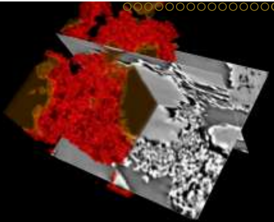
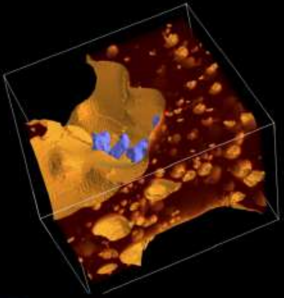
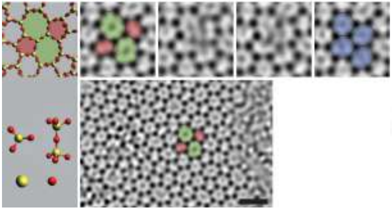




In-situ tomographic imaging of glasses and melts

Emmanuelle Gouillart
Joint Unit CNRS/Saint-Gobain SVI

not much to be seen !



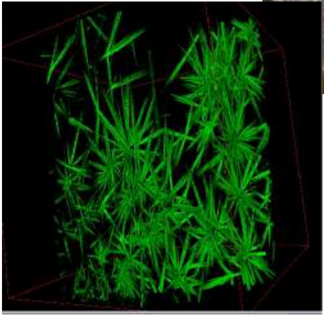
[Huang et al., Science 2013]

HRTEM of 2D silica

Not in this talk !

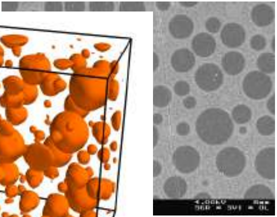
Imaging glasses and melts
Looking at heterogeneous systems

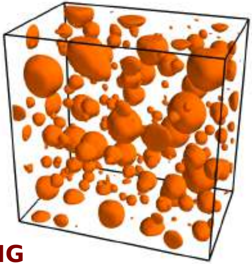
CRYSTALLIZATION



Sophie Schuller, Elise Régnier (CEA)

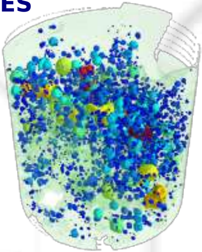
PHASE SEPARATION



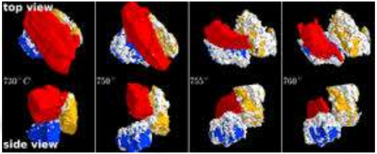


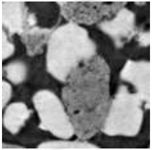
IMAGING PHASES

BUBBLES

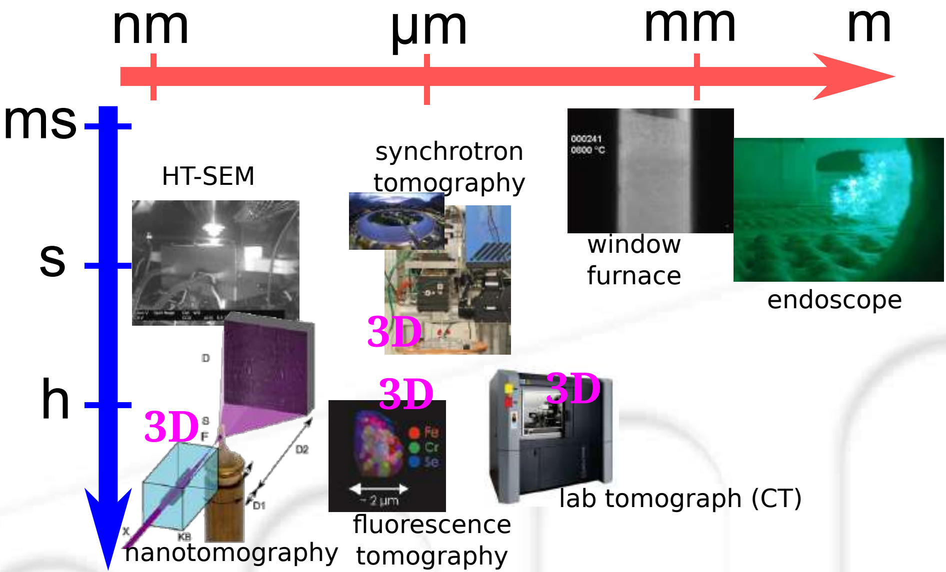


GLASS-FORMING BATCHES





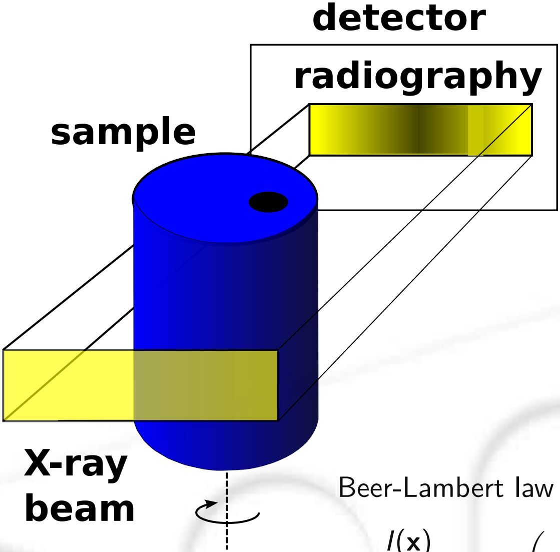
Different imaging modalities : speed vs. resolution



Outline

1. **In-situ tomography**

2. Applications
   - Phase separation
   - Glass reactive melting : reaction paths between granular raw materials



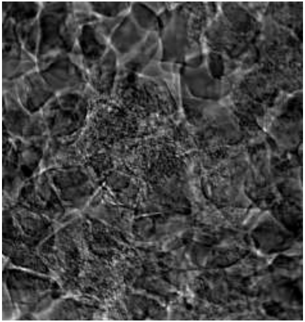
X-ray radiography

+ rotation

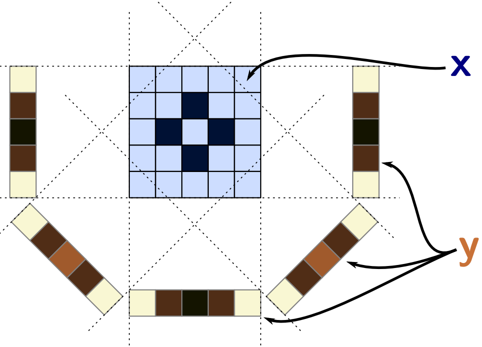
Beer-Lambert law

$$\frac{I(\mathbf{x})}{I_0(\mathbf{x})} = \exp\left(-\int \mu(s)\,\mathrm{d}s\right)$$

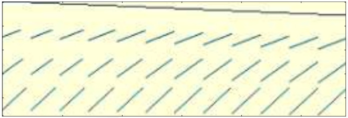
Obtaining 3-D absorption maps from 2-D radiographies



$\Rightarrow$



$$\mathbf{y} = \mathbf{A}\,\mathbf{x}$$



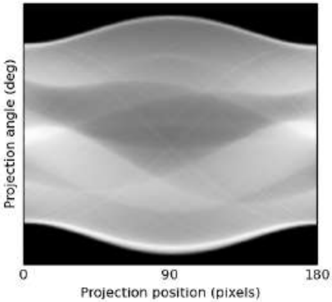
$$\mathbf{y} \in \mathbb{R}^n, \quad \mathbf{A} \in \mathbb{R}^{n \times p}, \quad \mathbf{x} \in \mathbb{R}^p$$

$n \propto$ number of projections

p : number of pixels in reconstructed image

We want to find x knowing A and y

Principle of synchrotron microtomography

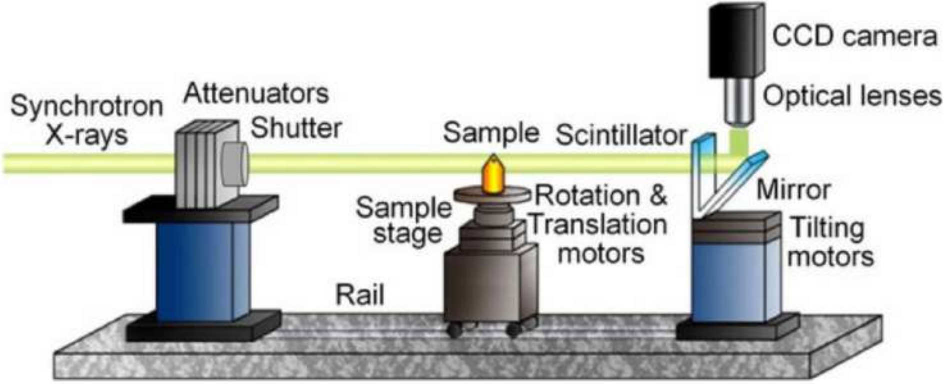


Unknown data :
One horizontal cut through sample

Measured data :
One line of the camera, at all projection angles

[Slaney and Kak, 1988]

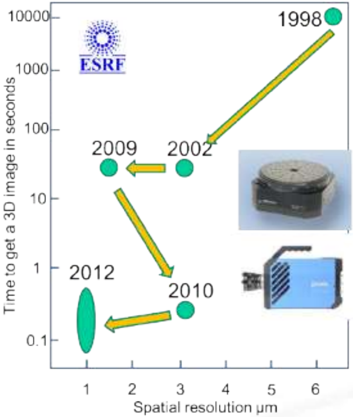
Synchrotron microtomography



Credit : Francesco De Carlo, APS

- **Spatial** resolution limited by
  - optical lenses (diffraction)
  - scintillator (blurring)
- **Time** resolution limited by
  - X-ray photon flux
  - camera sensitivity, . . .

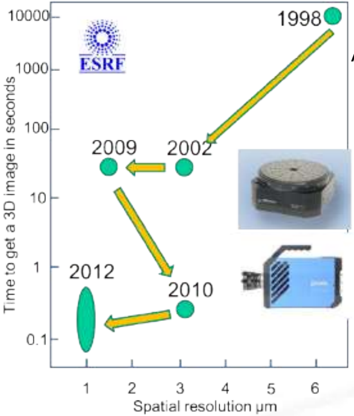
In-situ acquisition is possible in 3-D



Ultrafast 3-D imaging available at ESRF, APS, SLS

Acquisition rate depends a lot on sample (absorption, dose sensitivity, ...)

In-situ acquisition is possible in 3-D

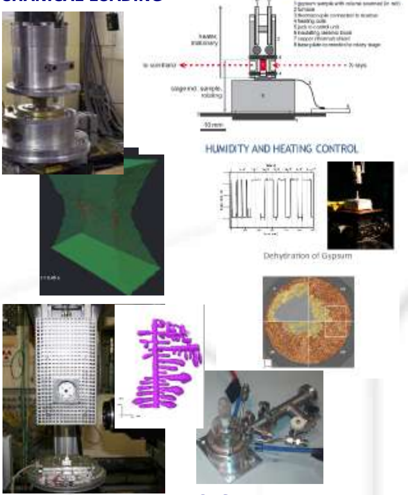


Ultrafast 3-D imaging available at ESRF, APS, SLS

Acquisition rate depends a lot on sample (absorption, dose sensitivity, ...)

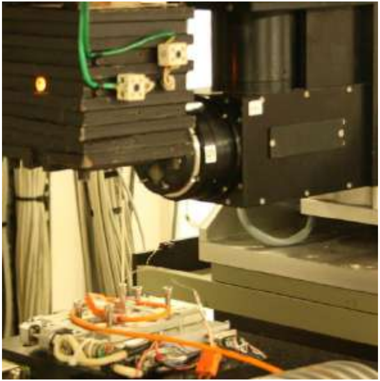
A variety of in-situ setups are available

MECHANICAL LOADING



FURNACES

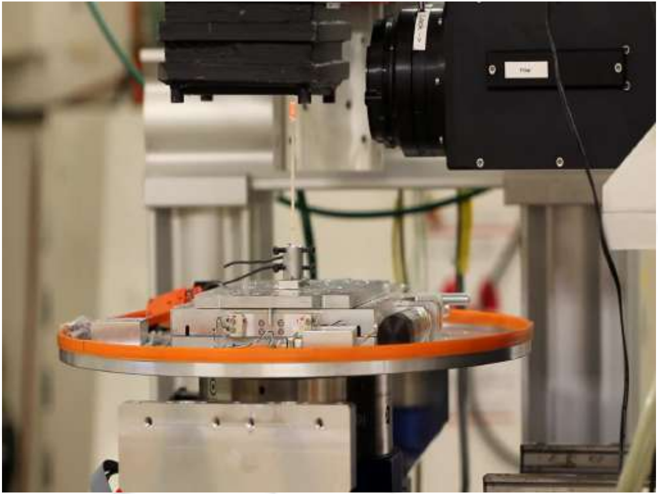
Simap furnace, 300-800°C
[Terzi et al., 2010]

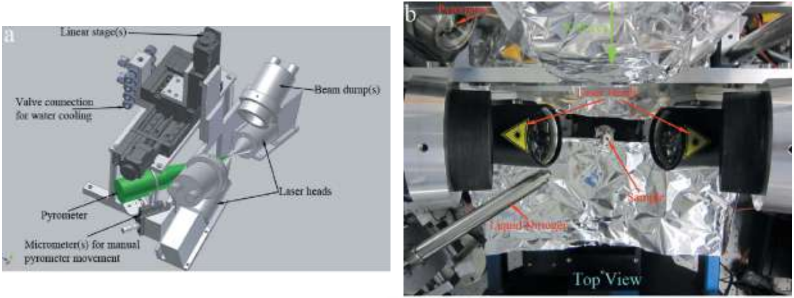


Ecole des Mines furnace, 700-1500°C
[Limodin et al., 2009]

Advantages	Drawbacks
homogeneity of temperature field	slow quenching
	bulky

Continuous rotation for ultrafast acquisition



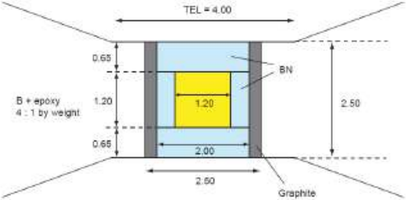


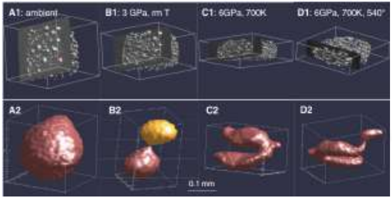
Tomcat beamline, SLS PSI [Fife et al., 2012]

Advantages	Drawbacks
fast heating	homogeneity of temperature field
minimal space requirement	

Advantages	Drawbacks
fast heating	homogeneity of temperature field
controlled atmosphere	

Argonne synchrotron (APS)





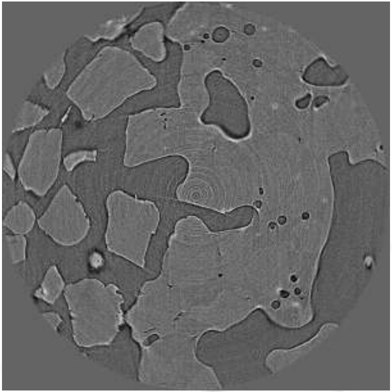
connectivity of Fe-Ni-S inclusions in a silicate (olivine) matrix

[Wang et al., 2005, Lesher et al., 2009, Wang et al., 2009, Wang et al., 2011]

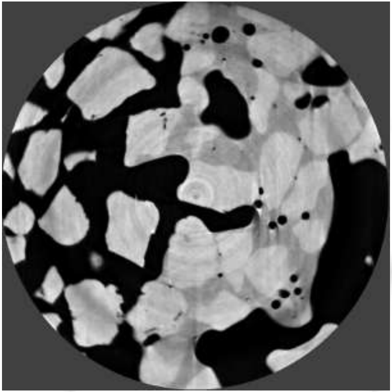
Absorption and phase contrast

Good spatial coherence @ ESRF → phase can be reconstructed

$$n = 1 - \delta + i\beta$$



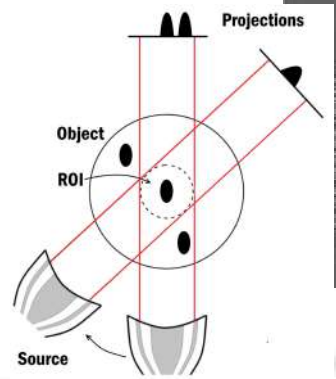
Absorption reconstruction

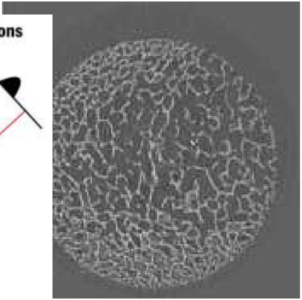


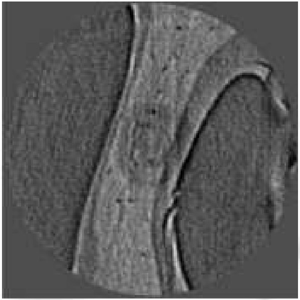
(approximate) phase reconstruction

[Paganin et al., 2002, Weitkamp et al., 2012]

Possible to zoom into the sample



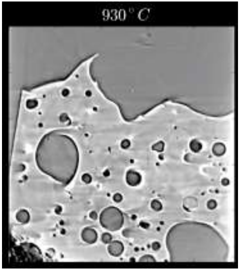


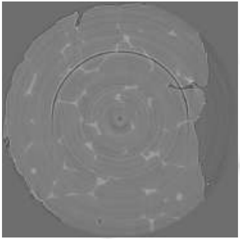


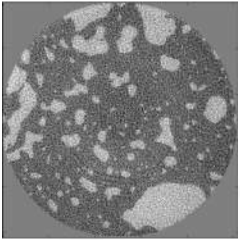
[Kyrieleis et al., 2011]

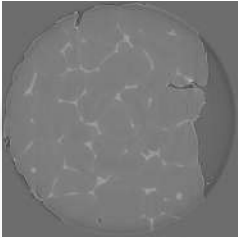
Artifacts

Multiple sources of artifacts : sample motion, sensor non-linearities, undersampling, etc.



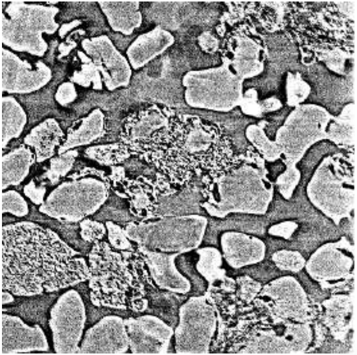






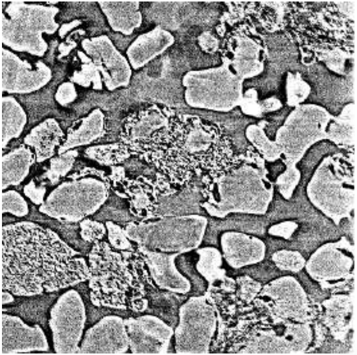
Images from Limodin, Salvo, Cloetens

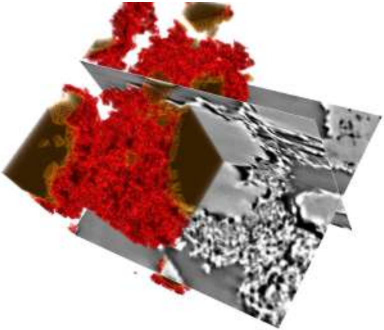
Heavy-duty image processing



- Large datasets : 100 Go for one experiment !
- Noisy images : tradeoff between speed and quality
- Longer to process the images than to acquire them !

Heavy-duty image processing





- Large datasets : 100 Go for one experiment !
- Noisy images : tradeoff between speed and quality
- Longer to process the images than to acquire them !

Outline

1 In-situ tomography

2 Applications

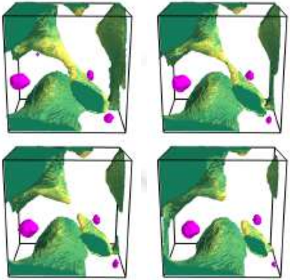
Phase separation

Glass reactive melting : reaction paths between granular raw materials

Advantages of (in-situ) X-ray tomography

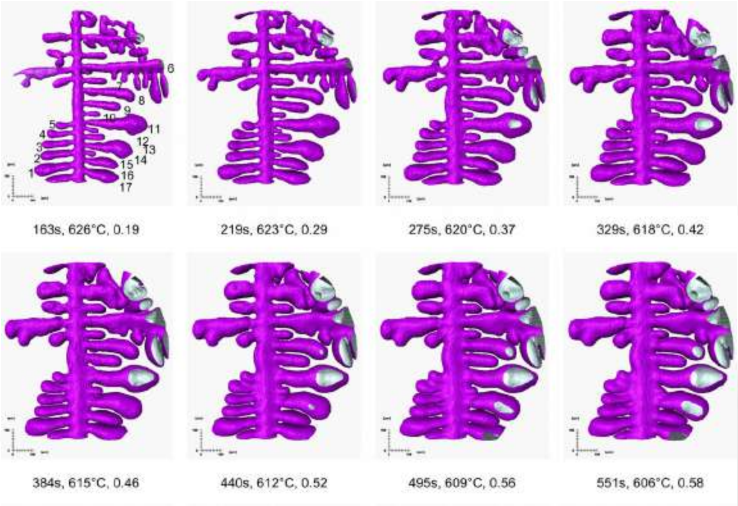
- 3-D images ⇒ connectivity, topology
- more statistics on objects/particles
- non-destructive : follow the same sample
- time resolution : don't miss important events

[Watson and Roberts, 2011]



[Bouttes et al., 2015]

Tomography has been used for a long time in metallic alloys



Mechanisms of dendritic growth in Al-Cu [Limodin et al., 2009]

Outline

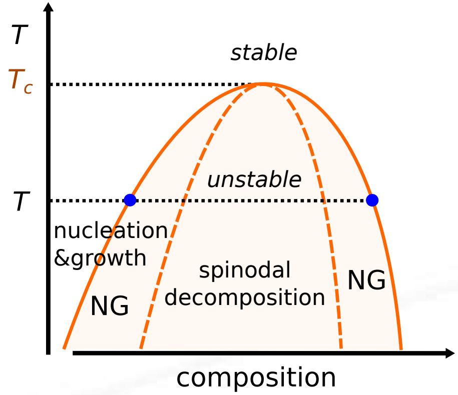
1 In-situ tomography

2 Applications

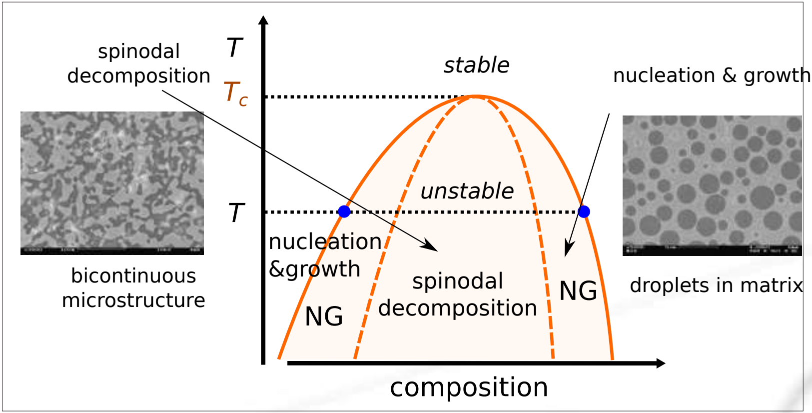
Phase separation

Glass reactive melting : reaction paths between granular raw materials

Principles of phase separation

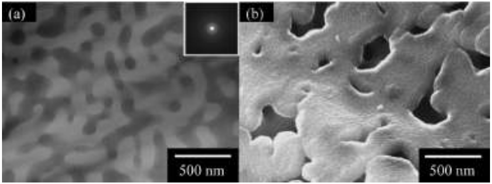


Principles of phase separation



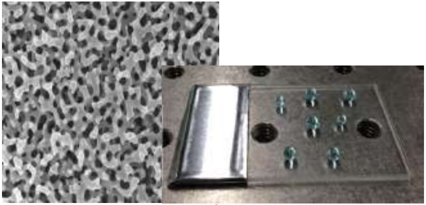
- No 1-to-1 correspondence between thermodynamic regime (SD, NG) and microstructure of phases
- Silicate melts : phase separation possible in stable or metastable liquids

Porous membranes: Vycor



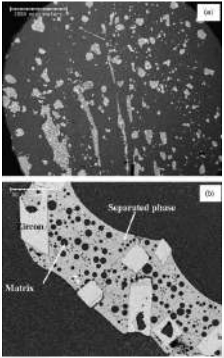
Suzuki et al. 2008

Super-hydrophobic porous films



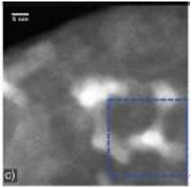
Aytug et al. 2013

Nuclear waste glasses



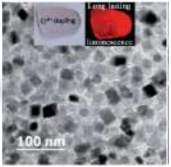
Martineau et al. 2010

Dargaud et al . 2012



Glass ceramics

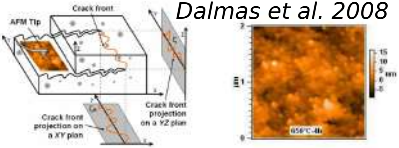
Luminescent glass



Chenu et al. 2014

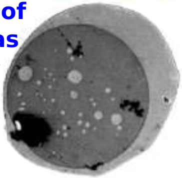
Model materials for crack propagation

Dalmas et al. 2008

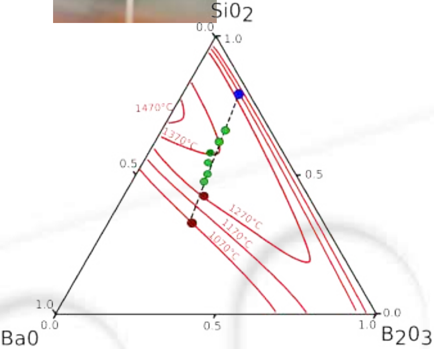


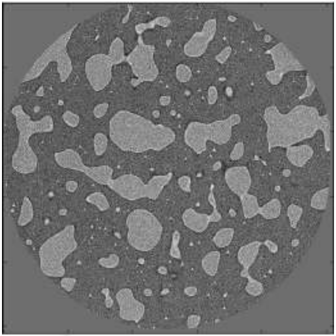
Microstructure of basaltic magmas

Veksler et al. 2007



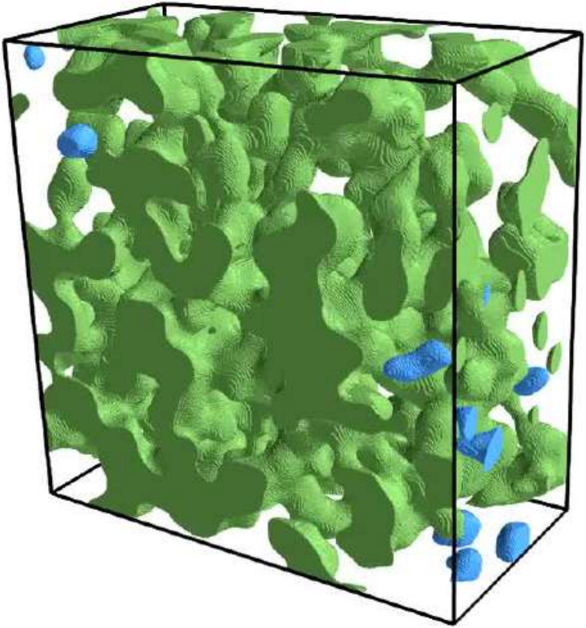
- pink beam 35 keV (barium is very absorbing !)
- pixel size 0.55 or 1.1 μm
- acquisition times 5s - 1min
- local (ROI) tomography
- temperatures : 1000 - 1300° C



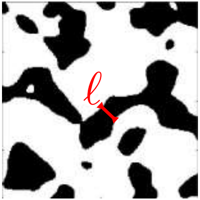


FOV : 2mm

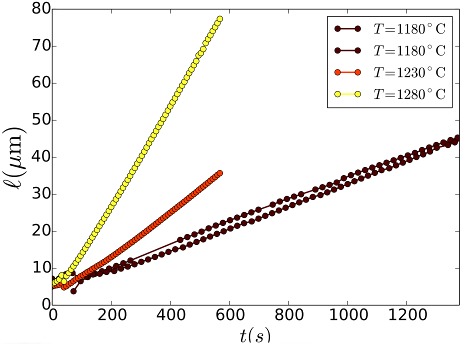
Coarsening : $\phi \leq 0.5$ case



Evolution of characteristic length



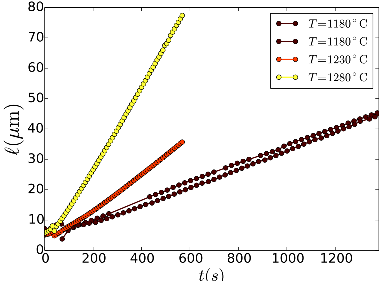
$$\ell = \frac{\mathcal{V}}{\mathcal{S}}$$

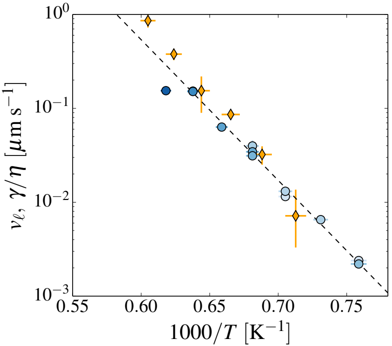


- linear evolution with time
- coarsening rate increases with temperature

⇒ is hydrodynamic flow the dominant mechanism ?

Evolution of characteristic length



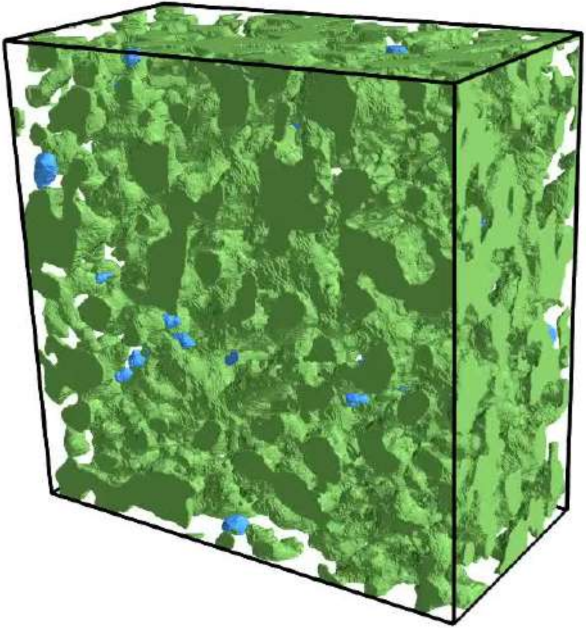


Evolution consistent with viscous coarsening :

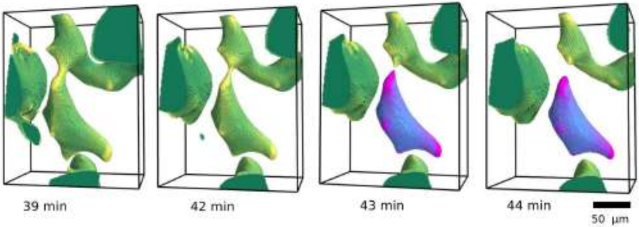
$$\dot{\ell}(t) \simeq \frac{\gamma}{\eta}$$

[Bouttes et al., 2014, Bouttes et al., 2015]

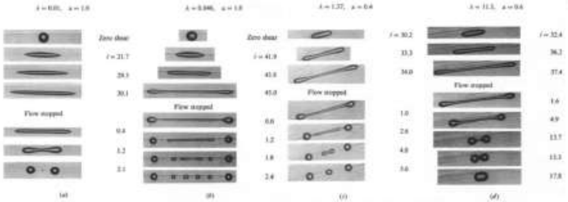
Coarsening and fragmentation



Close-up on domain break-up



Only the barium-rich phase breaks up in domains

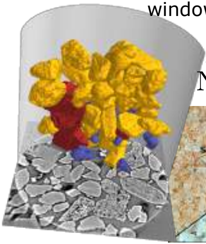


- Viscous filaments tend to retract
- Less viscous filaments tend to break

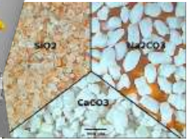
[Stone and Leal, JFM 1989]

1 In-situ tomography

2 Applications
Phase separation
Glass reactive melting : reaction paths between granular raw materials



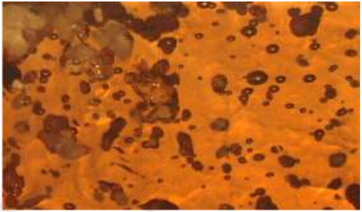
window glass raw materials:
66% silica sand
33% carbonates
Na_2CO_3 $CaCO_3$

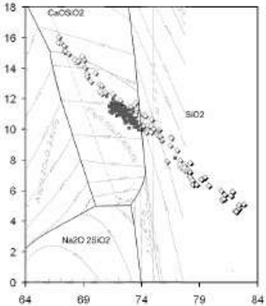


grain sizes ~ 100s of microns

$$SiO_2 + Na_2CO_3 + CaCO_3 \text{ " } \rightarrow \text{ " } (SiO_2, Na_2O, CaO) + CO_2$$

Reaction paths and geometry ?



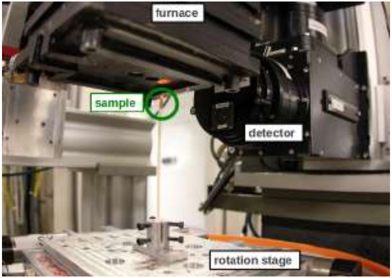


[Chopinet et al., 2010]
Large calcium carbonate grains lead to poor quality.

Why ?

Tomographic imaging of glass melting

ID19, PCO camera



- Pink beam 19 keV
- Tomo in 1-4 s
- Pixel size of 1.1 μm
- Samples of a few mg
- Furnace : T $\leq 1500^\circ$ C

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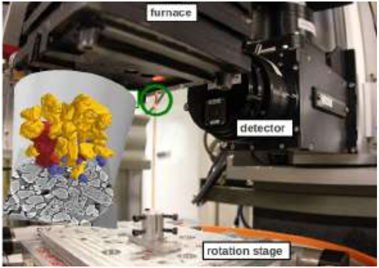

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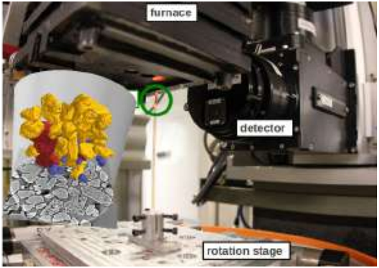

lab tomograph (RX Solutions)

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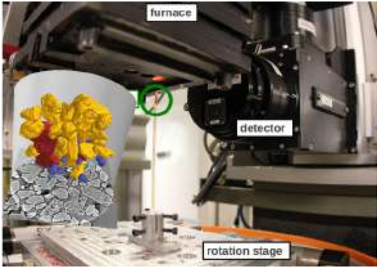

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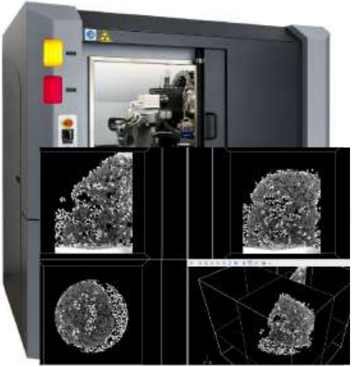
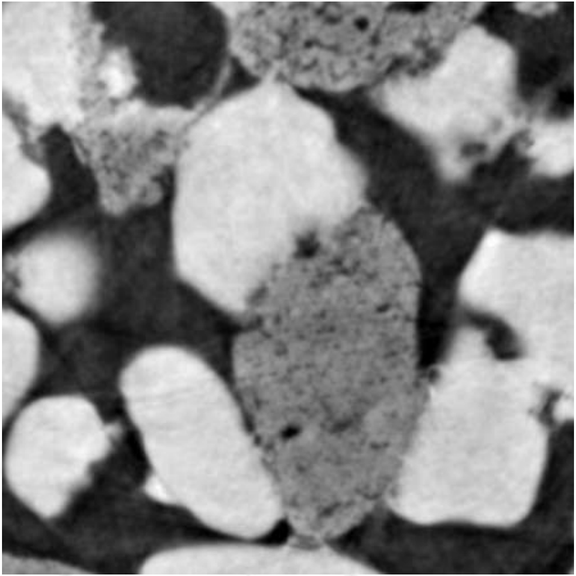


- Polychromatic beam
- Tomo in one hour or more
- Pixel size $\geq$ 5 microns
- Samples of a few grams

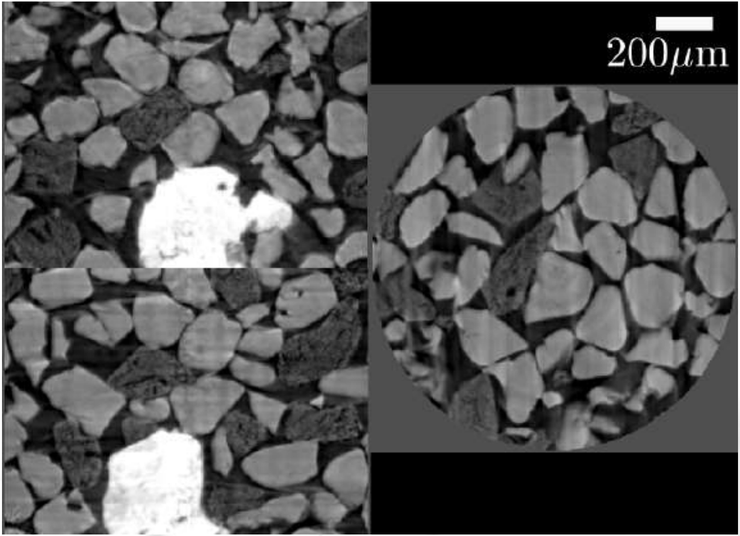
Geometry of solid-state reactions



Binary mixture of SiO_2 - Na_2CO_3, 800°C

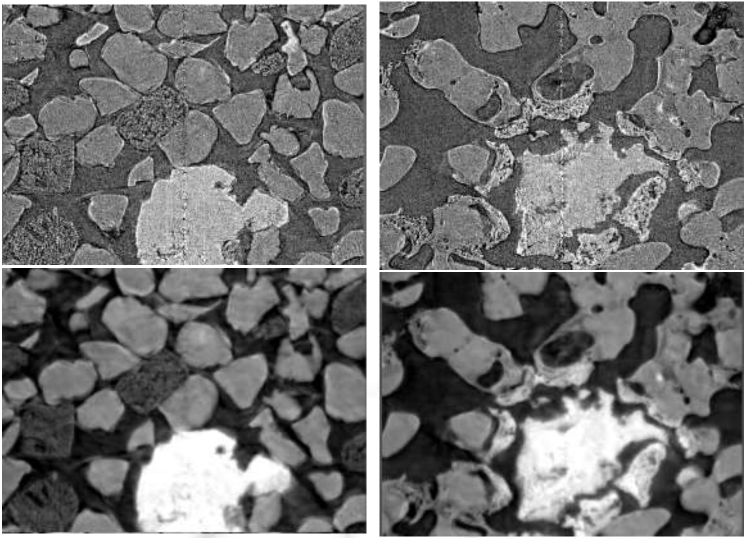
[Gouillart et al., 2012], [Grynberg et al., 2015]

The fate of calcium carbonate

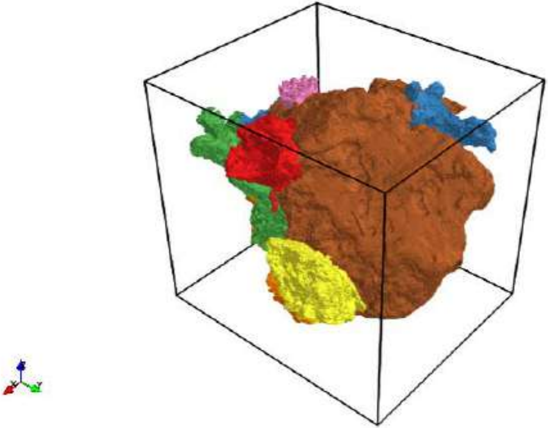


Fast heating at 900°C, ternary mixture of SiO_2, Na_2CO_3, $CaCO_3$

The fate of calcium carbonate

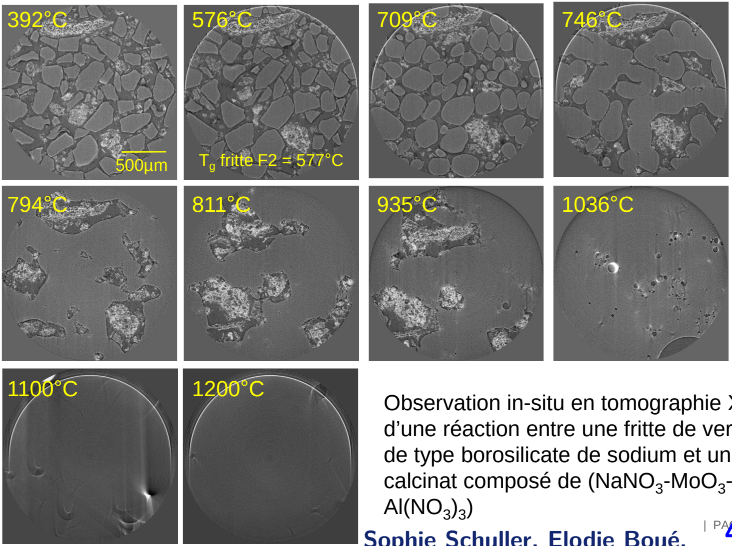


Only outer parts of the grain react
Core of CaO stays unreacted for a long time
Delay in the formation of molten silicates



Quantifying the reacted parts of the grain

Observation de l'élaboration d'un verre nucléaire de composition simplifiée – microtomographie in situ



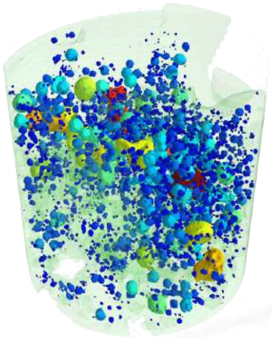
Observation in-situ en tomographie X d'une réaction entre une fritte de verre de type borosilicate de sodium et un calcinat composé de ($NaNO_3$-MoO_3-$Al(NO_3)_3$)

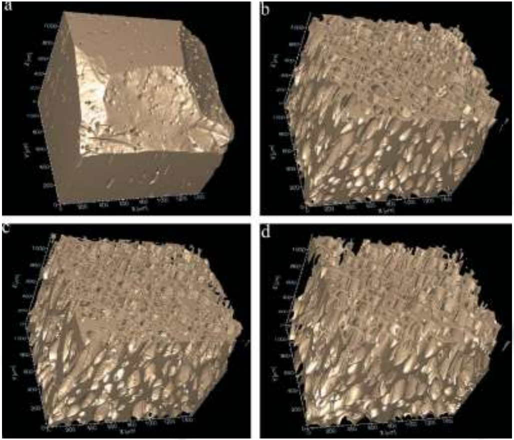
Sophie Schuller, Elodie Boué,

CEA Marcoule

Bubbles and vesicularity

Good contrast between gas and glass/melt

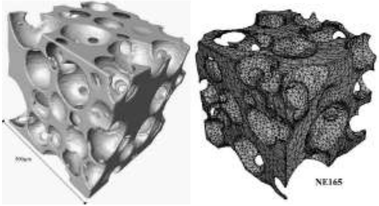




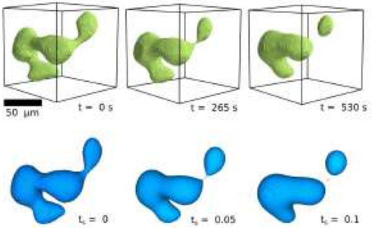
[Fife et al., 2012] : fast heating at 15 $K.s^{-1}$ of obsidian glass in range $800 - 1000°C$.

Ex-solution of water from the melt generates bubbles.

Combination with 3-D modelling

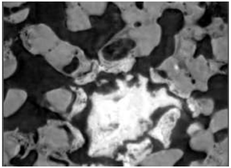


Mechanics : finite-element simulation on mesh determined from tomography
[Youssef et al., 2005]



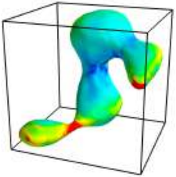
Estimation of surface tension thanks to hydrodynamic simulations
[Bouttes et al., 2015]

Conclusions



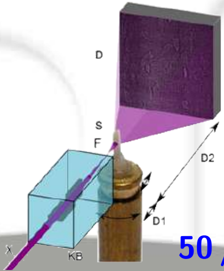
In-situ imaging

- Whole evolution for one sample
- Capture turning points



3D imaging

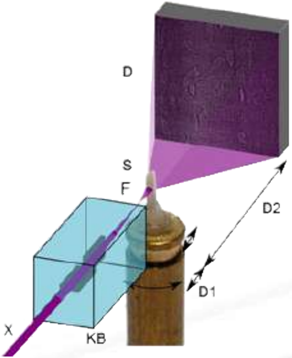
- 3-D information : connectivity, topology
- Whole sample : don't miss where the action is taking place
- More statistics



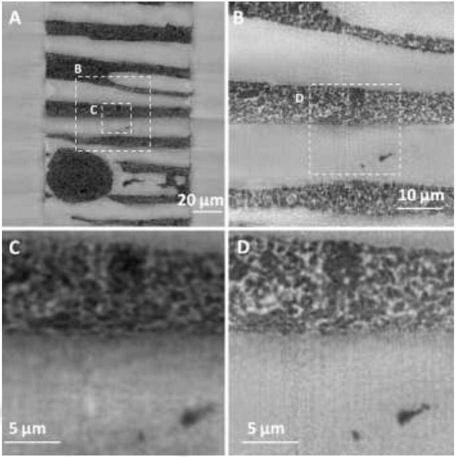
Challenges

- Spatial resolution
- Realistic sample environment
- Data processing
- Link/combine w/ other techniques

Nanotomography



ESRF, ID16a



[Villanova et al., 2014]

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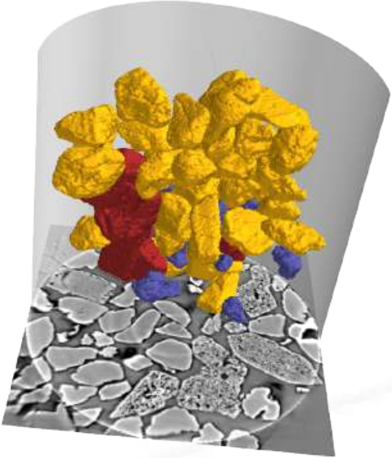
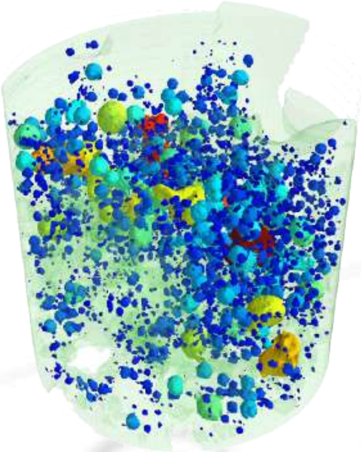
- Funding agencies : Saint-Gobain Recherche, CNRS INP, ANR project EDDAM
- Beamtime : ESRF projects HD501, SC3724, MA1839 and MA1876

Thank you for your attention !





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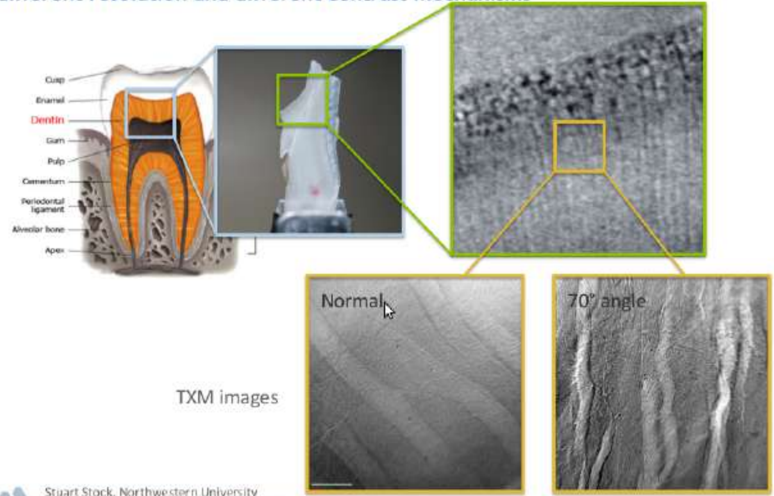
Data Fusion Study of Mineralized Tissue

Integration of data from instruments with different resolution and different contrast mechanisms



Stuart Stock, Northwestern University