Simulation of Aging in SiO$_2$: Single Particle Jump Analysis

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Model: BKS Potential

[B.W.H. van Beest et al., PRL 64, 1955 (1990)]

\[
\phi(r_{ij}) = \frac{q_i q_j e^2}{r_{ij}} + A_{ij} e^{-B_{ij} r_{ij}} - \frac{C_{ij}}{r_{ij}^6}
\]

112 Si & 224 O \quad \rho = 2.32 \text{ g/cm}^3

\(T_c = 3330 \text{ K}\)

Dynamics:

Simulation Runs:

\(T\)

- 5000 K
- 3760 K
- 3250 K
- 3000 K
- 2750 K
- 2500 K

\(T_i\)

- 0.33 ns
- 33 ns

\(T_f\)

(Nosé Hoover)

(NVE)

(NVT)

aging:

waiting time \(t_w\)

[SiO\textsubscript{2}, GeO\textsubscript{2}, Glycerol]

Tg/T

\(\log \eta \text{ [Poise]}\)

strong glass former

fragile glass former

(compare below)

SiO\textsubscript{2} (here)

[C.A. Angell et al. 1976]
Generalized Intermediate Incoherent Scattering Function

\[ C_q(t_w, t_w + t) = \frac{1}{N_\alpha} \sum_{j=1}^{N_\alpha} e^{i\vec{q} \cdot (\vec{r}_j(t_w + t) - \vec{r}_j(t_w))} \]

\[ q=1.7 \text{ Å}^{-1} \]
\[ T_i=5000 \text{ K} \]

\( t_w = 0 \text{ ns} \)
\( t_w = 24.0 \text{ ns} \)

- \( C_q(t_w, t_w + t) \) depends on waiting time \( t_w \) (colors)
- three time windows:
  - too short
  - intermediate (scaling)
  - equilibrium \( (t_{eq}^C) \)
- equilibrium curve included in scaling

[KVL, J. Roman, J.Horbach, PRE 81, 061203 (2010)]
Mean Square Displacement

\[ \Delta r^2(t_W, t_W + t) = \frac{1}{N} \sum_{j=1}^{N} (r_j(t_W + t) - r_j(t_W))^2 \]

Goal:

Single Particle Picture

\(T_i = 2500\, \text{K}\)
\(T_f = 5000\, \text{K}\)

O-atoms

- \(\Delta r^2(t_W, t_W + t)\) depends on waiting time \(t_W\) (colors)
- three time windows

Waiting time \(t_W\)
Jump Definition

\[ \Delta \bar{r}_n \]

\[ \{ \bar{r}_n \}, \sigma \quad \{ \bar{r}_n \}, \sigma \quad \{ \bar{r}_n \}, \sigma \quad \{ \bar{r}_n \}, \sigma \]

\[ \sigma \geq 3 \Delta \bar{r}_n \]

[\text{KVL, R. Bjorkquist, L.M. Chambers, PRL 110, 017801 (2013)}]
Jump Definition: Aging Dependence

\[ \bar{r}_n(t) \]

- Waiting time \( t_w \)
- Waiting time windows \( \Delta t_w \)
Average Jump Length

- **O-atoms jump farther than Si-atoms**
- **compare:**
  \[ d_{\text{SiO}} = 1.6\,\text{Å}, \quad d_{\text{OO}} = 2.6\,\text{Å}, \quad d_{\text{SiSi}} = 3.1\,\text{Å} \]
- **\( \Delta R \) mostly independent of \( t_w \)**
strong glass former SiO$_2$:

- $P(\Delta R)$ independent of $t_w$
- exponential decay
- compare fragile glassformer binary LJ (& polymer)

[Warren & Rottler, EPL(2009)]
Time Averages: Jump Duration $\Delta t_d$ & Time in Cage $\Delta t_b$

- $\Delta t_b \gg \Delta t_d$
- $t_w \approx 10$ artifact due to finite simulation run
- $\Delta t_b$ independent of $t_w$!
Distribution of Time in Cage $P(\Delta t_b): t_w$ varied

strong glass former SiO$_2$:

- $P(\Delta t_b)$ independent of $t_w$!
- compare fragile glassformer (binary LJ &) polymer

[Warren & Rottler, EPL(2009)]
Distribution of Time in Cage $P(\Delta t_b)$: $T_f$ varied

strong glass former SiO$_2$:

crossover
- power law to exponential
- at $t_{\text{cross}} \approx t_{\text{eq}}$

compares fragile glassformer
binary LJ

[Warren & Rottler, PRL 2013]
Number of Jumping Particles per Time

\[ \frac{N_p}{\Delta t_{w}} \] depends strongly on waiting time \( t_w \)

- \( \frac{N_p}{\Delta t_{w}} \) decreasing with increasing \( t_w \)
- compare: soft colloids [Yunker et al., PRL (2009)]

\[ t_j^{\text{eq}} \approx t_{\text{eq}}^{C} \] (arrows)
Summary: Microscopic Picture of Aging

Aging of SiO$_2$:
- Only $t_w$-dependence: $N_p/\Delta t_w$ (not $P(\Delta R)$ and $P(\Delta t_b)$)
- $P(\Delta t_b)$ crossover power law to exponential
  - at $t_{\text{cross}} \approx t_{\text{eq}}^j \approx t_{\text{eq}}^C$

Compare with Fragile Glassformer:
- Surprising similar jump dynamics of strong and fragile glass formers
  - $P(\Delta R)$ and $P(\Delta t_b)$
  - $t_w$-independent
  - $P(\Delta t_b)$ crossover

[KVL, R. Bjorkquist, L.M. Chambers, PRL 110, 017801 (2013)]

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