Dynamic fracture of alkali silicate glasses: insights from large scale atomistic simulations

Zhen ZHANG*, Walter KOB

Simona ISPAS

*Department of Physics, College of Mathematics and Physics, Chengdu Univ. of Technology, China

Outline

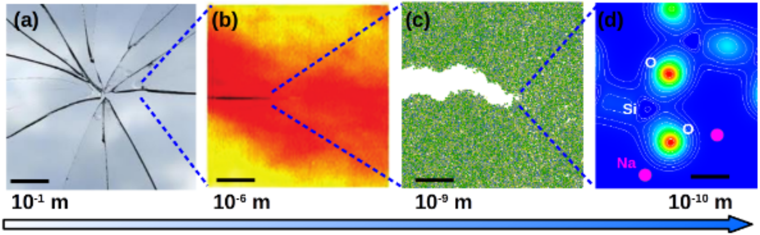
1. Motivations. Simulation details
2. Dynamic fracture
   - Stress-strain response
   - Cavitation
3. Evolution of atomic-level properties
4. Surface topography: melt formed *vs* fracture surface
5. Conclusions

Silicate glasses:

- No doubt that we are now living in the Glass Age
- Increasing need to control the glass properties and their mechanical reliability
- Silicate glasses have many exceptional properties BUT they are **brittle**!
- Fracture and deformation properties probed in a considerable number of experimental and computational studies

Silicate glasses

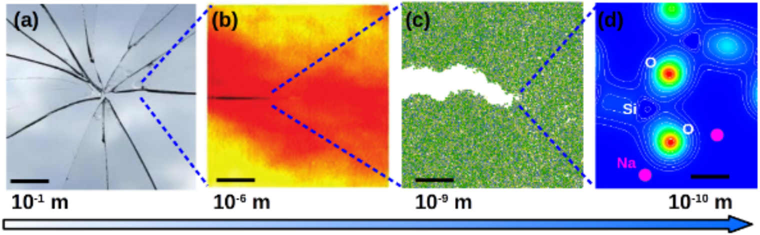
Archetypical brittle materials (macroscopic scale)



- Glass fracture: a multiscale phenomenon, involving different mechanisms
- Microscopic mechanisms of glass fracture and deformation are still under debate.
  Is fracture accompanied by the formation/growth and coalescence of microscopic cavities?
- More microscopic insights are needed to understand glass fracture and deformation

Silicate glasses

Archetypical brittle materials (macroscopic scale)



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Use **large scale computer simulations** to obtain a deeper understand of glass fracture

Large scale molecular dynamics simulations

Compositions, potential choice & system size

- Compositions
  - pure silica
  - sodium binary silicates
    Na$_2$O - xSiO$_2$ (**NS**x, $x = 20, 10, 7, 5, 4, 3$, i.e. from 5 to 25% mol)
  - alkali binary silicates
    A$_2$O - 3 SiO$_2$ (**AS3**), **A**=Li, Na, or K (25% mol)
- **SHIK** pair potential

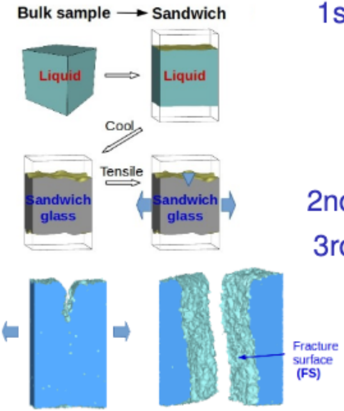
$$V(r_{ij}) = \frac{q_i q_j}{r_{ij}} + A_{ij} \exp\left(-B_{ij} r_{ij}\right) - \frac{C_{ij}}{r_{ij}^6} + \frac{D_{ij}}{r_{ij}^{24}}$$

Sundararaman, Huang, Ispas & Kob, JCP **148** (2018) & **150** (2019)

- System size: **up to** 2.3 **milion atomes**, 20nm×30nm×50nm

Large scale molecular dynamics simulations

Simulation protocol



1st **High temperature equilibration:** bulk liquid sample + introduction of vacuum layers
$\Longrightarrow$ **sandwich sample** with free dry surfaces

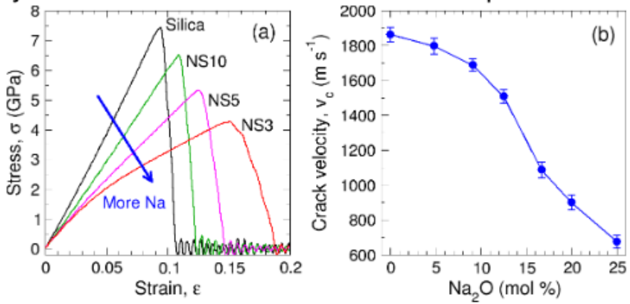
2nd Quench and relaxation at 300 K

3rd Fracture sample using tensile stress (dynamic fracture, 0.5 ns^{-1})
$\Longrightarrow$ fracture surface (**FS**)

Zhang, Ispas & Kob, PRM **6** (2022) & JCP **158** (2023), Z. Zhang Phd Thesis, Univ. Montpellier, 2020

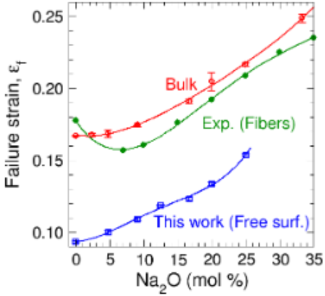
Dynamic fracture: composition dependence

Crack velocity is fast and thus environment has no impact



- With increasing Na concentration glass becomes more ductile (increasing failure strain)
- Significant variation of crack velocity with Na concentration; 10% Na_2O threshold composition

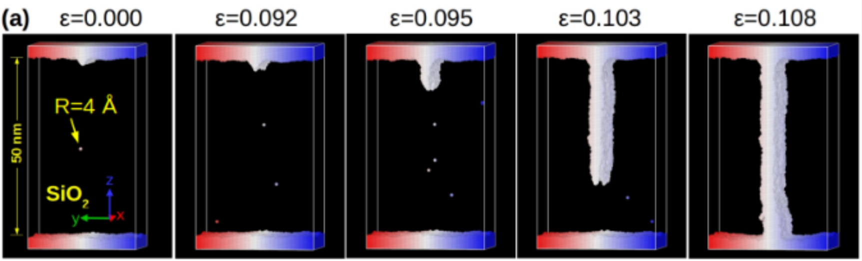
Failure point: composition dependence

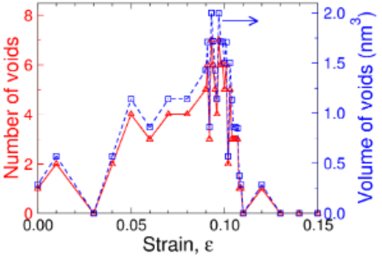


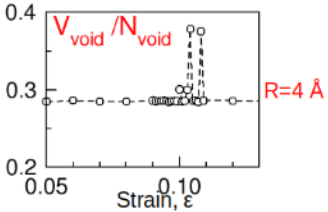
- Composition influences strongly the failure point
- Experimental trend reproduced
- Surface flaw: reduction of failure strain for the sandwich glasses.

Sim. bulk data: Zhang, Ispas & Kob Acta Mater. 231 (2022)
Exp. Lower et al. JNCS (2004)

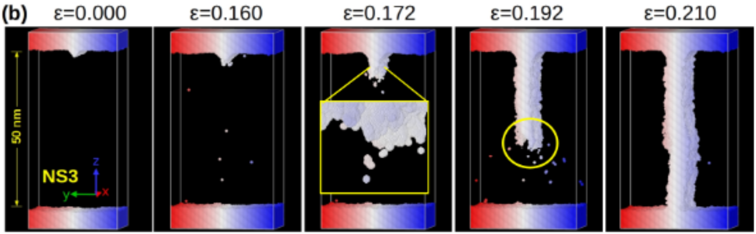
Silica glass: completely brittle

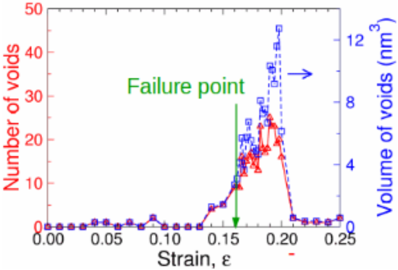






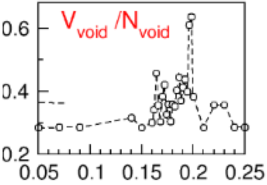
Sodium rich glass NS3: quasi-brittle



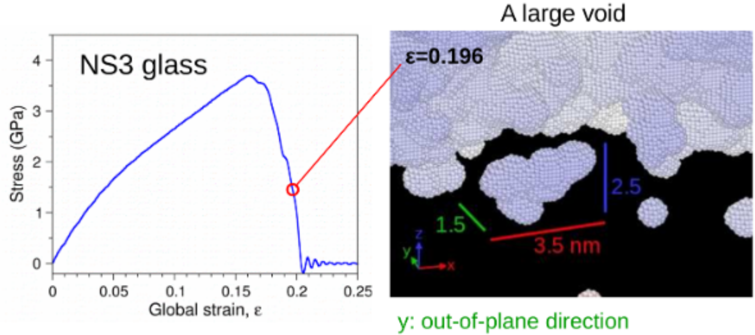


Micro-branching

Voids ahead of the crack front



Sodium rich NS3 glass: voids

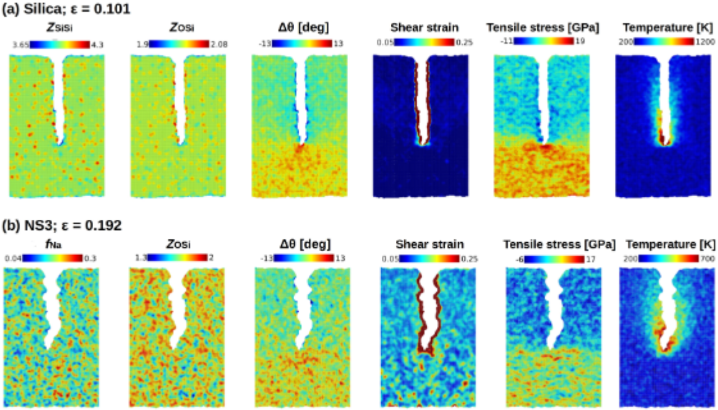


- Voids grow to several nanometers and have very irregular shapes
- Hard to detect the voids by comparing post-mortem fracture surfaces

Atomic-level properties

- Na fraction: $f_{\mathrm{Na}} = N_{\mathrm{Na}}/N_{\mathrm{tot}}$ (nominal $f_0 = 0.167$ for NS3)
- Local network connectivity: Z_{SiSi}, Z_{OSi}
- Angular change: $\Delta\theta$(SiOSi), (reference: strain=0)
- Temperature: $T = \frac{2E_{kin}}{3Nk_B}$ (kinetic energy $\rightarrow$ T)
- Tensile stress: component of the per-atom stress tensor in the pulling direction

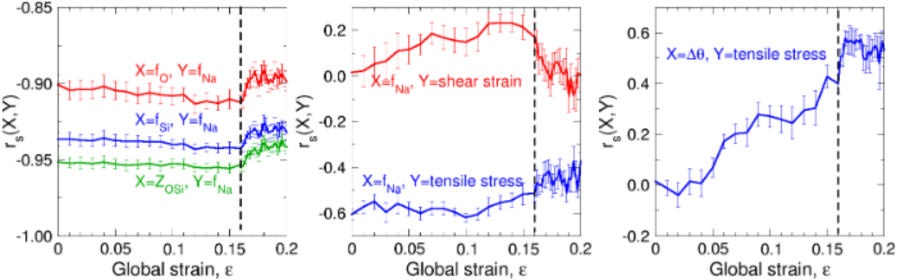
Local properties are heterogeneous



- **Silica:** more stress concentration & local heating ($T_c > 1200$ K)
- NS3: more heterogeneity & dissipation ($T_c \approx 700$ K)

Local properties are correlated (Na rich glass)

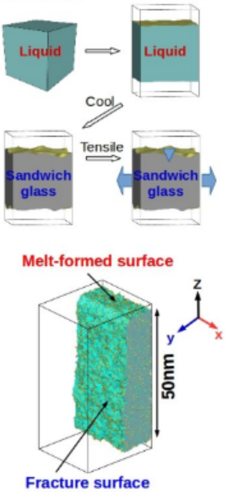
r_S = Spearman's rank correlation coefficient



- Strong correlations among the local elemental concentrations and network connectivity
- Na-rich regions more flexible/depolymerized: lower the stress & promote plastic deformation
- Increasing correlation between $\Delta\theta$(SiOSi) and stress due to the activation of soft modes

Large scale molecular dynamics simulations

Simulation protocol



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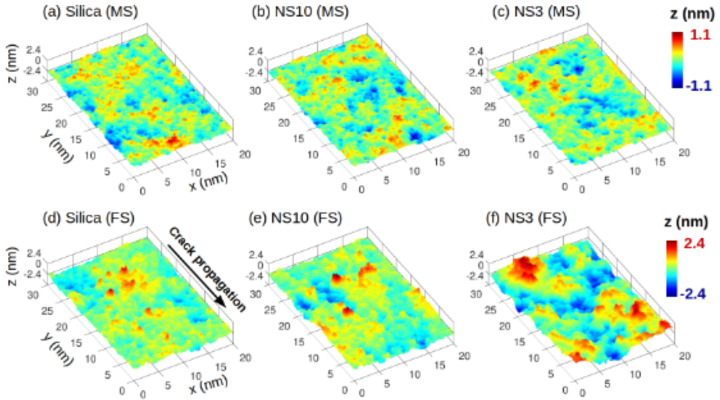
Surface topography: melted vs fracture surface



Melt formed surface

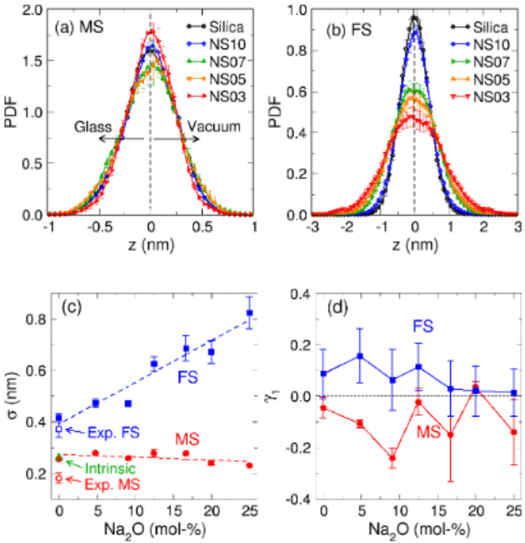
Fracture surface

Morphology depends on surface type & composition

Zhang, Ispas & Kob, PRL **126** (2021)

Surface topography

Roughness: peaks & valleys



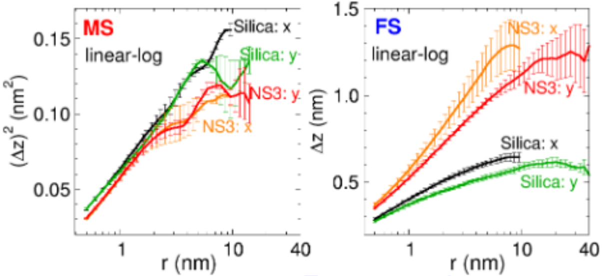
$$\sigma = \sqrt{\langle z^2 \rangle - \langle z \rangle^2}$$

- MS weak dependence on Na content
- FS rougher than MS and rougher with increasing Na content
- MS more deep holes
- Intrinsic roughness: frozen capillary waves prediction
  Exp.:Gupta et al. 2000

Scaling properties at the nanoscale

1D height-height correlation function

$$\Delta z(r) = \sqrt{\langle [z(r+x) - z(x)]^2 \rangle_x}$$



Melt surface

– isotropic
– capillary wave theory works well

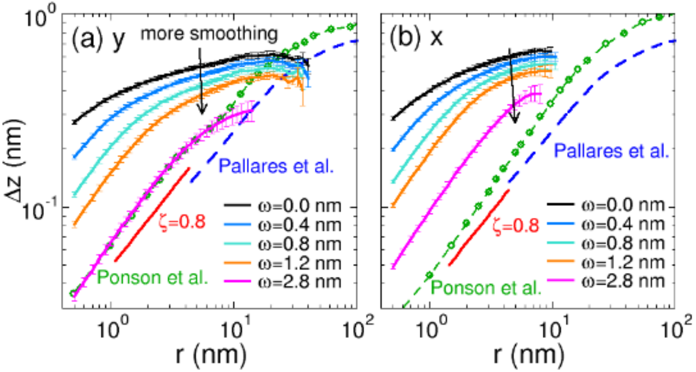
$$(\Delta z)^2 \propto \ln(r)$$

Fracture surface

– anisotropic and composition dependent
– logarithmic growth: $\Delta z \propto \ln(r)$
– surface is not a fractal on these length scales

Spatial resolution plays a key role on the nanoscale

Fracture surface



- smoothing of the surface profile: one switches from a **logarithmic** dependence to **power-law** scaling (self-affine)

Exp.: Ponson et al. 2006, Pallares et al. 2018

Summary

- **Dynamic fracture of silicate glasses:**
  - Fracture: complex phenomenon depending on multiple factors, such as length scale, loading condition, and composition.
  - Silica is completely brittle while Na-rich glasses show a certain degree of ductility
  - Exploration of spatial and temporal evolution of various atomic-level properties: notable crack tip heating, heterogeneity and correlations among these properties
- **Surfaces of silicate glasses**
  - Properties of glass surface depend strongly on production history (melt surface vs fracture surface)
  - Topography of fracture surface is not a fractal on the length scale of a few nm

Zhang, Ispas & Kob, PRL **126** (2021), PRM **6** (2022) Zhang, Ispas & Kob, JCP **153** (2020) & **158** (2023)

Z. Zhang Phd Thesis, Univ. Montpellier, 2020

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Thank you for your attention!