Diffusion and phase separation in silicate melts – physics problems inspired by glass industry

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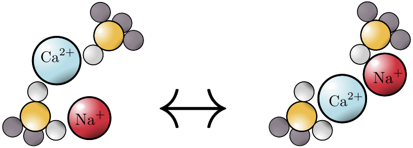

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A bridge between academic research and R&D

Basic research inspired by Saint-Gobain's products and processes

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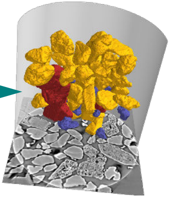

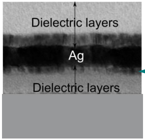

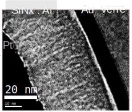

Viscous liquids quenched into amorphous solids

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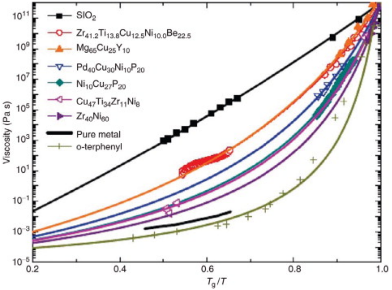
Glass transition and viscosity

Avoiding rearrangements :
fast quenching rates or
low mobilities



Silicate glasses are strong network-forming glasses : good glass-forming ability
Downside : low mobility, high viscosity
Origin : polymerization of (alumino)-silicate network

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Avoiding rearrangements :
fast quenching rates or
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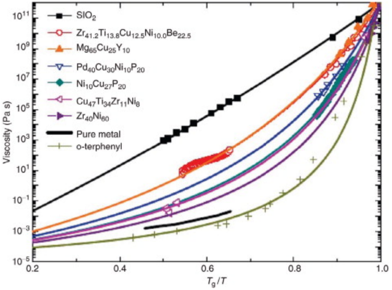

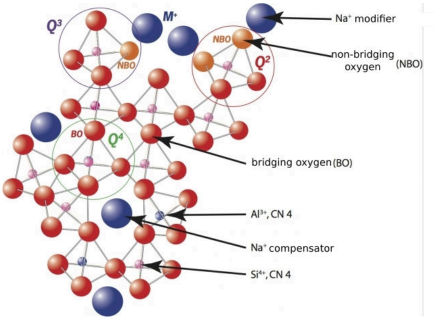

Silicate glasses are strong network-forming glasses : good glass-forming ability

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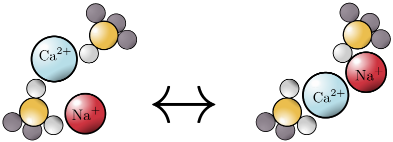
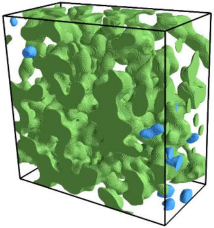
Mass transport phenomena in silicate glasses
Outline

1. Diffusion couplings in silicate melts

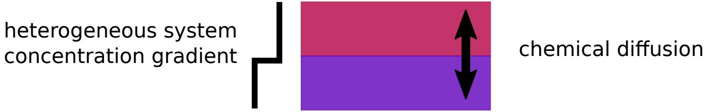
2. Morphology of phase separation





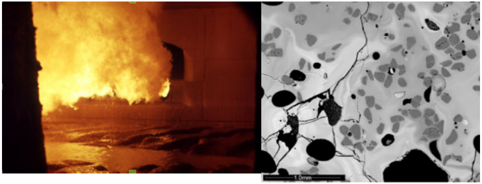
Outline

1. **Diffusion couplings in silicate melts**
2. Morphology of phase separation



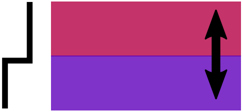
How to predict diffusive exchanges in heterogeneous systems?

glass melting

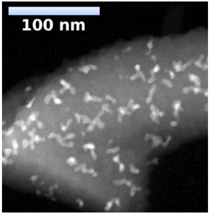


refractory corrosion

heterogeneous system
concentration gradient



chemical diffusion



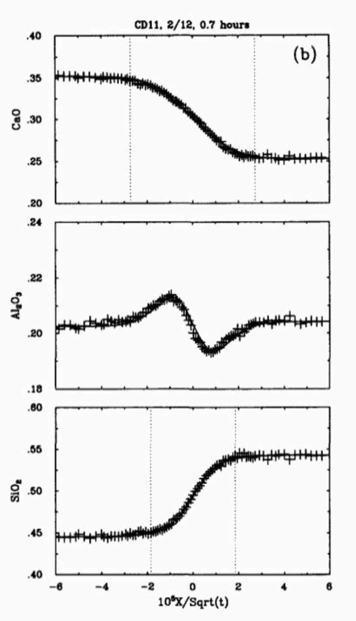
Dargaud 2011

crystallization
phase separation

thin films on glass substrate

How to predict diffusive exchanges in heterogeneous systems ?

Interdiffusion effects : uphill diffusion



← [Liang et al., 1996]

uphill diffusion

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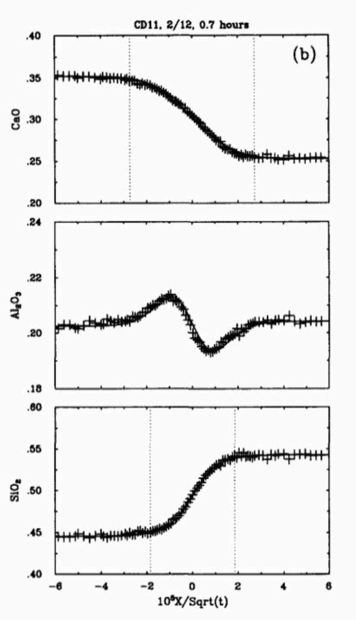

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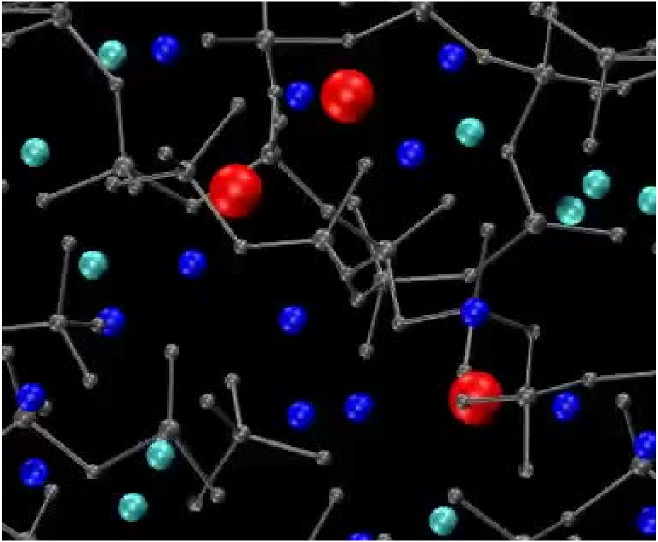
uphill diffusion

Diff Couple	$D(SiO_2)$ ($\mu m^2/s$)
Si–Ti	19.5 ± 2.8
Si–Al	15.7 ± 1.5
Si–Mg	30.0 ± 1.7
Si–Ca	28.7 ± 2.8
Si–Na	44.2 ± 4.0
Si–K	102.9 ± 19.5
Ti–Mg	
Mg–Ca	
Ca–Na	
An diss	

[Guo and Zhang, 2016]

Diffusion coefficient depends on counter-diffusing species

Diffusion and reorganizations of silicate network



https://www.youtube.com/watch?v=SOUIMspT4jw
A. Tilocca

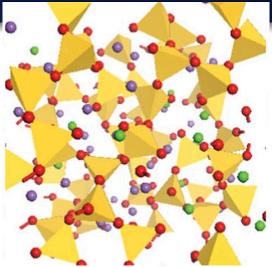
Diffusion matrix formalism

Fick's law

$$\mathbf{j} = -D \nabla C$$

$$\frac{\partial C}{\partial t} = D \Delta C$$

Diffusion matrix formalism



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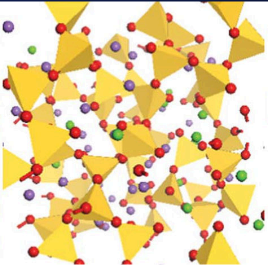
Diffusion matrix

$$\mathbf{j} = -\mathbf{D}\nabla\mathbf{C}$$

$$\mathbf{j}_i(\mathbf{x}) = -\sum_k D_{ik}\nabla C_k(\mathbf{x})$$

$$\frac{\partial}{\partial t}\begin{pmatrix} C_{\mathrm{Na}} \\ C_{\mathrm{Ca}} \\ C_{\mathrm{Al}} \\ C_{\mathrm{Si}} \end{pmatrix} = \begin{pmatrix} D_{\mathrm{Na,Na}} & D_{\mathrm{Na,Ca}} & D_{\mathrm{Na,Al}} & D_{\mathrm{Na,Si}} \\ D_{\mathrm{Ca,Na}} & D_{\mathrm{Ca,Ca}} & D_{\mathrm{Ca,Al}} & D_{\mathrm{Ca,Si}} \\ D_{\mathrm{Al,Na}} & D_{\mathrm{Al,Ca}} & D_{\mathrm{Al,Al}} & D_{\mathrm{Al,Si}} \\ D_{\mathrm{Si,Na}} & D_{\mathrm{Si,Ca}} & D_{\mathrm{Si,Al}} & D_{\mathrm{Si,Si}} \end{pmatrix} \Delta \begin{pmatrix} C_{\mathrm{Na}} \\ C_{\mathrm{Ca}} \\ C_{\mathrm{Al}} \\ C_{\mathrm{Si}} \end{pmatrix}$$

Diffusion matrix formalism



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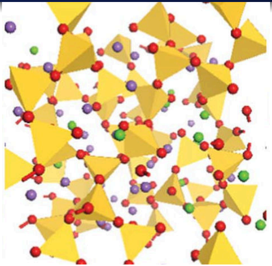

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Measured in several ternary systems, mostly in geosciences
[Liang et al., 1996], [Richter et al., 1998] : $\mathrm{CaO/MgO - Al_2O_3 - SiO_2}$

Also used in multicomponent metallic alloys

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Questions

What are the diffusion matrices in systems of industrial interest ?

- $Na_2O - CaO - SiO_2$ (NCS, W. Woelffel)
- $Na_2O - Al_2O_3 - SiO_2$ (NAS, V. Pukhkaya)
- $Na_2O - CaO - Al_2O_3 - SiO_2$ (NCAS, C. Claireaux)
- $Na_2O - CaO - Al_2O_3 - SiO_2 - ZrO_2$ (NCASZ, M. Ficheux)
- $Na_2O - B_2O_3 - SiO_2$ (NBS, H. Pablo)

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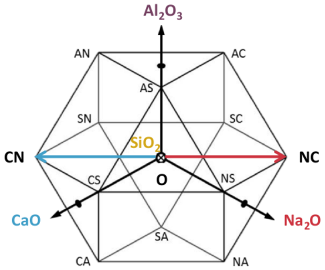
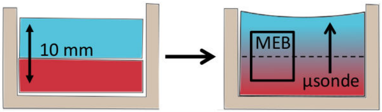


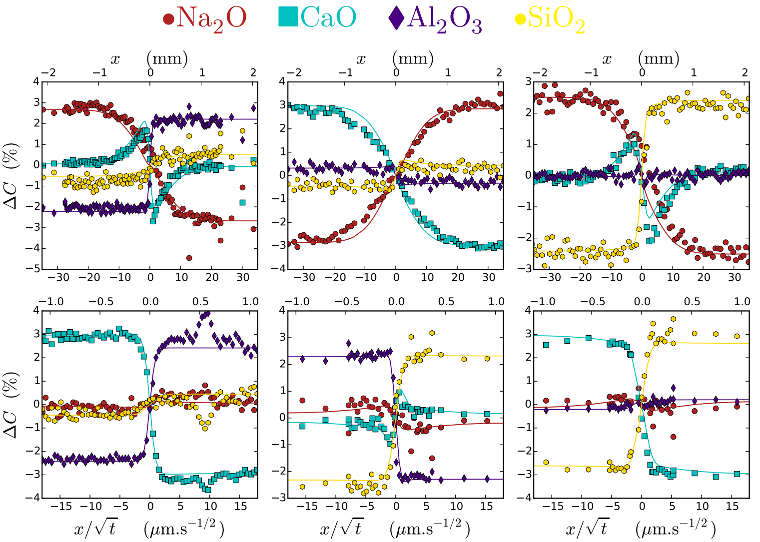


Large number of experiments

How do diffusion matrices depend on composition & temperature ?

Can we predict them ?

NCAS system at 1200°C



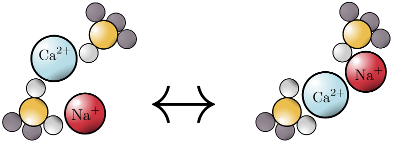
[Claireaux et al., 2016] GCA, Claireaux JNCS 2018

Python package to fit and simulate diffusion profiles : `multidiff`.

Diffusion eigenvectors at 1200° C

Dominant eigenvector

$$\frac{1}{2}Ca^{2+} \leftrightarrow Na^{+}$$



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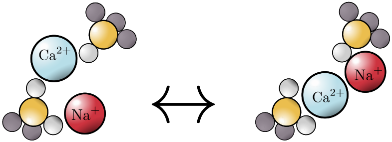
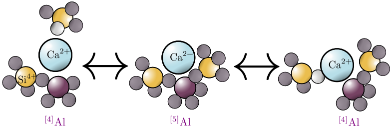


Second eigenvector (52x less frequent)

$$\mathrm{Ca}^{2+} \leftrightarrow 0.5\mathrm{Al}^{3+} + 0.5\mathrm{Si}^{4+}$$



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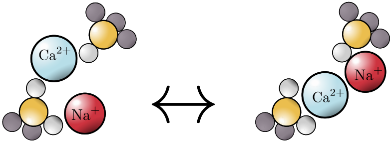

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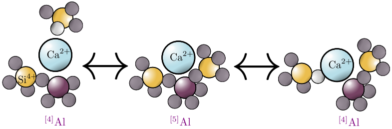
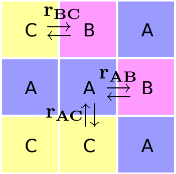


Third eigenvector (155x less frequent)

$$Al^{3+} \leftrightarrow 0.5Ca^{2+} + 0.5Si^{4+}$$

A toy model for multicomponent diffusion

Random exchange of neighbors with fixed probability r_{AB}



Coll. with B. Seoane

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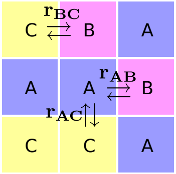

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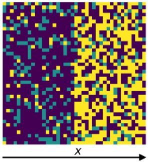
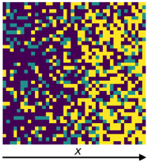


$t = n$ iterations



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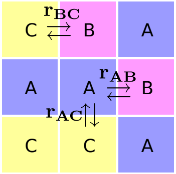

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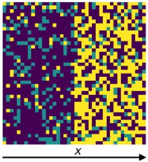

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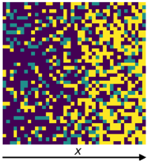

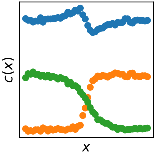

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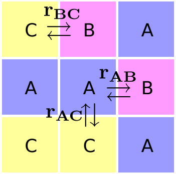

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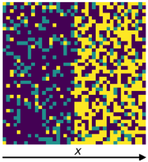

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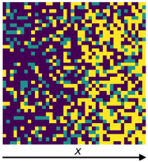

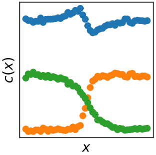

Analytical form of diffusion matrix from rates r_{ij}

$$\mathbf{D} = \frac{1}{3}\begin{pmatrix} (1 - c_2)r_{13} + c_2 r_{12} & c_1(r_{13} - r_{12}) \\ c_2(r_{23} - r_{12}) & (1 - c_1)r_{23} + c_1 r_{12} \end{pmatrix}$$

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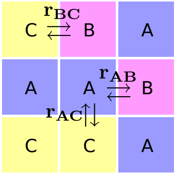

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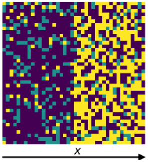

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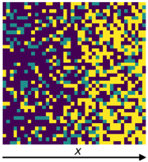

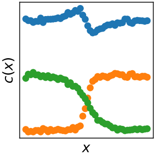

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composition	exchange rates		
NCS	$r_{NC} = 3.2$	$r_{NS} = 1.3$	$r_{CS} = 0$
NCAS	$r_{NC} = 1$	$r_{NA} = 0.4$	$r_{NS} = 0.4$
	$r_{CA} = 0$	$r_{CS} = 0$	$r_{AS} = 0$
BNS	$r_{BN} = 0.2$	$r_{BS} = 0$	$r_{NS} = 0.2$

← experiments

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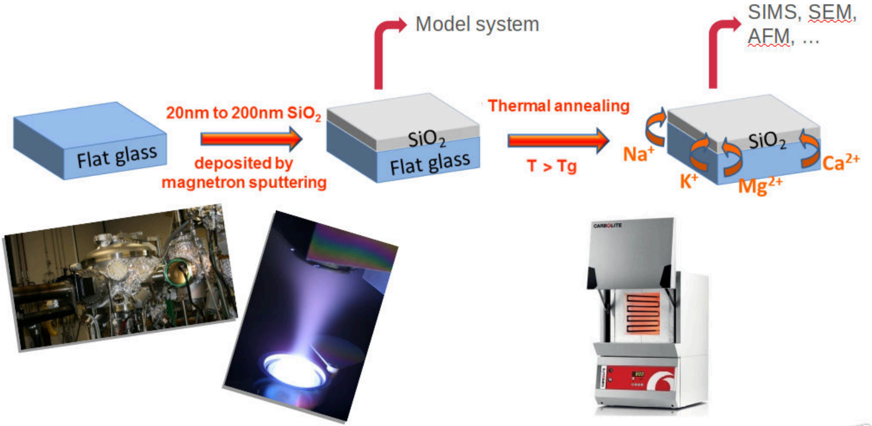
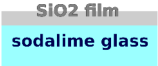
Annealing of PVD-sputtered silica layers on soda-lime substrate (Planiclear)



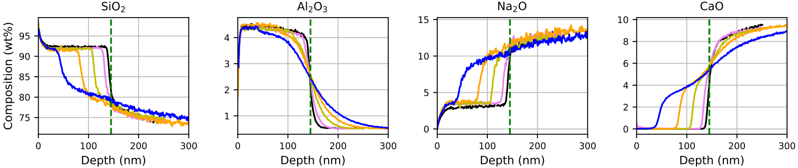
Pure and Al-doped silica thin films

Diffusive dissolution of thin film and multicomponent effects



Al-doped SiO_2 thin film on glass, different annealing times at 650° C

SIMS profiles



PhD JT Fonné ; Fonné et al., JACS 2017, JACS2018

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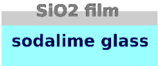

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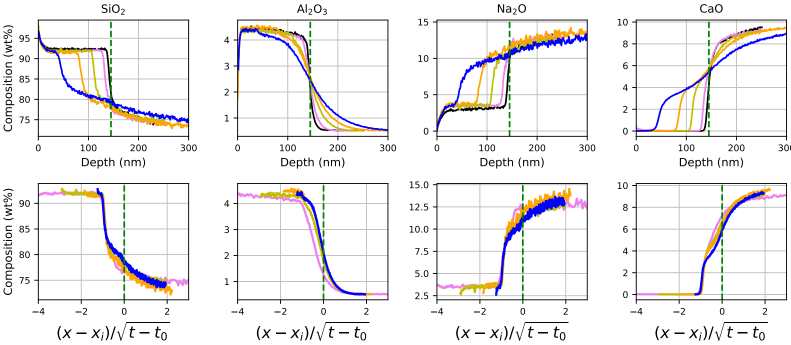
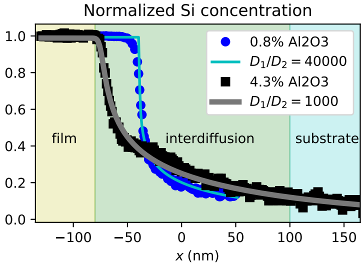


- Diffusion distance of Al smaller than for Si
- Na coupled to Si, Ca to both Si and Al.
- Can we use the bulk diffusion matrix to explain these results ?

PhD JT Fonné ; Fonné et al., JACS 2017, JACS2018

Fitting asymmetric diffusion profiles



High Si diffusivity (& viscosity) ratio between substrate and film

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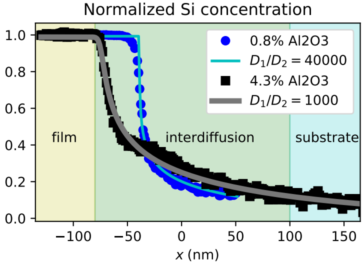
Fitting asymmetric diffusion profiles



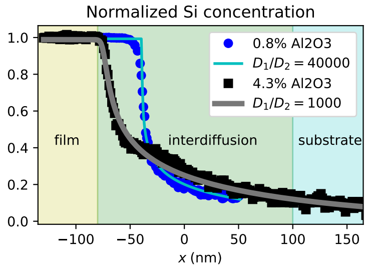
High Si diffusivity (& viscosity) ratio between substrate and film

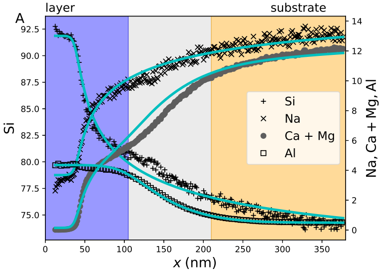
Using Crank's model to fit profiles :

$$D_{\mathrm{Si}} = D_0 \exp(-\beta C_{\mathrm{Si}})$$

Fitted values of β consistent with Eyring's law and viscosity model

Fitting asymmetric diffusion profiles





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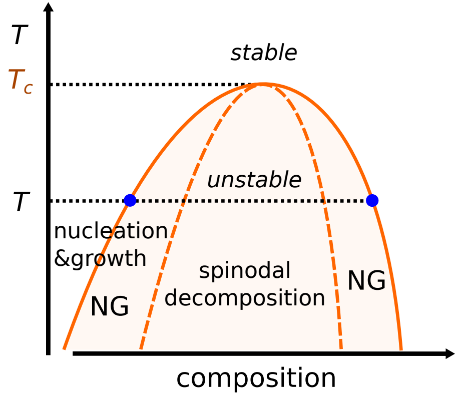
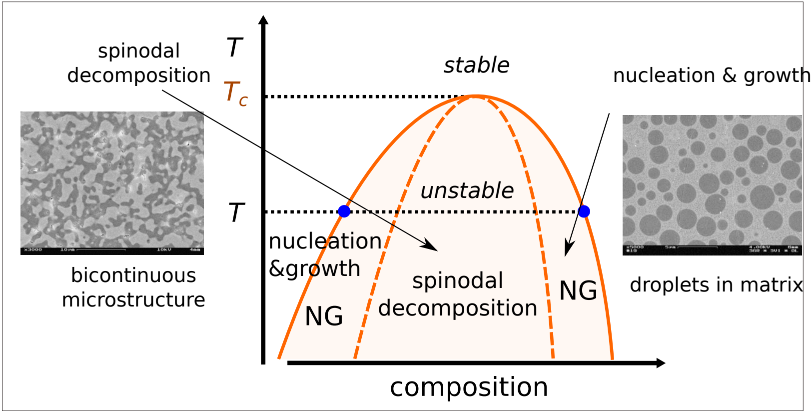
Use bulk eigenvectors to fit profiles

Outline

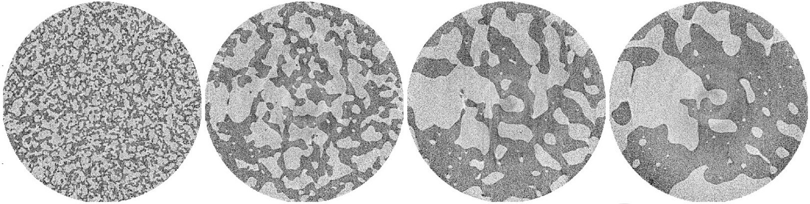
Principles of phase separation



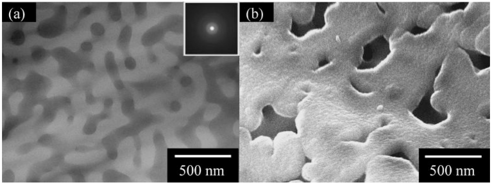
Principles of phase separation



Microstructure coarsening : decrease interfacial energy

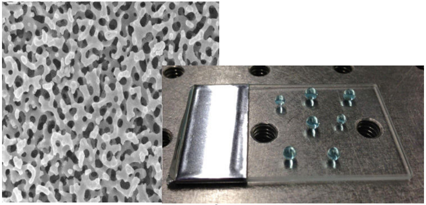


Porous membranes: Vycor



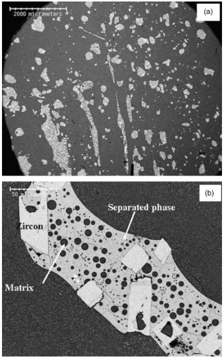
Suzuki et al. 2008

Super-hydrophobic porous films



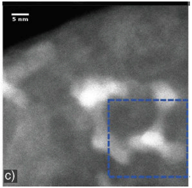
Aytug et al. 2013

Nuclear waste glasses



Martineau et al. 2010

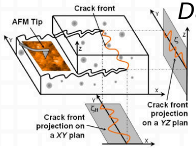
Dargaud et al . 2012

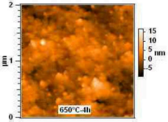


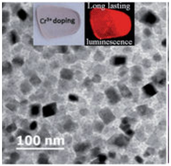
Glass ceramics

Model materials for crack propagation

Dalmas et al. 2008

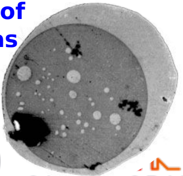






Microstructure of basaltic magmas

Veksler et al. 2007



Luminescent glass

Chenu et al. 2014

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A classical topic of statistical physics

Cahn-Hilliard equation

`https://www.youtube.com/watch?v=sysya3Lo78Y`

Fabio Garofalo

A classical topic of statistical physics

Cahn-Hilliard equation

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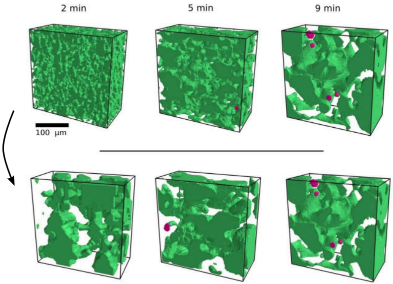
Fabio Garofalo

Dynamic scaling

$\times \frac{1}{\ell(t)}$



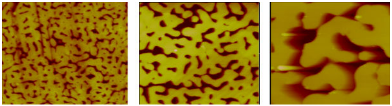
Is it the same microstructure ?

Coarsening mechanisms

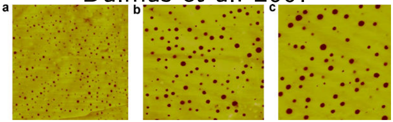
diffusion

$$\ell(t) \sim \left(\frac{\gamma D\Omega}{kT} t\right)^{\frac{1}{3}}$$

Regime observed in borosilicates

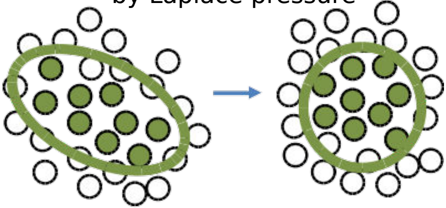


Dalmas et al. 2007



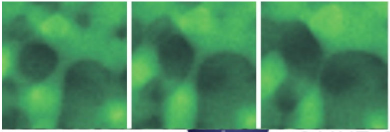
Wheaton et al. 2007

viscous flow induced
by Laplace pressure



Laplace pressure : $\gamma \left(\frac{1}{R_1} + \frac{1}{R_2}\right)$

$$\ell(t) \sim \frac{\gamma}{\eta} t$$



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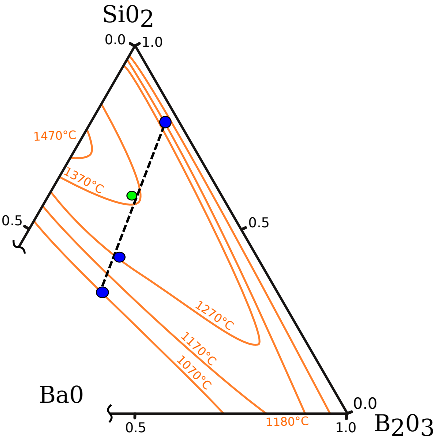
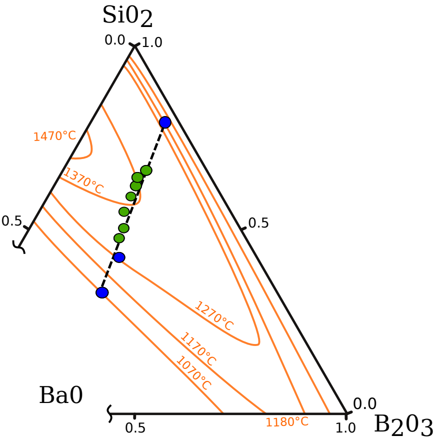
The system : barium borosilicates



Liquid-liquid phase separation

Different compositions separating into the same phases : different volume fractions.

The system : barium borosilicates



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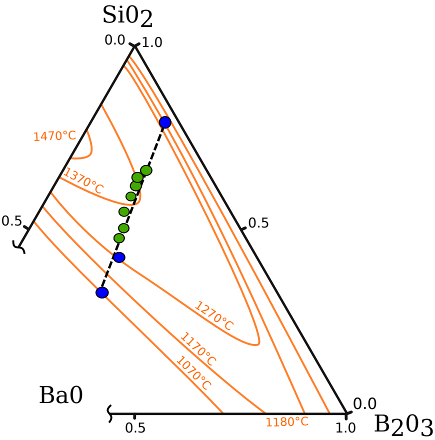

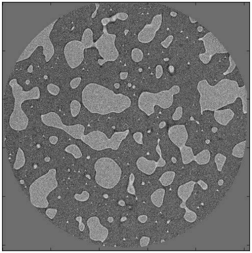

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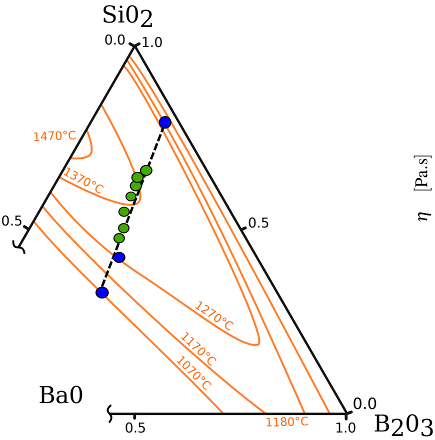
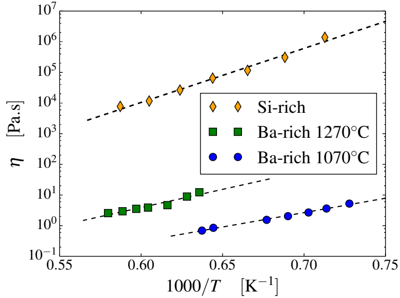


strong viscosity contrast

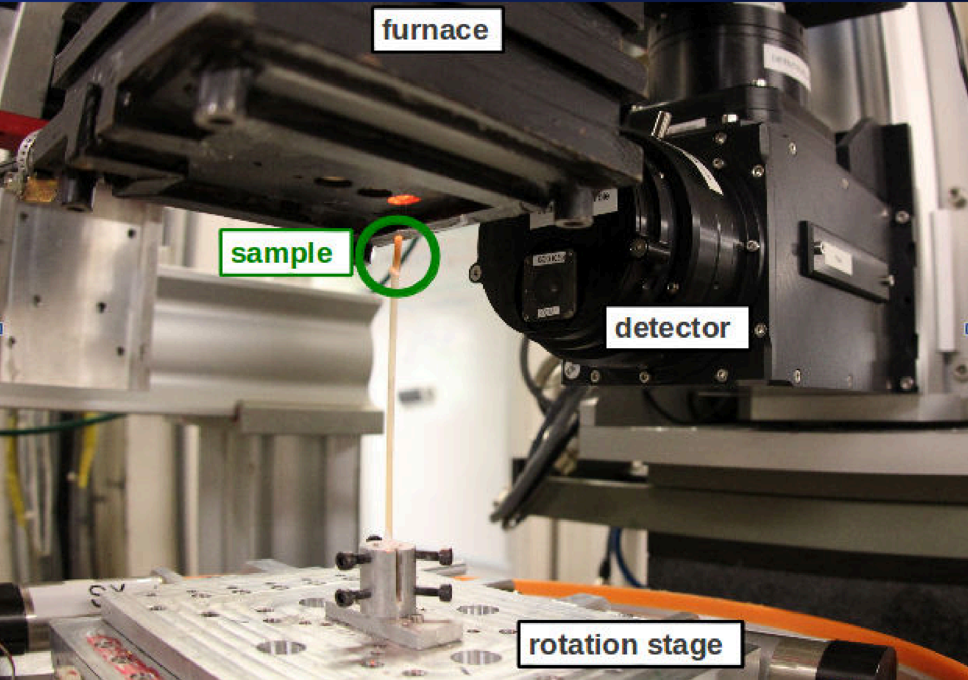


[Bouttes et al., 2015], Acta Mat. **92**

Liquid-liquid phase separation

Different compositions separating into the same phases : different volume fractions.

In-situ tomography on ID19 beamline, ESRF



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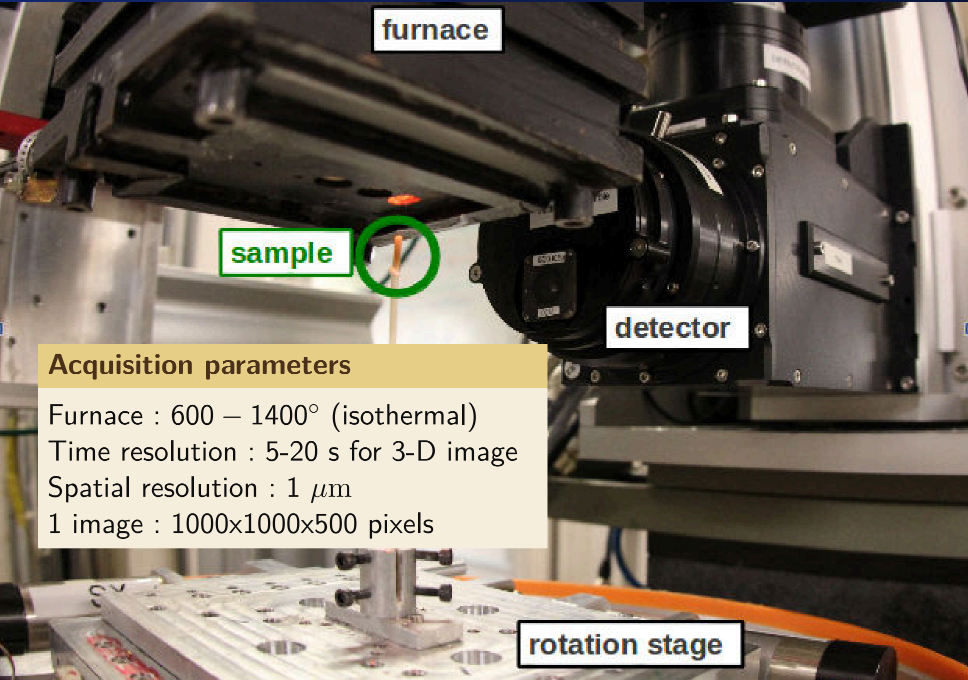
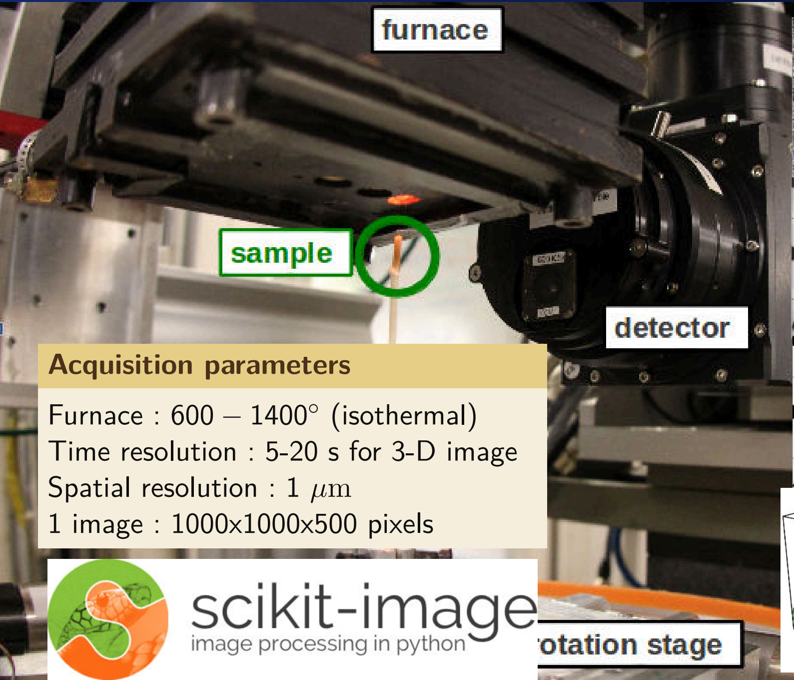


Acquisition parameters

Furnace : 600 – 1400° (isothermal)
Time resolution : 5-20 s for 3-D image
Spatial resolution : 1 μm
1 image : 1000x1000x500 pixels

In-situ tomography on ID19 beamline, ESRF



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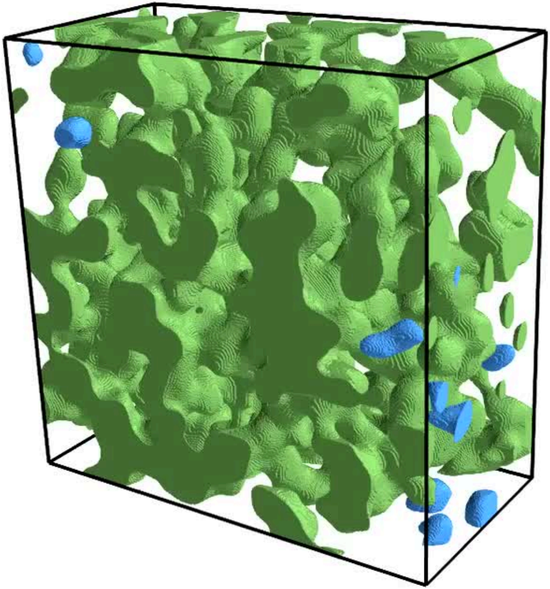
Spatial resolution : 1 μm
1 image : 1000x1000x500 pixels

scikit-image
image processing in python

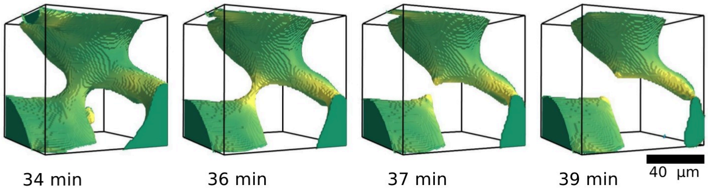
Coarsening : $\phi \leq 0.5$ case, 1200° C

box size : 400 μm, barium-rich phase represented



Only the less viscous phase breaks up

Barium-rich phase (less viscous) : liquid bridge breaks up



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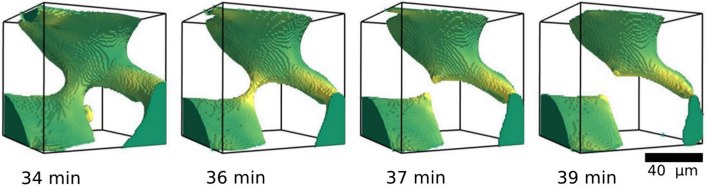
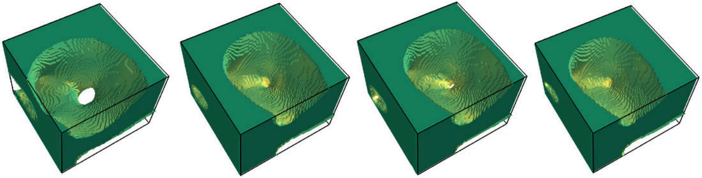


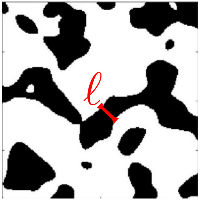
Silica-rich phase (more viscous) : loop is filled in



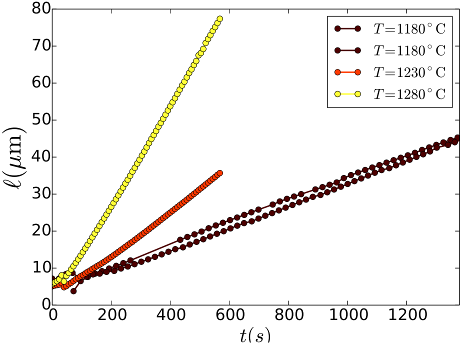
Break-up : strong shear preferentially in more fluid phase

$$\frac{\eta_{\text{viscous}}}{\eta_{\text{fluid}}} > 10^4$$

Evolution of characteristic length

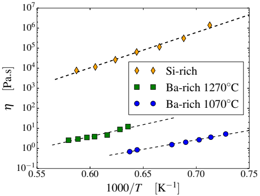


$$\ell = \frac{\mathcal{V}}{\mathcal{S}}$$

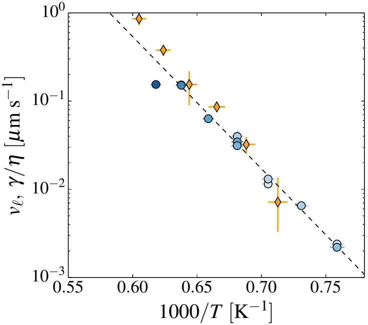


- linear evolution with time
- coarsening rate increases with temperature

Coarsening rate vs. temperature



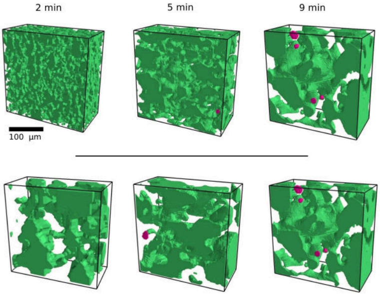
- coarsening rate $\dot{\ell}$, ♦ $\frac{\gamma}{\eta_v}$



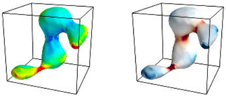
$$\dot{\ell}(t) \simeq \frac{\gamma}{\eta_v}$$

[Bouttes et al., 2015], Acta Mat. **92**

Dynamic scaling : self-similarity of microstructure

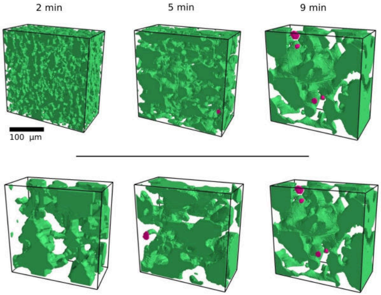


Curvatures



$$H = \frac{\kappa_1 + \kappa_2}{2}$$
$$K = \kappa_1 \kappa_2$$

Dynamic scaling : self-similarity of microstructure



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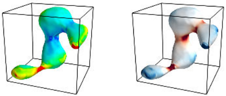

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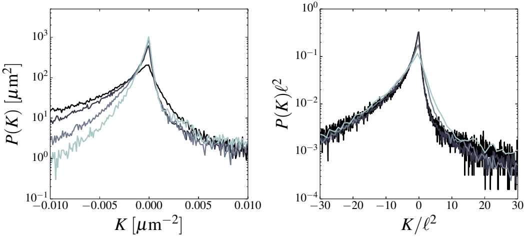
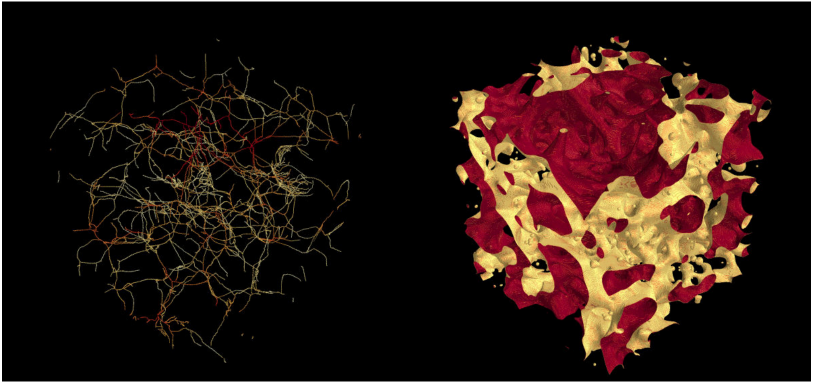
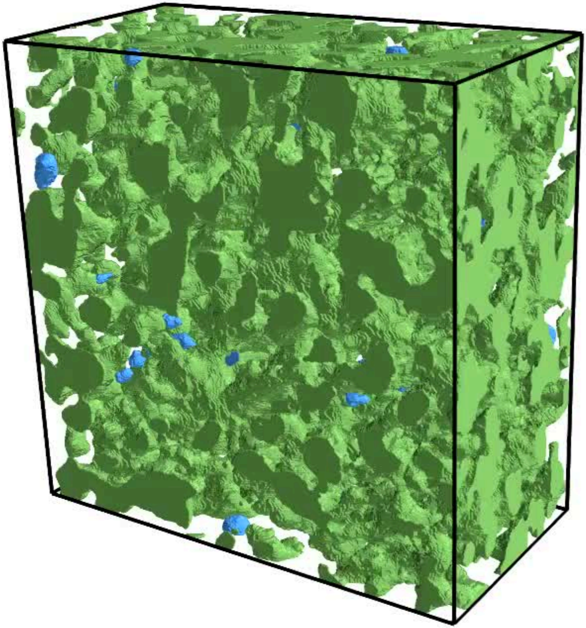


[Bouttes et al., 2014], PRL 112

Towards local statistics of break-ups



Coarsening and fragmentation, 1200° C



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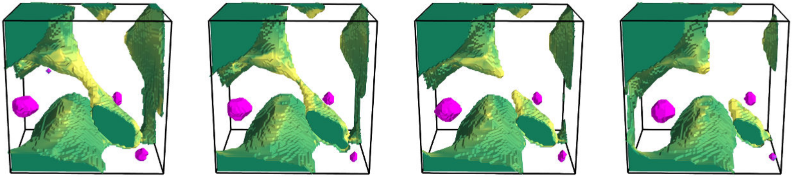
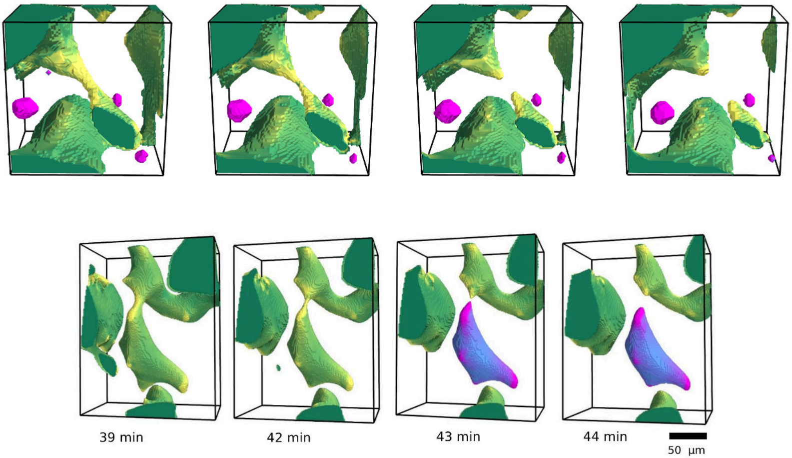
Break-up and fragmentation



Break-up and fragmentation

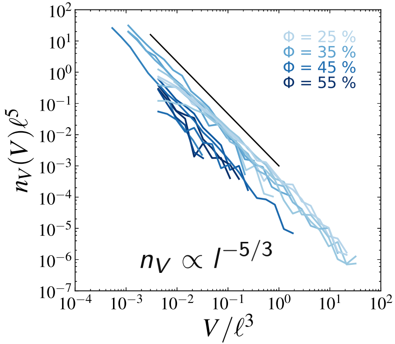


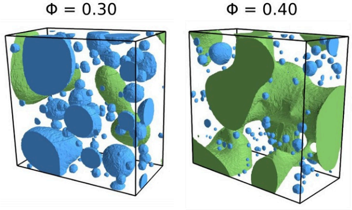
Only the barium-rich phase breaks up in domains

[Bouttes et al., 2014] PRL, [Bouttes et al., 2015] Acta Mat.

A broad distribution of domain sizes

Histogram of domain sizes



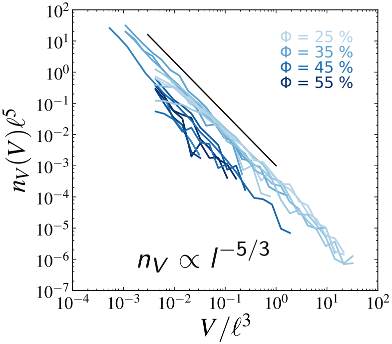


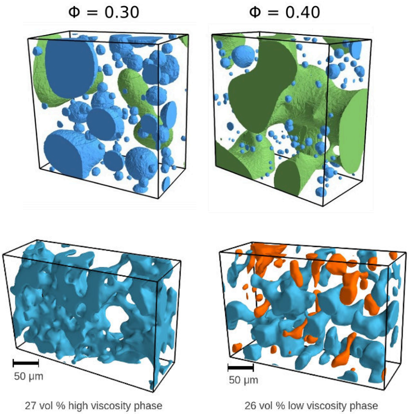
Different fragmentation times ⇒ different domain sizes

[Bouttes et al., 2014] PRL, [Bouttes et al., 2016] PRL

A broad distribution of domain sizes

Histogram of domain sizes





Viscous phase : long thin bridges

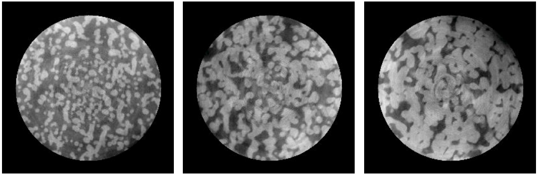
Fluid phase : rounded ends

Different fragmentation times ⇒ different domain sizes

[Bouttes et al., 2014] PRL, [Bouttes et al., 2016] PRL

Perspectives : towards smaller scales

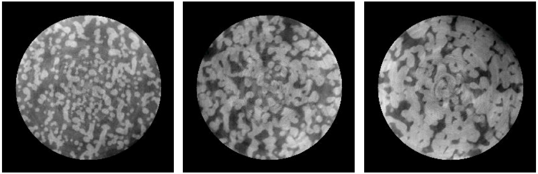
Coupling between diffusion and hydrodynamic transport



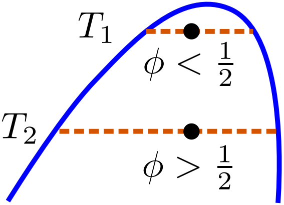
Nanotomography, C. Brillatz, FOV 100 μm

Perspectives : towards smaller scales

Coupling between diffusion and hydrodynamic transport

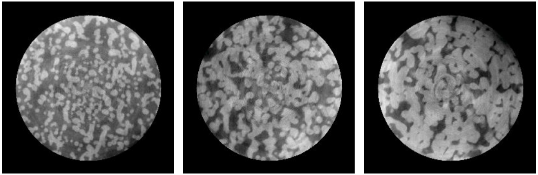


Nanotomography, C. Brillatz, FOV 100 μm

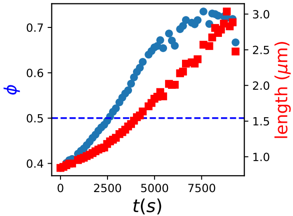


Perspectives : towards smaller scales

Coupling between diffusion and hydrodynamic transport

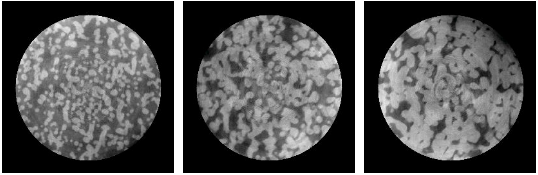


Nanotomography, C. Brillatz, FOV 100 μm

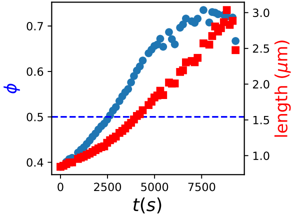


Perspectives : towards smaller scales

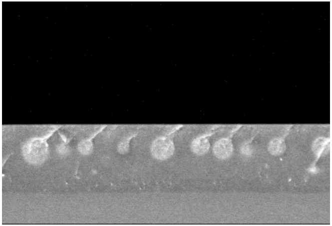
Coupling between diffusion and hydrodynamic transport



Nanotomography, C. Brillatz, FOV 100 μm



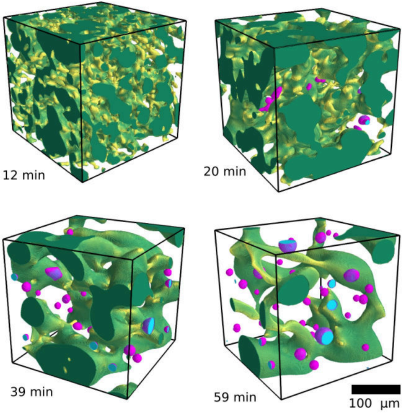
Phase separation in silicate thin films

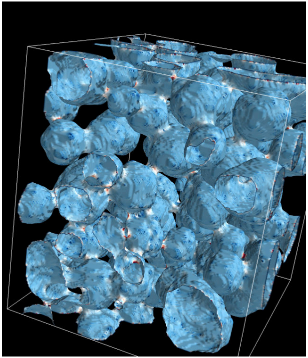


PhD JT Fonné, B Bouteille

- Droplet size depends on film thickness
- Interaction with substrate

Morphology evolution : beyond the simplistic picture





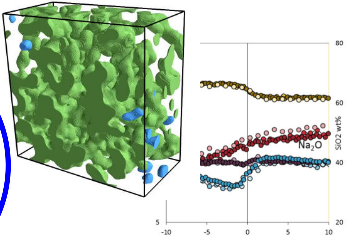
Importance of **hydrodynamic** effects and **viscosity contrast**

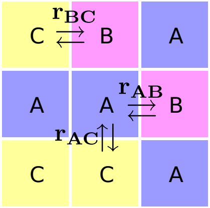
In situ and 3-D imaging needed

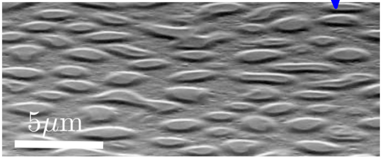
Conclusions

Industrial systems & questions

Controlled experiments, tools development







Materials properties and design

Microscopic understanding

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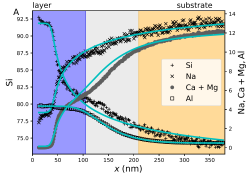
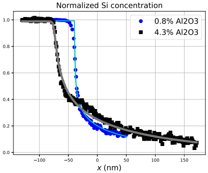
Conclusions

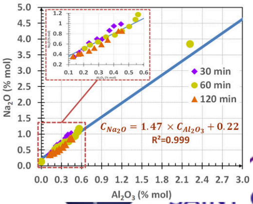
- Diffusion matrices : a powerful tool (useful outside of geochemistry !)



- Multicomponent effects modeled on bulk and thin films



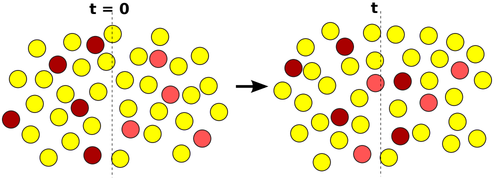
- Contrast of transport properties have to be modeled



- Exchanges with atmosphere cannot be neglected for thin films, role of water and Al content

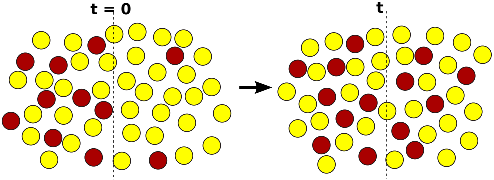
Different configurations for diffusion

Isotopic diffusion : marked tracer



[Jambon and Carron, 1976, Richter et al., 1999]

Chemical diffusion : gradient of chemical concentration



[Trial and Spera, 1994, Chakraborty et al., 1995a, Liang et al., 1996]