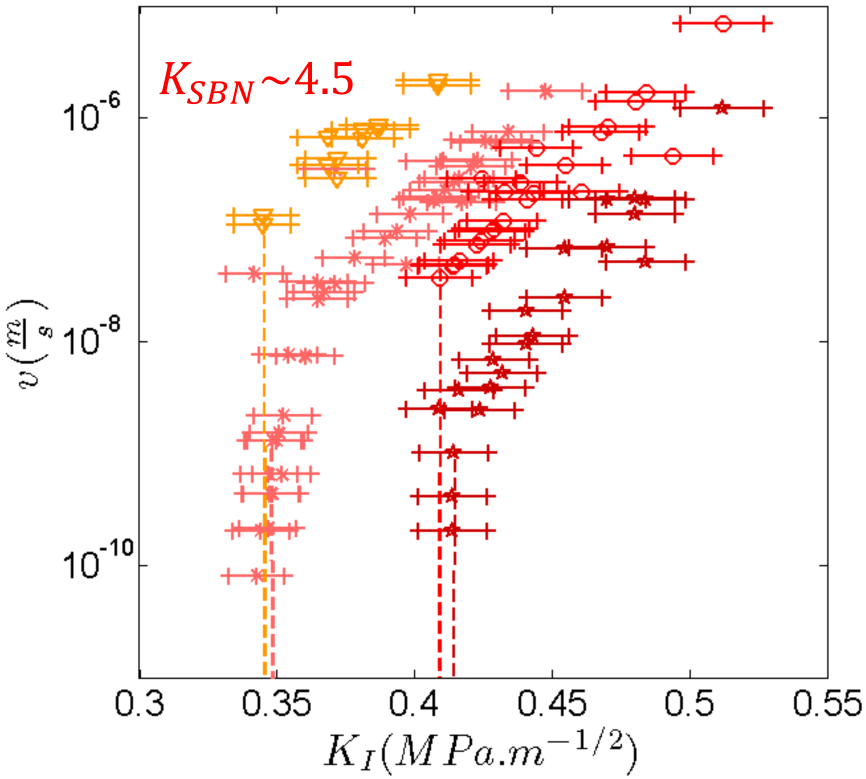
EVOLUTION OF SCC PROPERTIES

$$K_{SBN} = \frac{[SiO_2]}{[B_2O_3]}$$

$[Na_2O]$: 16.5 → 28.9 → 34.5

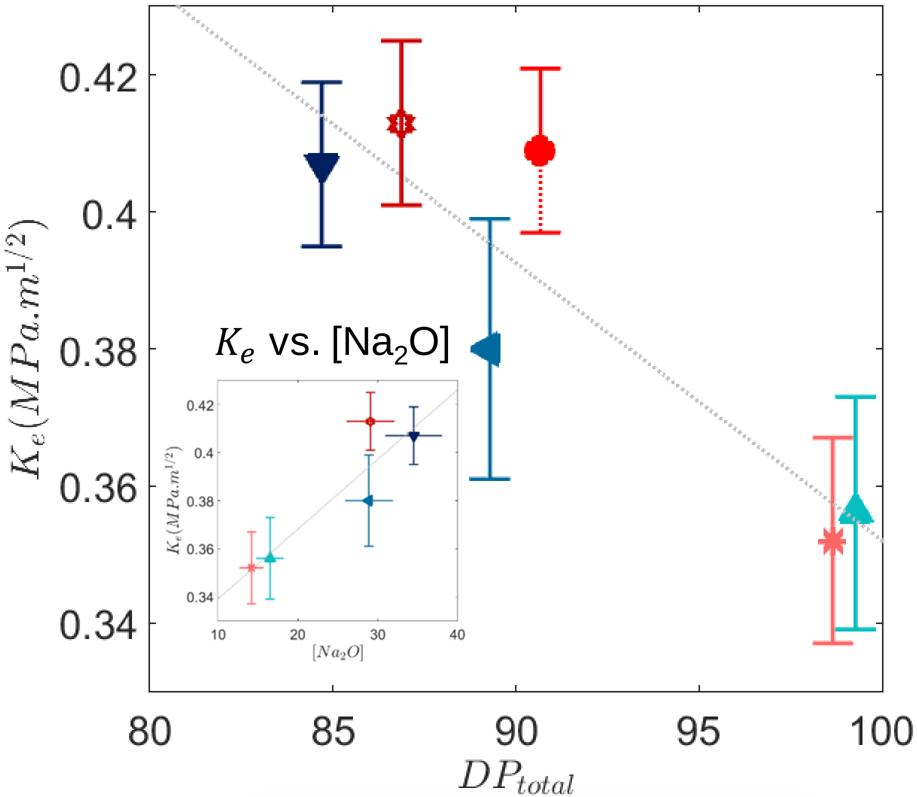
$[Na_2O]$: 14.2 → 19.2 → 25.5 → 29.1

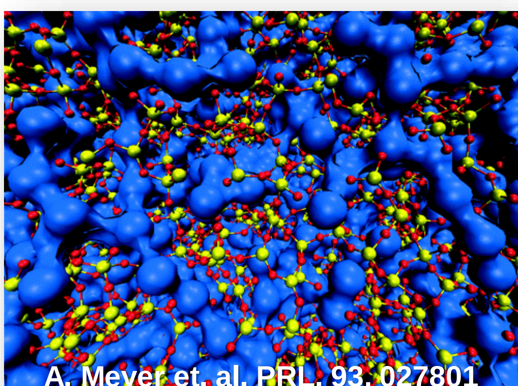




M. Barlet et al, JNCS doi: 10.1016/j.jnoncrysol.2016.07.017

EVOLUTION OF K_e





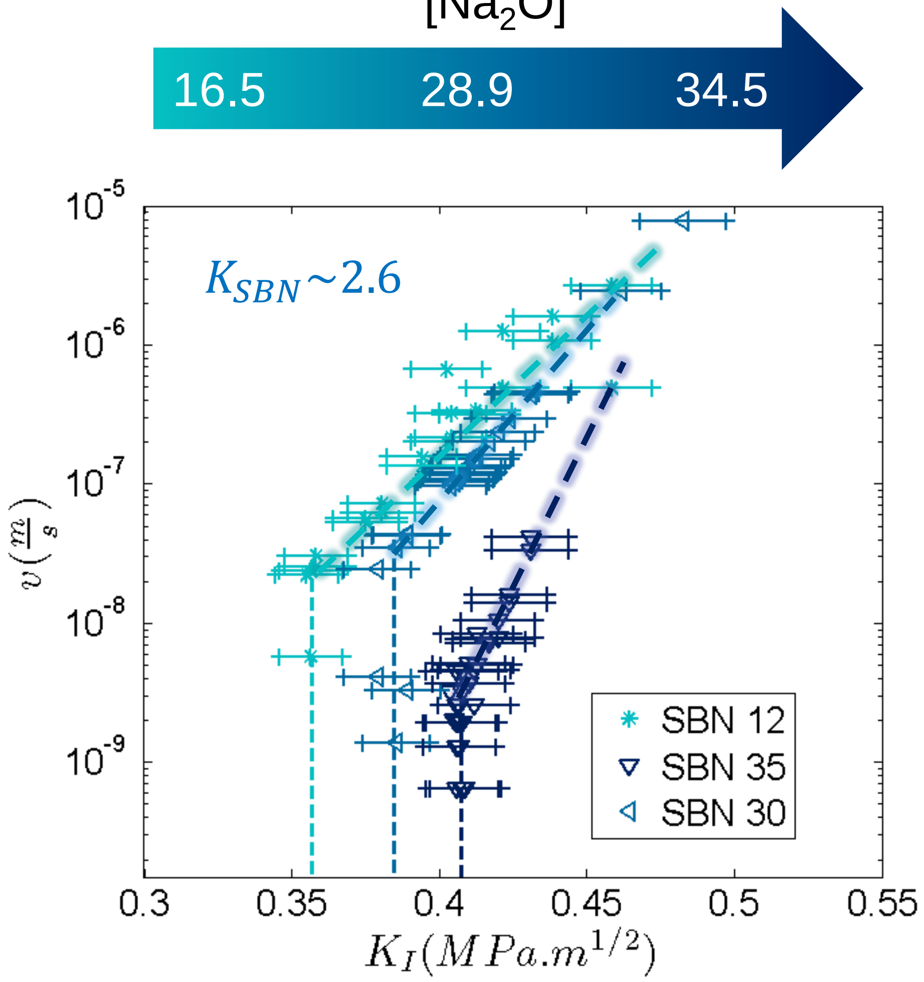
- K_{SBN}~constant:
  ↑ [Na_2O] → ↑ K_e
- [Na_2O] forms pockets or "channels" (Na^+ rich regions)
- Pockets acts as stress "sinks"
- Stresses are redistributed around these sinks and do not concentrate solely at the crack tip
- This delays the onset of crack propagation causing K_e to increase

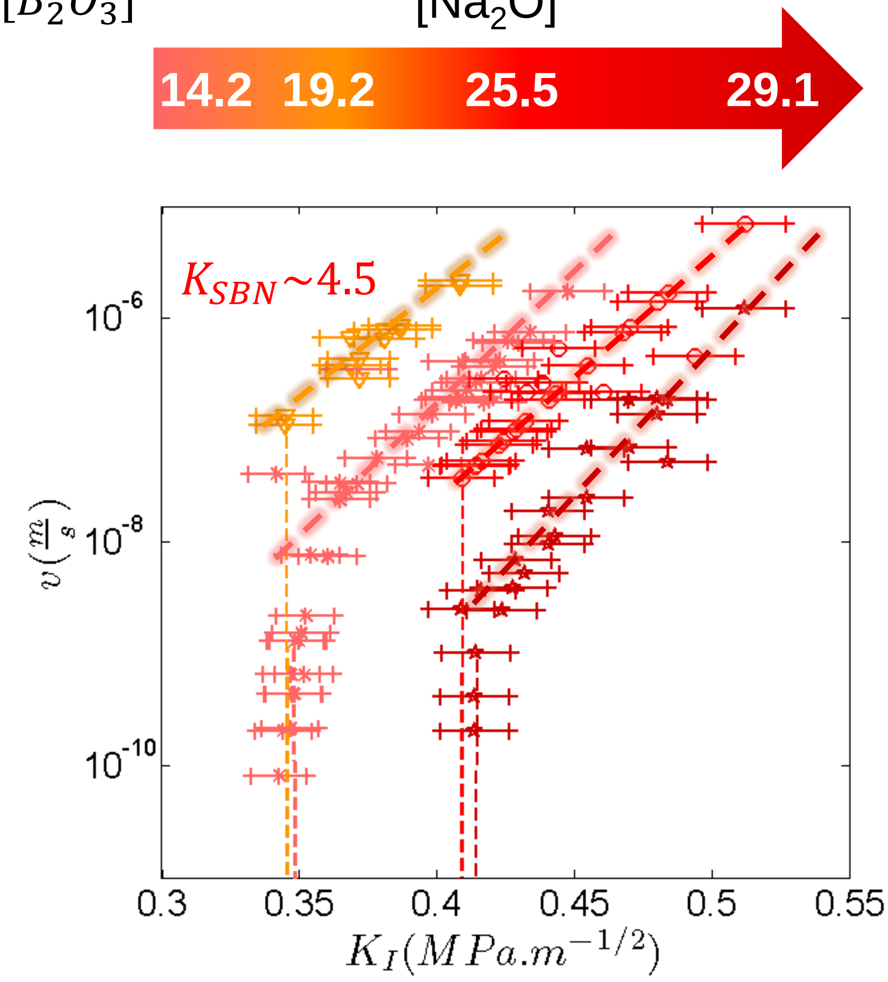
M. Barlet et al, JNCS doi: 10.1016/j.jnoncrysol.2016.07.017

DE LA RECHERCHE À L'INDUSTRIE

EVOLUTION OF FRACTURE PROPERTIES

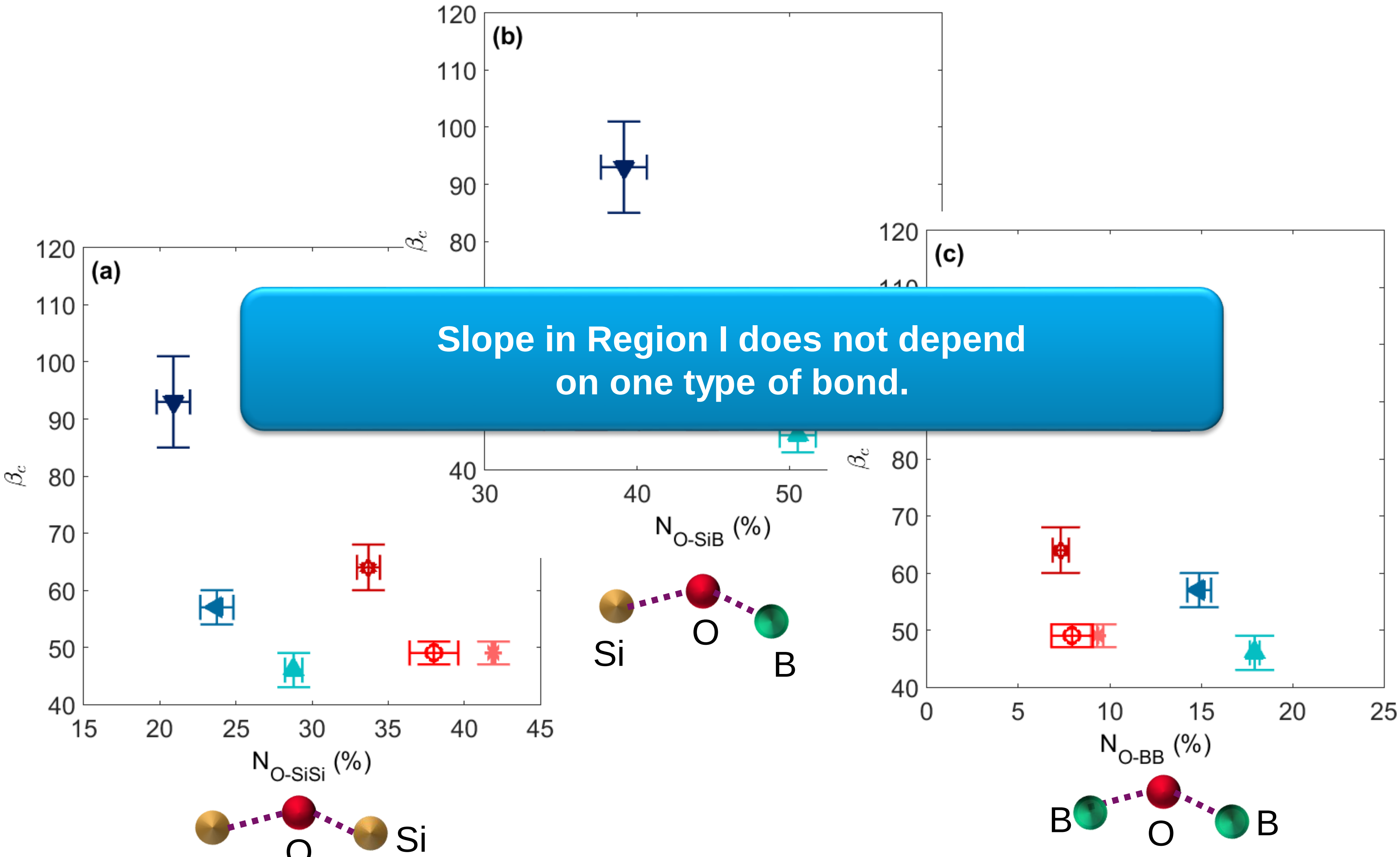
$$K_{SBN} = \frac{[SiO_2]}{[B_2O_3]}$$





M. Barlet et al, JNCS 2016. doi: 10.1016/j.jnoncrysol.2016.07.017

BOND TYPE & CRACK PROPAGATION



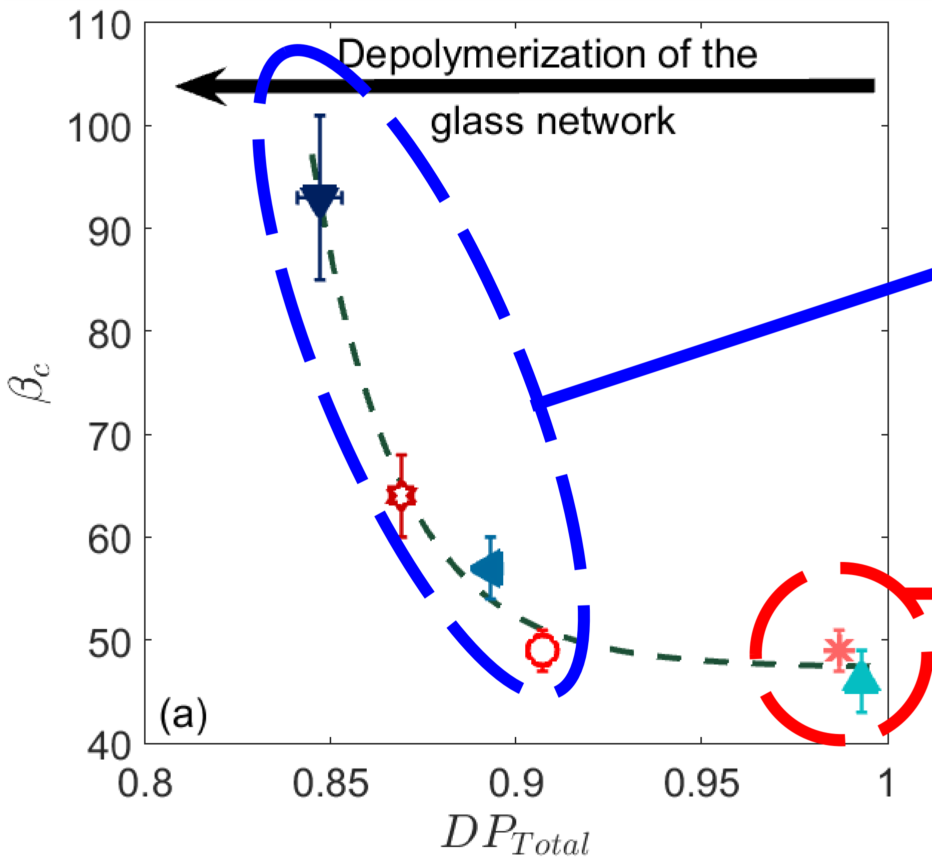
Slope in Region I does not depend on one type of bond.

M. Barlet et al, JNCS 2016. doi: 10.1016/j.jnoncrysol.2016.07.017

RETICULATION LEVEL & CRACK PROPAGATION

Degree of Polymerization (DP) at the atomic level

$$DP_{total} = \frac{2\ SiSi + 2\ SiB + 2\ BB + 3\ SiSiSi + 3\ SiSiB + 3\ SiBB + 3\ BBB}{Si + B + 2\ SiSi + 2\ SiB + 2\ BB + 3\ SiSiSi + 3\ SiSiB + 3\ SiBB + 3\ BBB}$$



- Na^+ ions act as network modifiers.
- # NBO atoms ↑ with $[Na_2O]$ and β_c (or n) ↑ significantly

- Na^+ ions network compensators
- # NBO atoms is independent of $[Na_2O]$, and hence, β_c (or n) are ~ constant

M. Barlet et al, JNCS 2016. doi: 10.1016/j.jnoncrysol.2016.07.017

RETICULATION LEVEL & CRACK PROPAGATION

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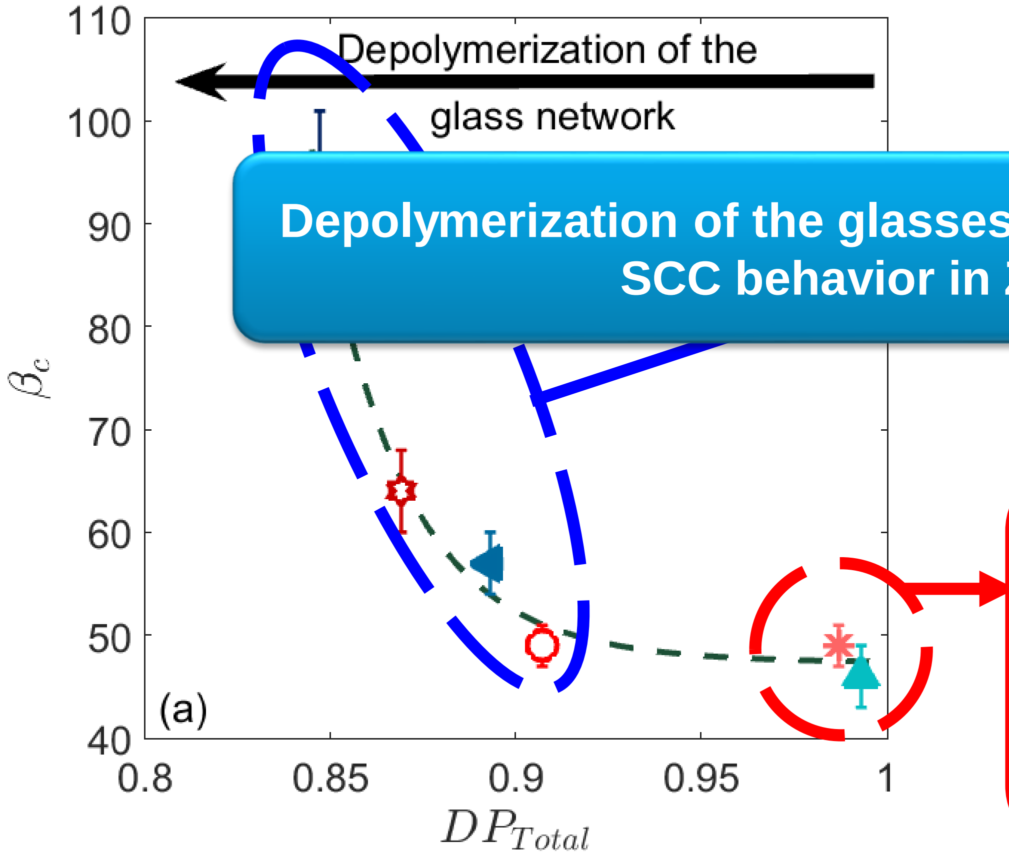


Depolymerization of the glasses network controls the SCC behavior in Zone I.

work

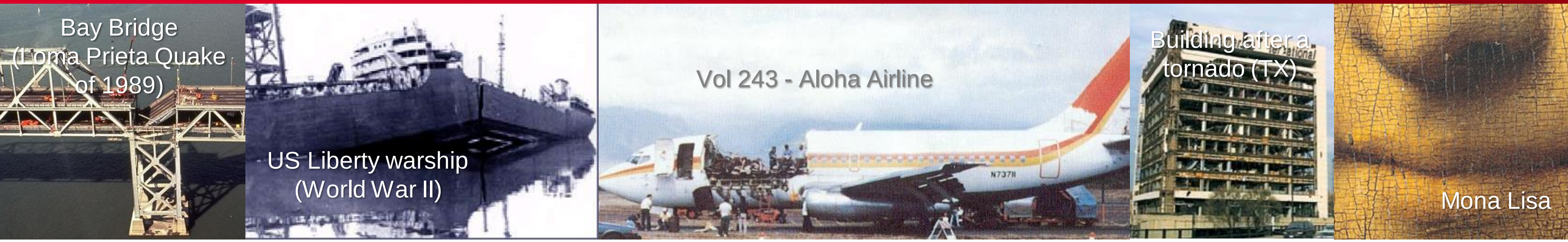
[Na_2O]

ificantly

- Na^+ ions network compensators
- # NBO atoms is independent of [Na_2O], and hence, β_c (or n) are ~ constant

M. Barlet et al, JNCS 2016. doi: 10.1016/j.jnoncrysol.2016.07.017

SUMMARY – SBN GLASSES



Na_2O plays an important non-constant role for different parameters

- ↑ Na_2O → ↑ Poisson's ratio: linear ratio
- Stress Corrosion cracking: Region 0
  - ↑ Na_2O → ↑ K_e: linear ratio
- Stress Corrosion cracking: Slope in region 1
  - Null dependence on Na_2O when Na^+ ions network compensators
  - ↑ slope when Na^+ ions network modifiers on Si and B networks

THANK YOU FOR YOUR ATTENTION

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Financement

- ANR ToughGlasses
- CEA-DEN-Vestale
- ANR-Corcosil
- RTRA-FracHet
- IDF-équipement mi-lourd 2011: IMAFMP
- RTRA-IMAFMP
- Dim-Map – AFM4aStory
- PALM - CoEuRs