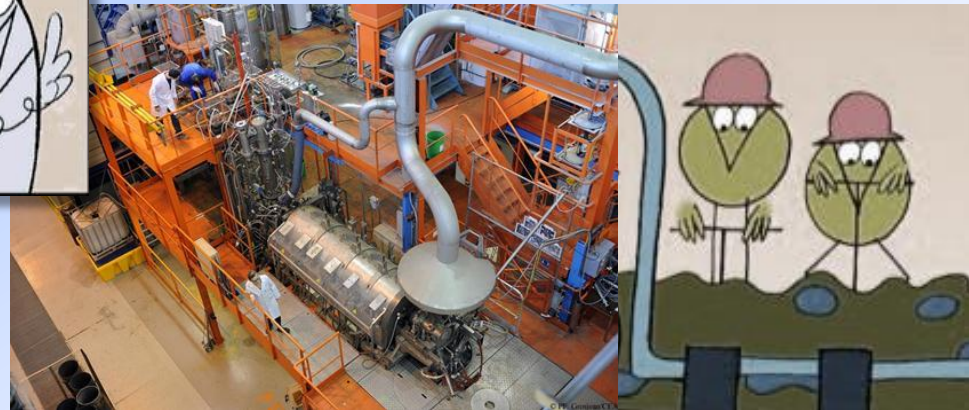
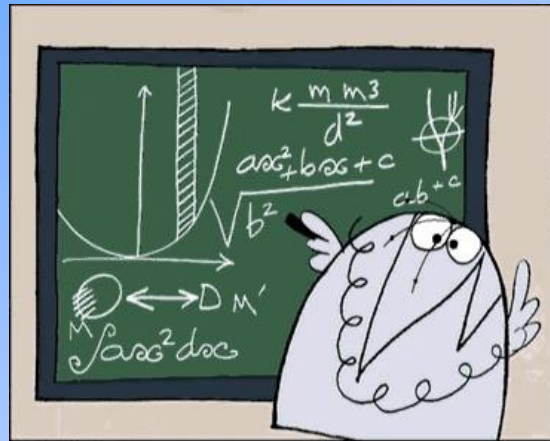


Modélisation du comportement rhéologique de verres nucléaires fondus contenant des agrégats de platinoïdes



2010-2023



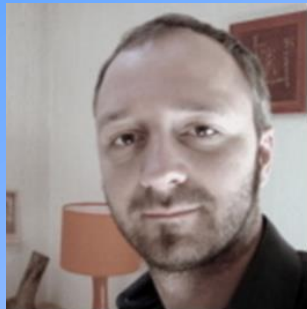
Cécile



Phil



Muriel



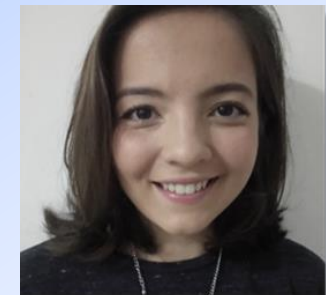
Jean-Philippe



Jean



Caroline



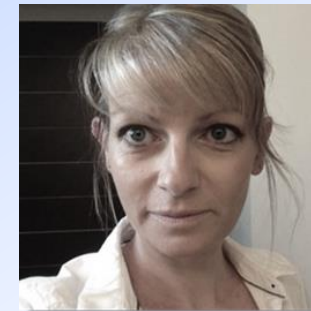
Norma



Aurélien



Olivier

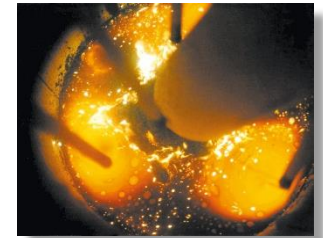
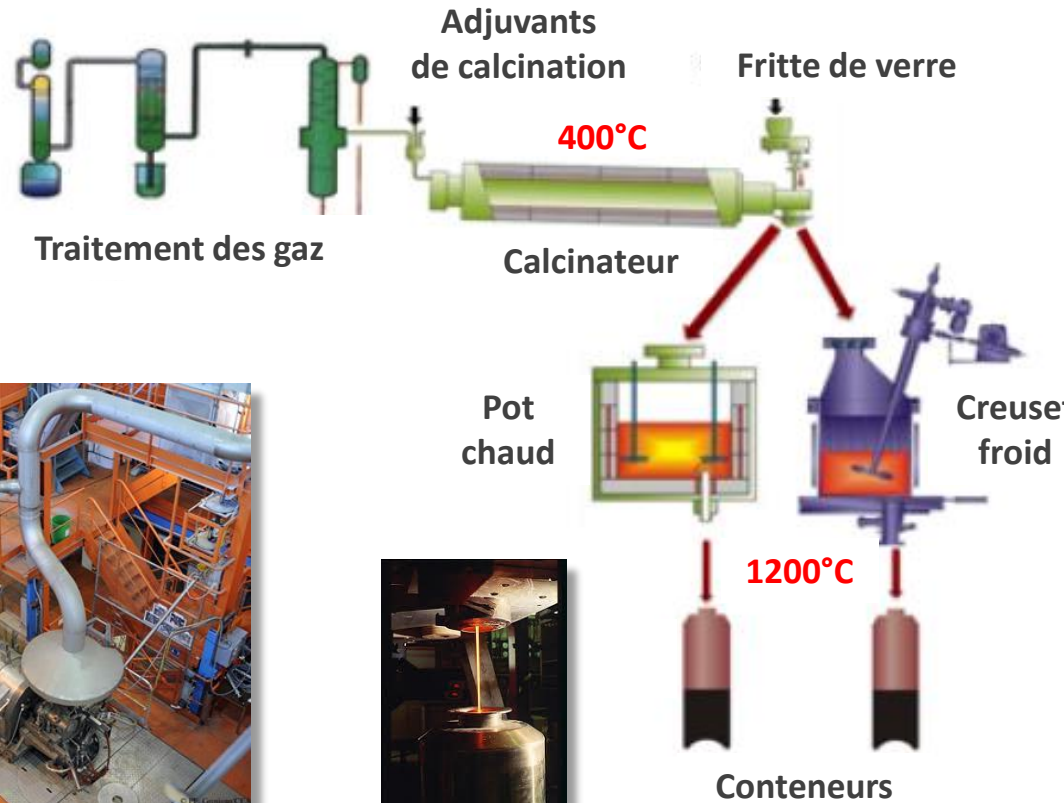


Isabelle



Elise

Vitrification des déchets de haute activité



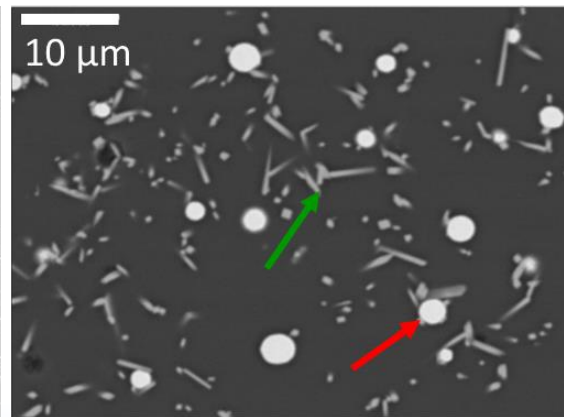
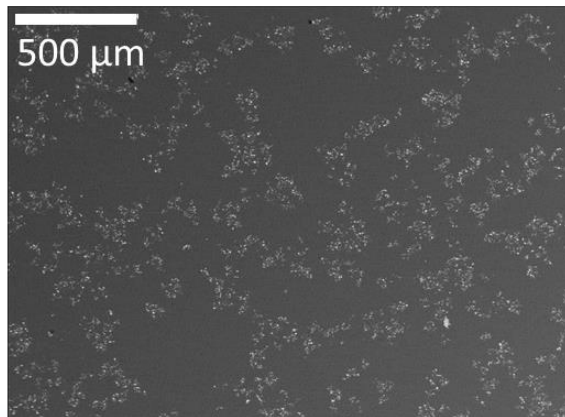
Stockage géologique
Comportement
à long terme



Le verre nucléaire et les platinoïdes

	wt% Min.	wt% Max.
SiO ₂	42.4	51.7
Al ₂ O ₃	3.6	6.6
B ₂ O ₃	12.4	16.5
Na ₂ O	8.1	11.0
FP* + Act* + Ptds + ZrO ₂	4.2	18.5
RuO ₂ + Pd + Rh	0	3.0

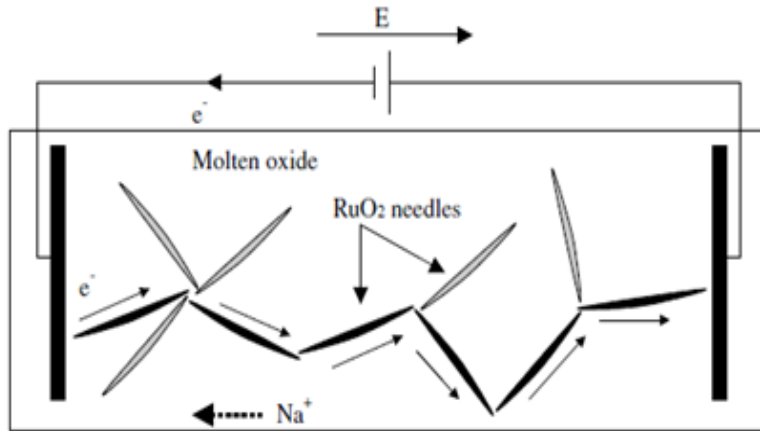
*FP: Produits de Fission; Act: Actinides Ptds : platinoïdes insolubles dans le verre



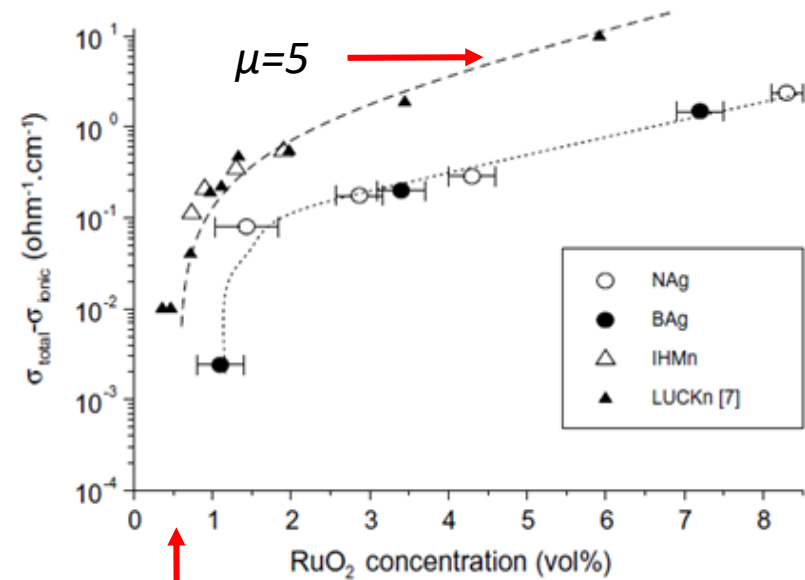
Platinoïdes
Billes de Pd-Te
 (2-5μm)
Aiguilles de RuO₂
 (1x10-20μm)

Regroupements et agrégation de particules

Le verre nucléaire et les platinoïdes

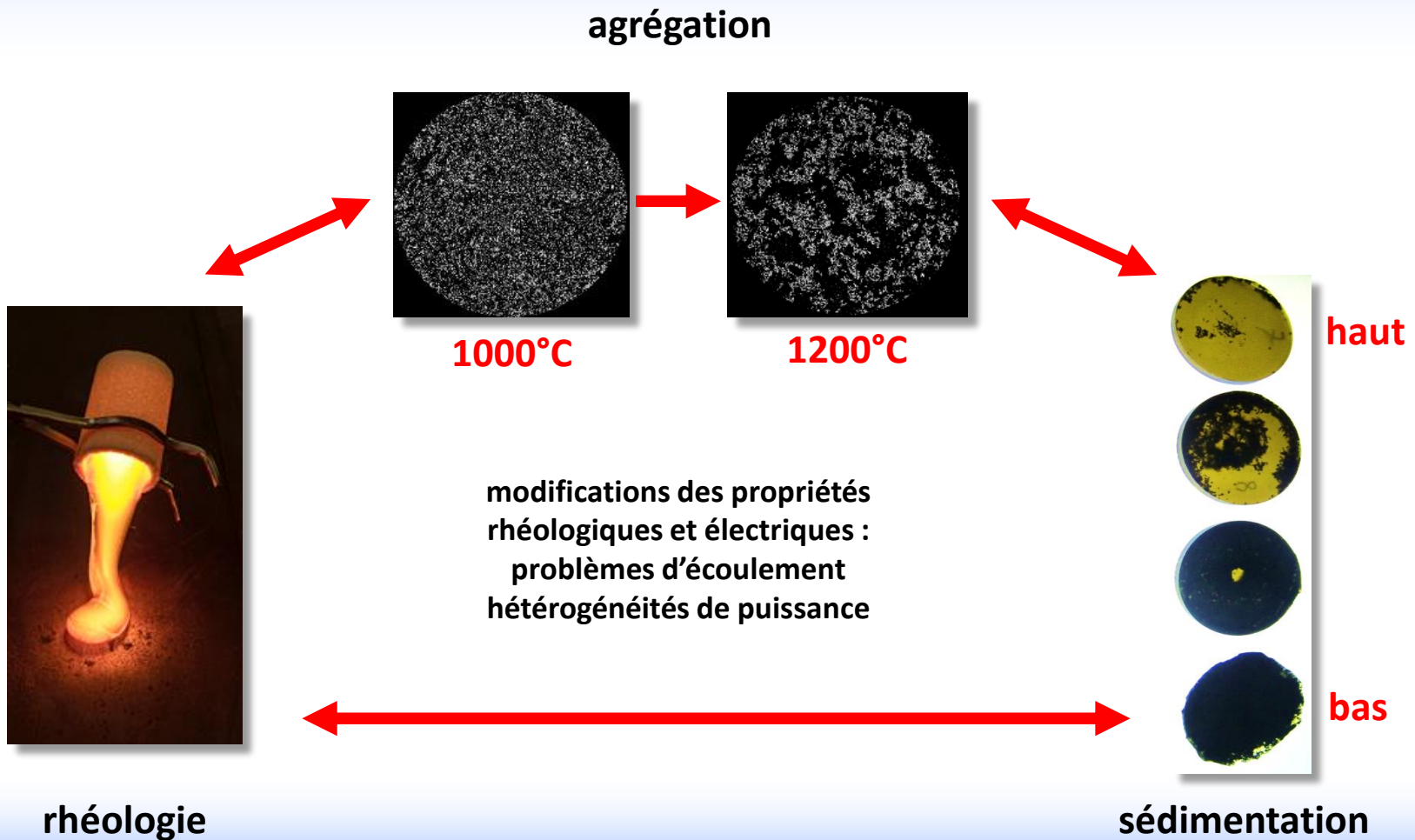


$$\sigma_e = \sigma_{e0} (x - x_c)^\mu$$



$$x_c = 0.5 \pm 0.2 \text{ vol}$$

Le verre nucléaire et les platinoïdes

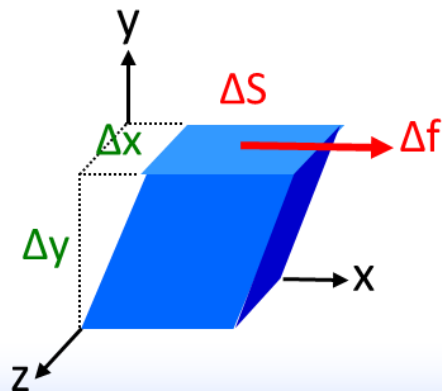


Rhéométrie à haute température

$$\tau = \Delta f / \Delta S$$

$$\gamma = \Delta x / \Delta y$$

$$\dot{\gamma} = \partial \gamma / \partial t$$

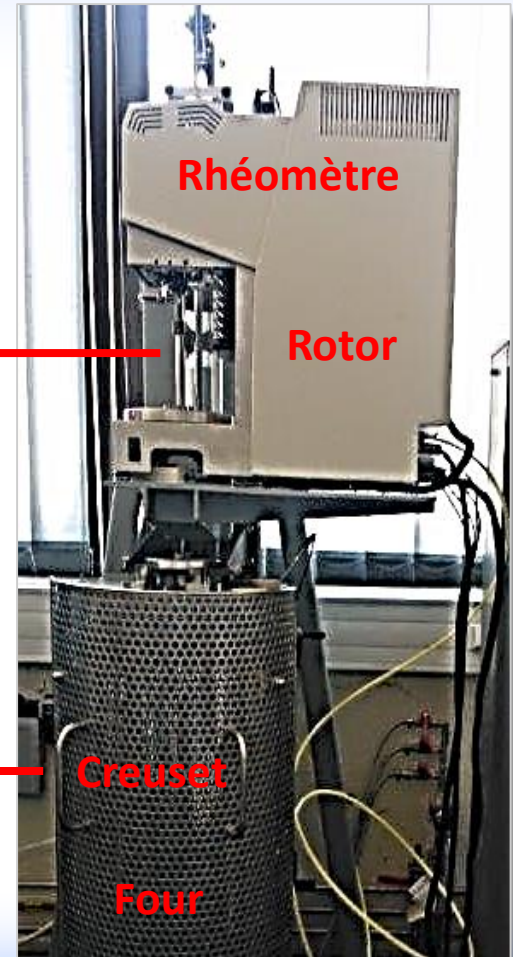


$$\tau = K_{\tau} C$$

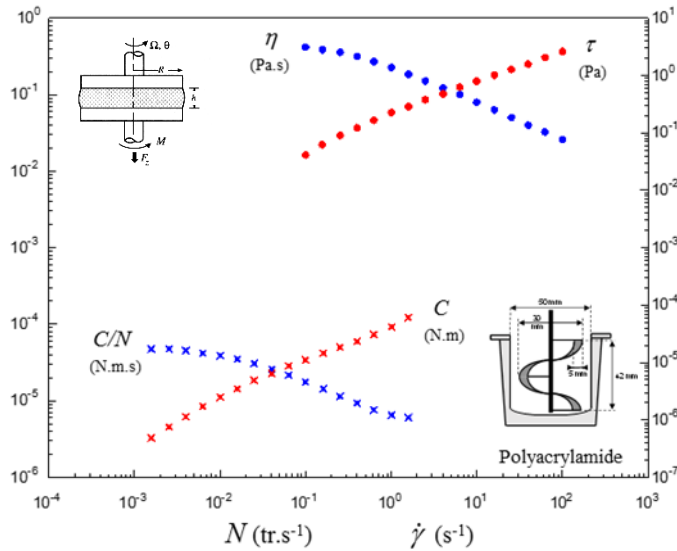
$$\dot{\gamma} = K_{\dot{\gamma}} \dot{\theta}$$

C

$$\dot{\theta} = 2\pi N$$



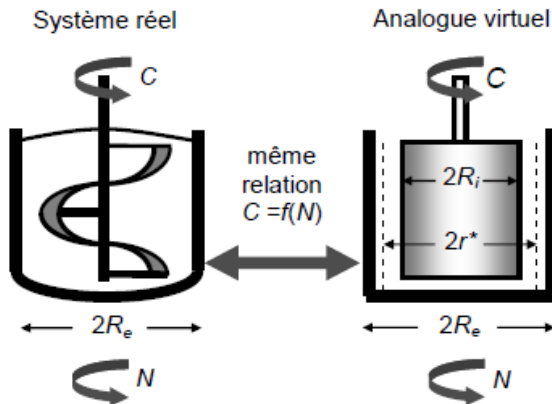
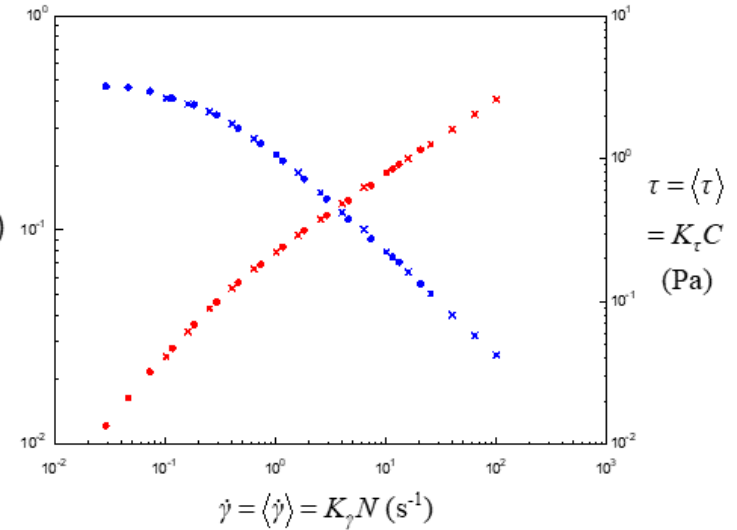
Analogie de Couette



$$C \propto \tau \quad N \propto \dot{\gamma}$$

$$C/N \propto \eta$$

$$\eta = \langle \eta \rangle = \frac{K_\tau}{K_\gamma} (C/N) \text{ (Pa.s)}$$

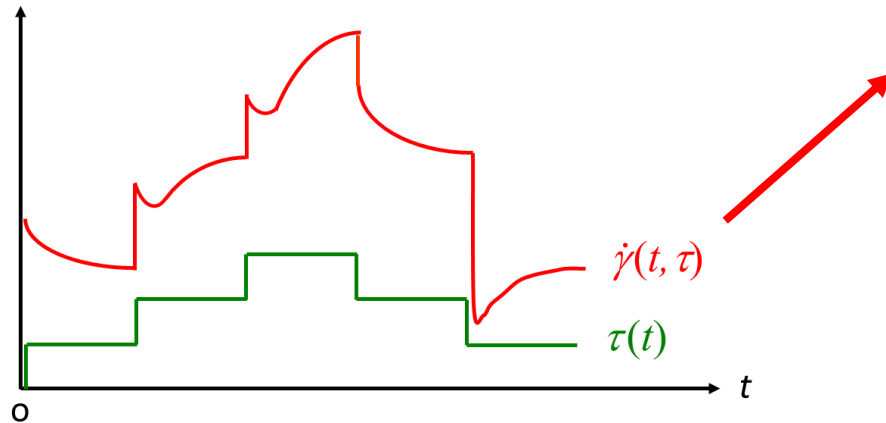
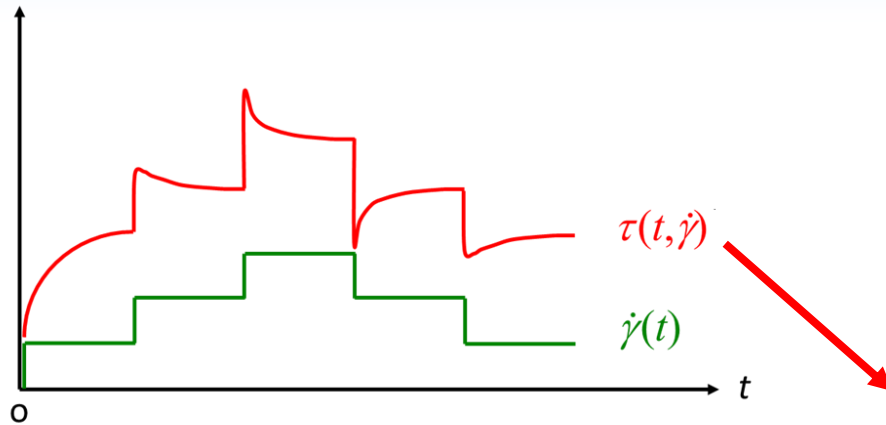


$$K_\tau = \frac{\tau}{C} = \frac{1}{2\pi L r^2} \quad K_\gamma = \frac{\dot{\gamma}}{N} = \frac{(4\pi n)(R_e/r)^{2/n}}{(R_e/R_i)^{2/n} - 1}$$

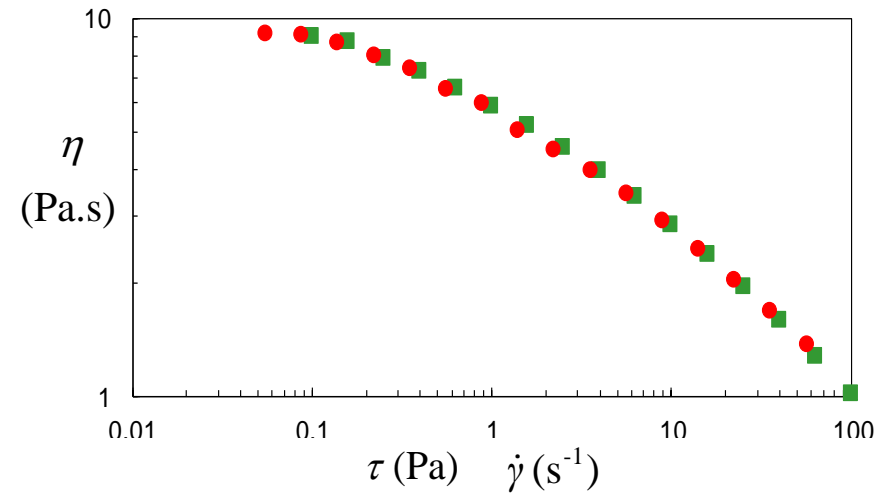
$$\langle \tau \rangle = K_\tau C \quad \Rightarrow \quad \langle \eta \rangle = \frac{\langle \tau \rangle}{\langle \dot{\gamma} \rangle} = \frac{K_\tau C}{K_\gamma N} = \frac{K_\tau}{K_\gamma} (C/N)$$

$$\langle \dot{\gamma} \rangle = K_\gamma N$$

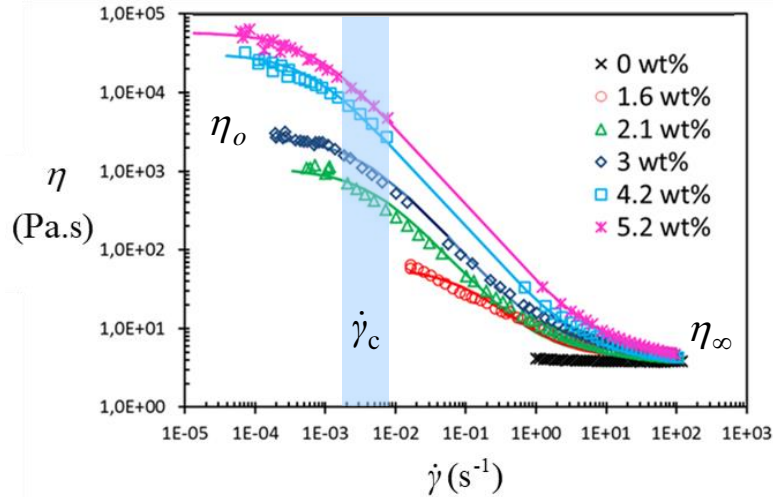
Comportement rhéologique



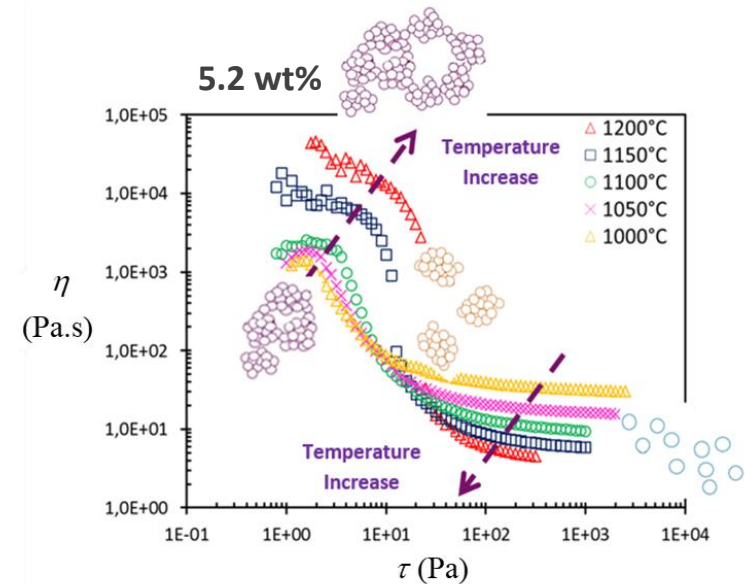
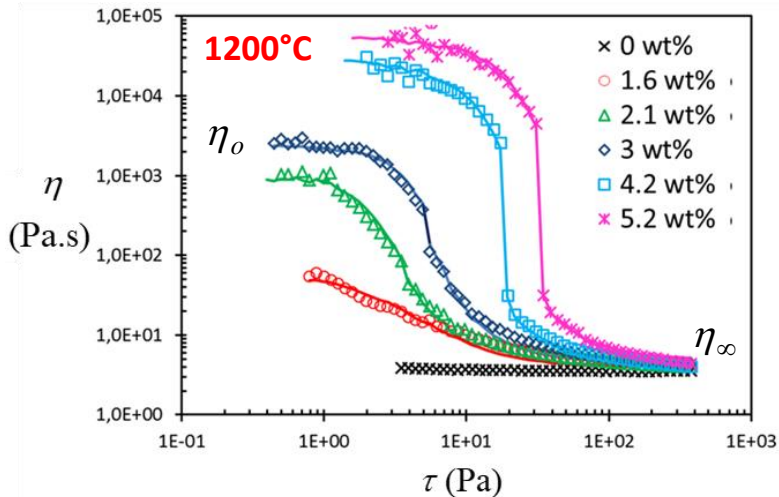
$$\eta(t, \dot{\gamma}) = \tau / \dot{\gamma}$$



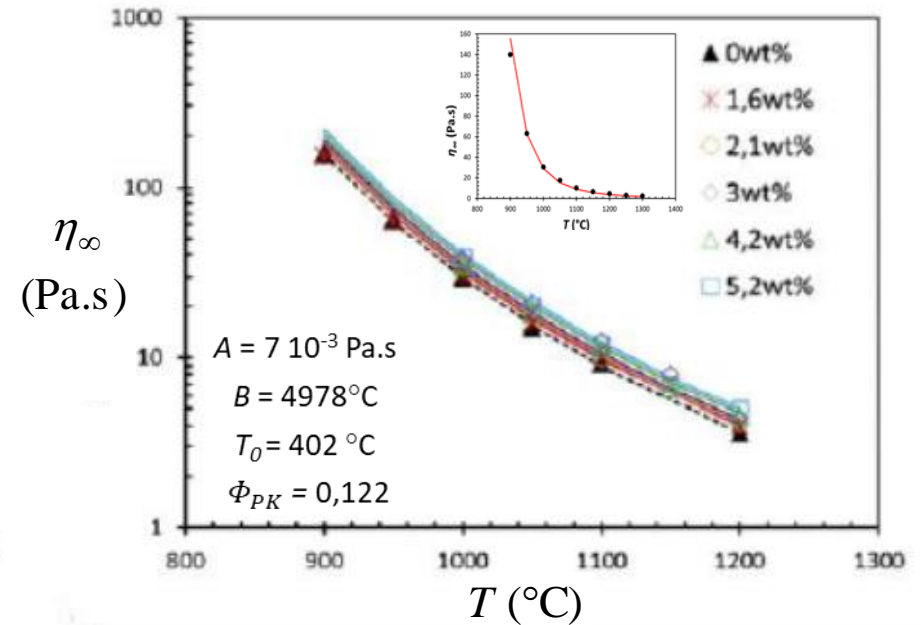
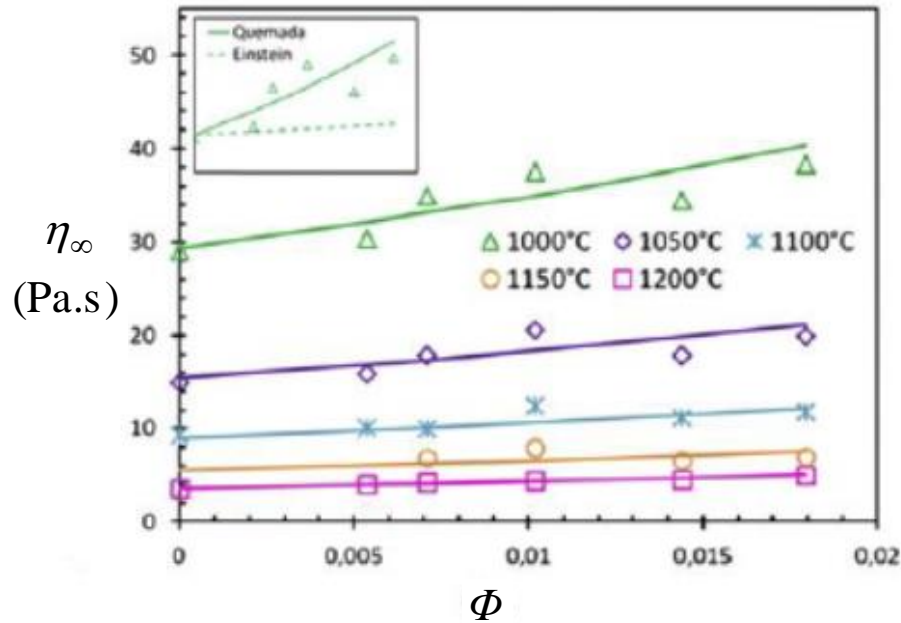
Régime permanent



$$\eta = \eta_\infty + \frac{\eta_0 - \eta_\infty}{1 + \dot{\gamma} / \dot{\gamma}_c}$$



Régime permanent - Haute vitesse



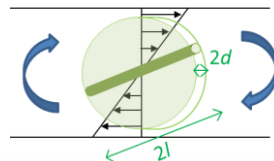
$$\eta_{\infty} = \eta_f \left(1 - \frac{\Phi_{eff}}{\Phi_m} \right)^{-2}$$

$$\Phi_{eff} \cong \Phi \frac{l}{d}$$

$$\eta_f = A \exp \left(\frac{B}{T - T_0} \right)$$

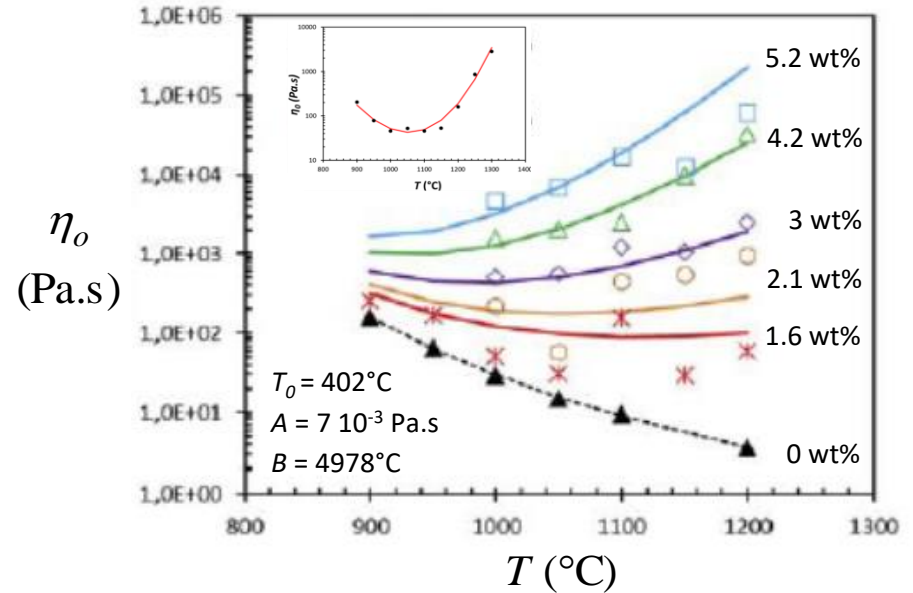
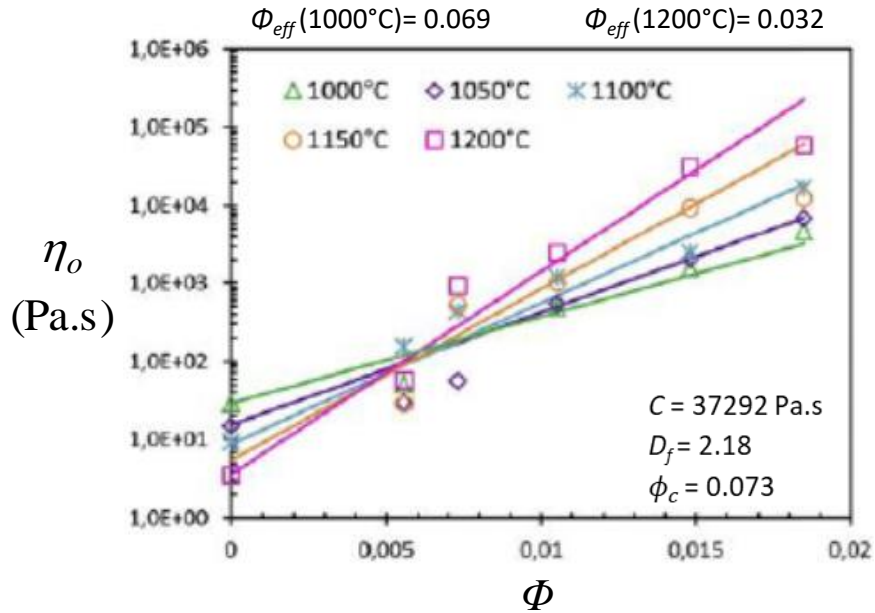
$$\Phi_{eff}(1000^{\circ}\text{C}) = 0.069 \quad \Phi_{eff}(1200^{\circ}\text{C}) = 0.032$$

$$\eta_{\infty} = \eta_f \left(1 - \frac{\Phi}{\Phi_{PK}} \right)^{-2}$$



$$\eta_{\infty} = A \exp \left(\frac{B}{T - T_0} \right) \left(1 - \frac{\Phi}{\Phi_{PK}} \right)^{-2}$$

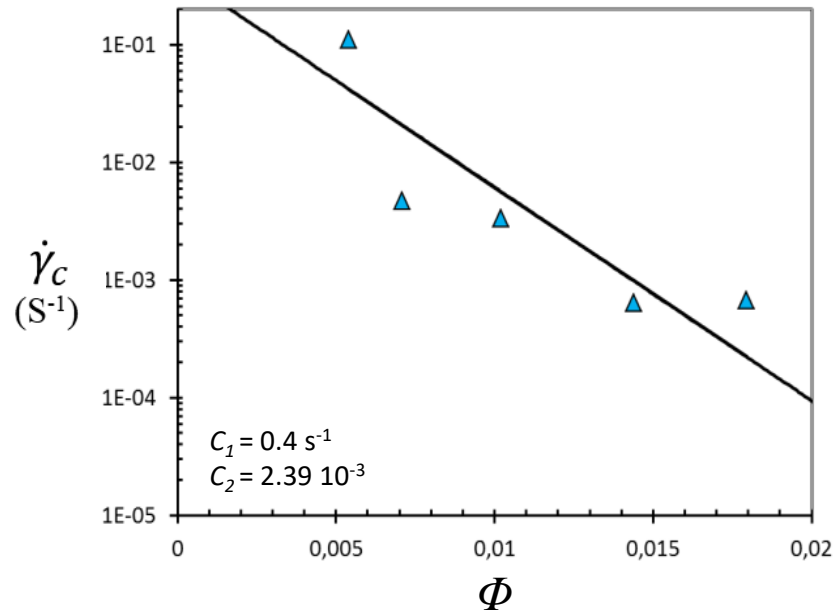
Régime permanent - Basse vitesse



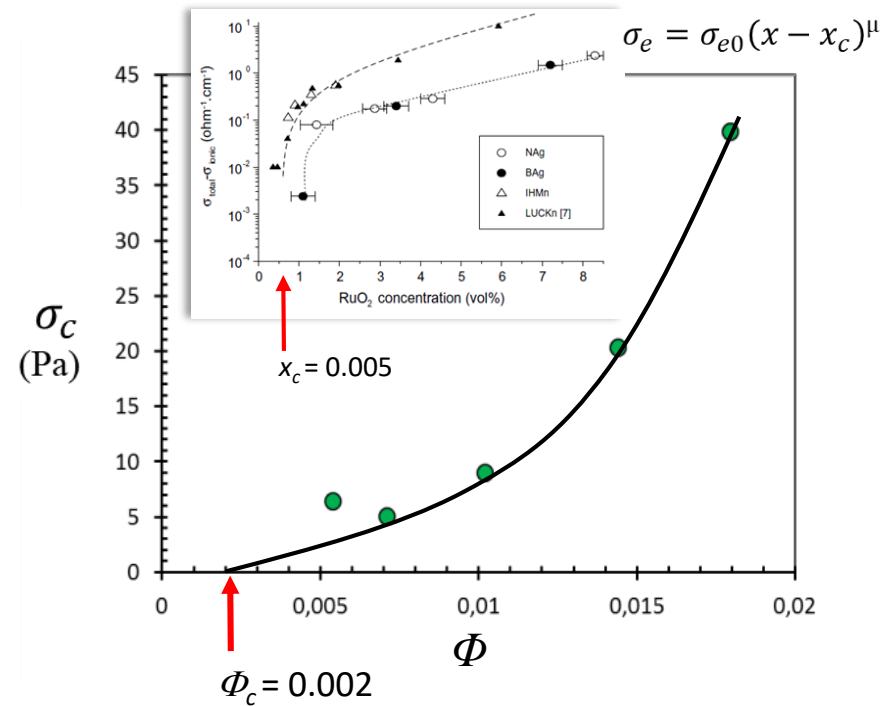
$$\Phi_{eff} = \Phi \left(\frac{R}{a} \right)^{3-D_f} \quad R = a \left(\frac{\Lambda}{a12h^2\eta_f\dot{\gamma}} \right)^{1/2} = a \left(\frac{C}{\eta_f} \right)^{1/2} \quad \Phi_{eff} = \Phi \left(\frac{C}{\eta_f} \right)^{3-D_f/2}$$

$$\eta_0 = \eta_f \exp \left(\frac{\phi_{eff}}{\phi_c} \right) = \eta_f \exp \left[\frac{\phi}{\phi_c} \left(\frac{C}{\eta_f} \right)^{3-D_f/2} \right] = A \exp \left(\frac{B}{T-T_0} \right) \exp \left[\frac{\phi}{\phi_c} \left(\frac{C}{A \exp \left(\frac{B}{T-T_0} \right)} \right)^{3-D_f/2} \right]$$

Régime permanent - Vitesse critique

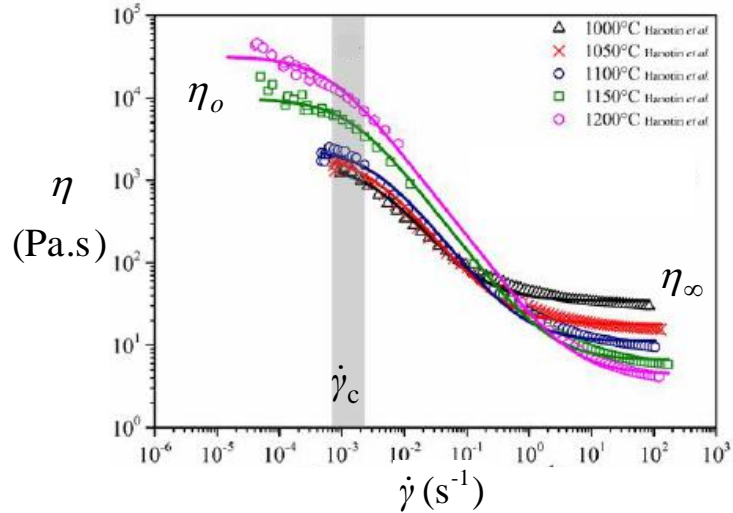


$$\dot{\gamma}_c = C_1 \exp\left(-\frac{\Phi}{C_2}\right)$$



$$\sigma_c = \sigma_{c0}(\Phi - \Phi_c)^\beta$$

Régime permanent - Modèle final

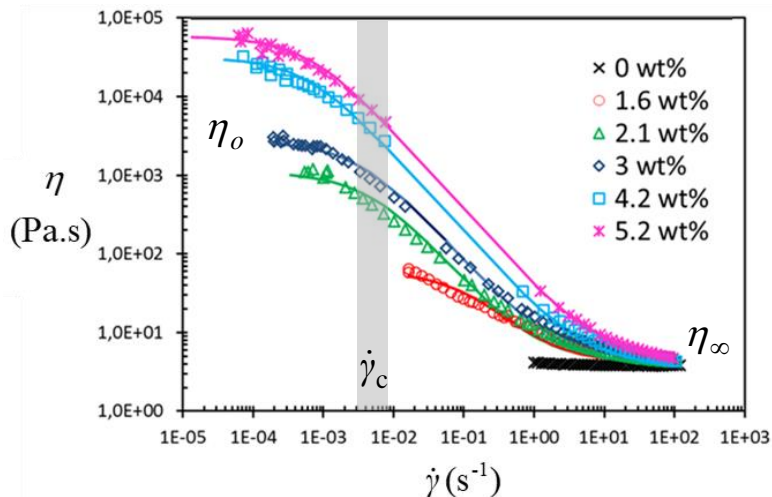


$$\eta(T, \phi, \dot{\gamma}) = \eta_{\infty}(T, \phi) + \frac{\eta_0(T, \phi) - \eta_{\infty}(T, \phi)}{1 + \frac{\dot{\gamma}}{\dot{\gamma}_c(\phi)}}$$

$$\eta_0 = A \exp\left(\frac{B}{T-T_0}\right) \exp\left[\frac{\phi}{\phi_c} \left(\frac{C}{A \exp\left(\frac{B}{T-T_0}\right)}\right)^{3-D_f/2}\right]$$

$$\eta_{\infty} = A \exp\left(\frac{B}{T-T_0}\right) \left(1 - \frac{\phi}{\phi_{PK}}\right)^{-2}$$

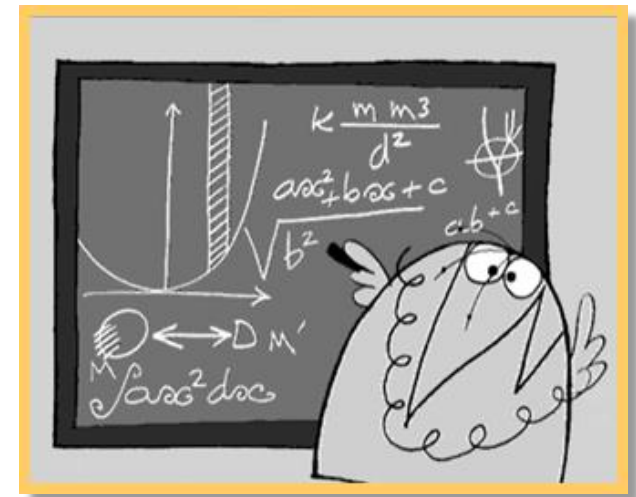
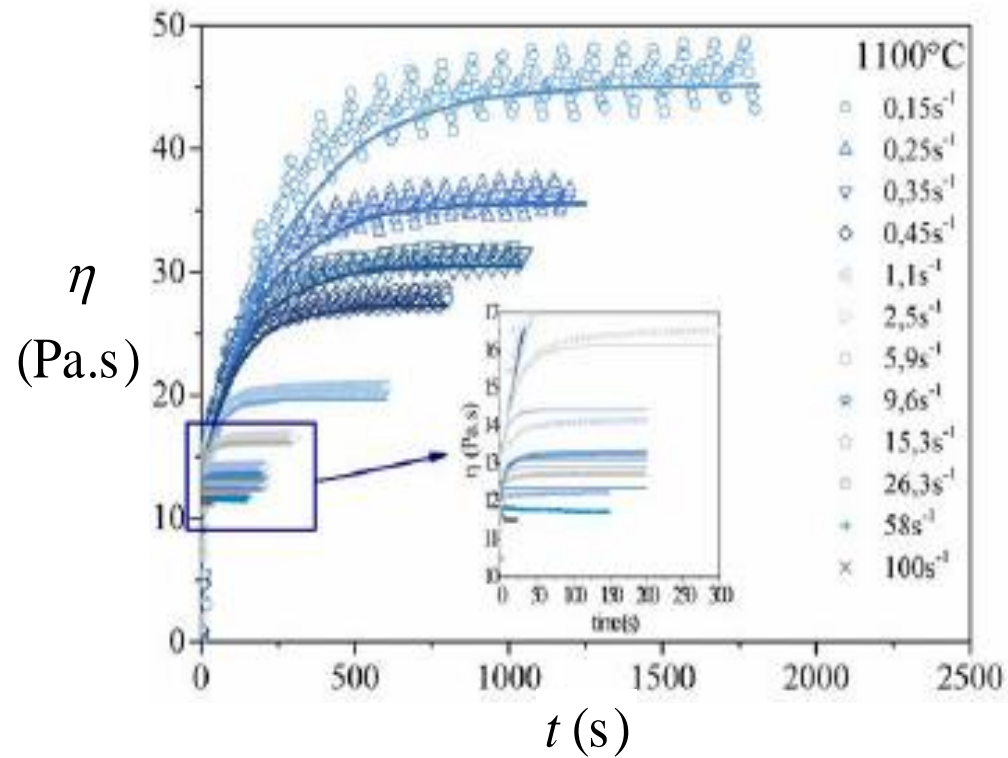
$$\dot{\gamma}_c = C_1 \exp\left(-\frac{\phi}{C_2}\right)$$



$A = 7 \cdot 10^{-3} \text{ Pa.s}$	$B = 4978^\circ\text{C}$	$C = 37292 \text{ Pa.s}$
$D_f = 2,18$	$\phi_c = 0,073$	$\phi_{PK} = 0,122$
$C_1 = 0,4 \text{ s}^{-1}$	$C_2 = 2,39 \cdot 10^{-3}$	$T_0 = 402^\circ\text{C}$

Régime transitoire

$$\eta = \eta(T, \phi, \dot{\gamma}, t) = ?$$



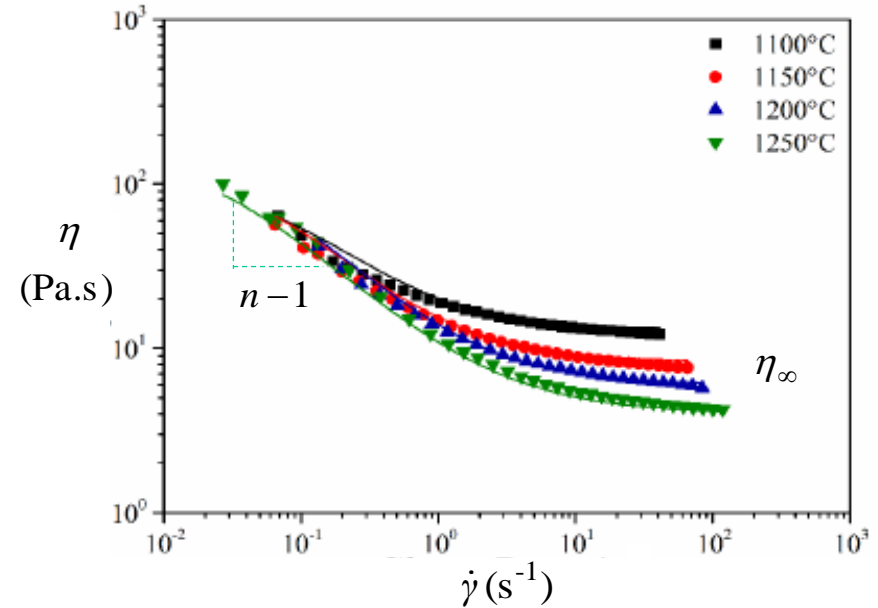
Modélisation du régime transitoire

$$\eta = \eta_{\infty} + k\dot{\gamma}^{n-1}$$

$$\eta = \eta_{\infty} + \lambda K_0 \dot{\gamma}^{\alpha-1}$$

$$\frac{d\lambda}{dt} = -k_1 \dot{\gamma} \lambda + k_2 (1 - \lambda)$$

$$\lambda(t) = \left\{ \frac{k_2}{\dot{\gamma}_0 k_1 + k_2} - \frac{k_2}{\dot{\gamma} k_1 + k_2} \right\} \exp[-(\dot{\gamma} k_1 + k_2)t] + \frac{k_2}{\dot{\gamma} k_1 + k_2}$$



$$\eta(t) = \eta_{\infty} + \left(\left\{ \frac{1}{\dot{\gamma}_0 K + 1} - \frac{1}{\dot{\gamma} K + 1} \right\} \exp[-k_2(\dot{\gamma} K + 1)t] + \frac{1}{\dot{\gamma} K + 1} \right) K_0 \dot{\gamma}^{\alpha-1}$$

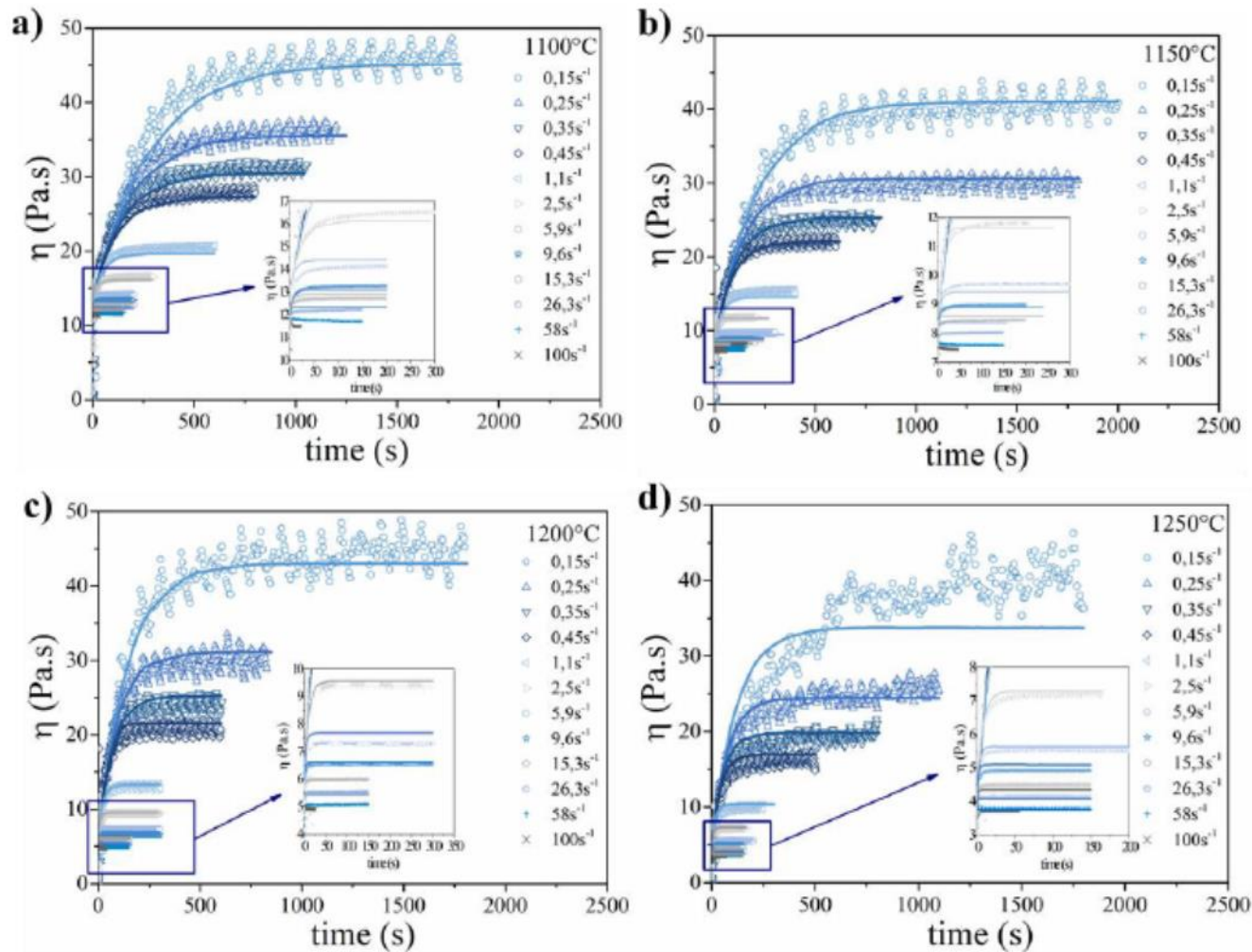
$$K = k_1/k_2 \quad k_2 = 1/t_c = \dot{\gamma}_c = 10^{-3} \text{ s}^{-1}$$

Parameters	1100 °C	1150 °C	1200 °C	1250 °C
η_{∞}	13	8.0	6.0	4.0
K	15	24	35	57
K_0	115	177	285	400
α	1.05	1.08	1.11	1.18

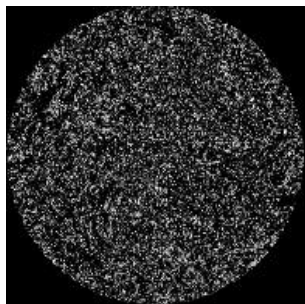
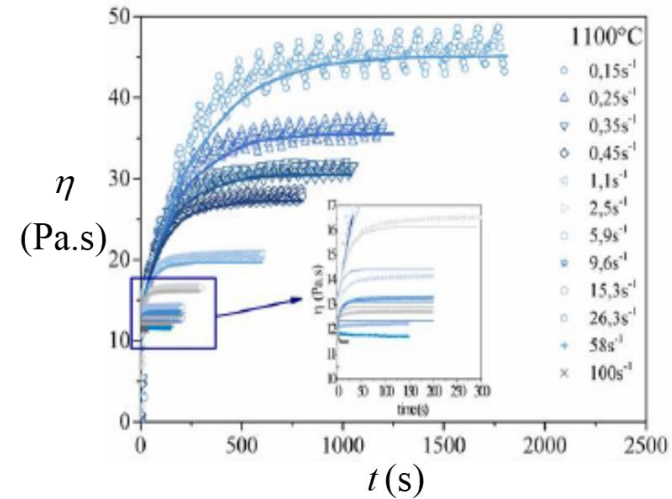
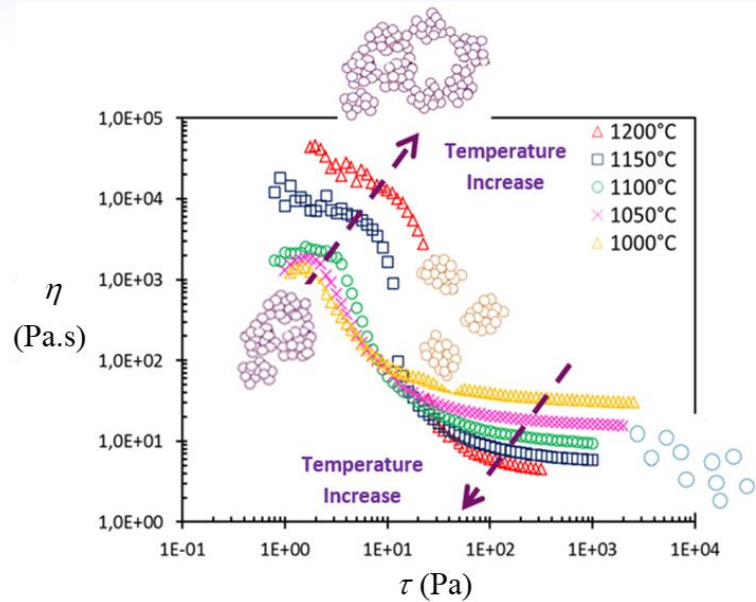
$$t \rightarrow +\infty \quad \eta(T, \phi, \dot{\gamma}, t) \rightarrow$$

$$\eta = \eta_{\infty} + \left(\frac{1}{\dot{\gamma} K + 1} \right) K_0 \dot{\gamma}^{\alpha-1}$$

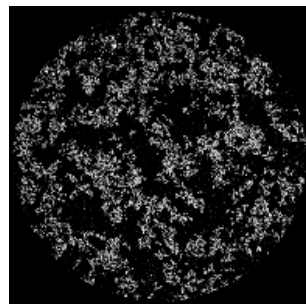
Modélisation du régime transitoire



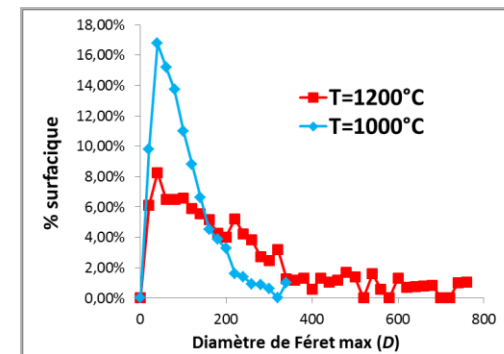
Agrégation - Facteurs influents



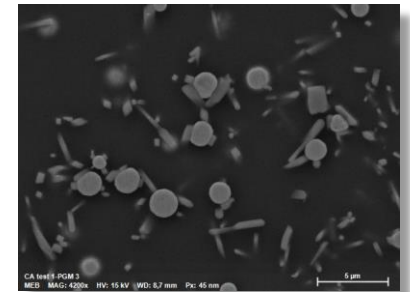
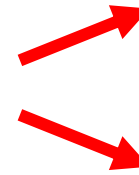
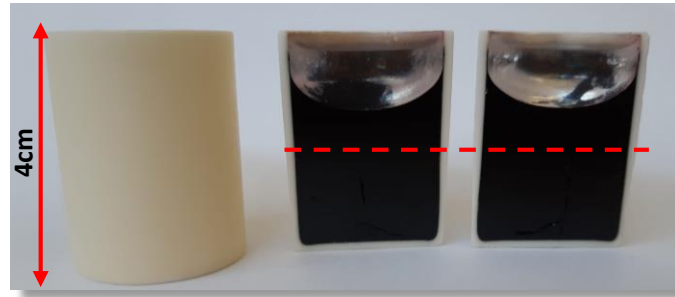
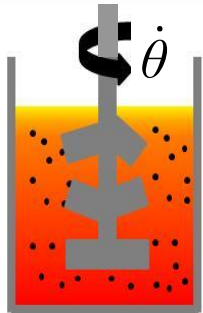
1000°C



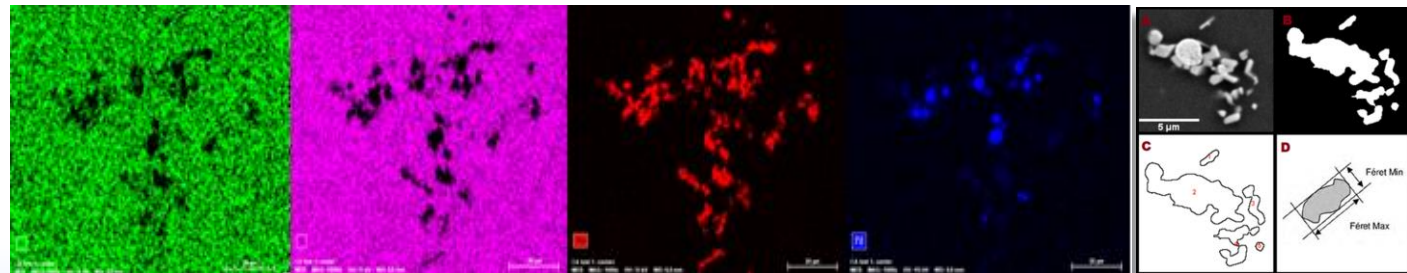
1200°C



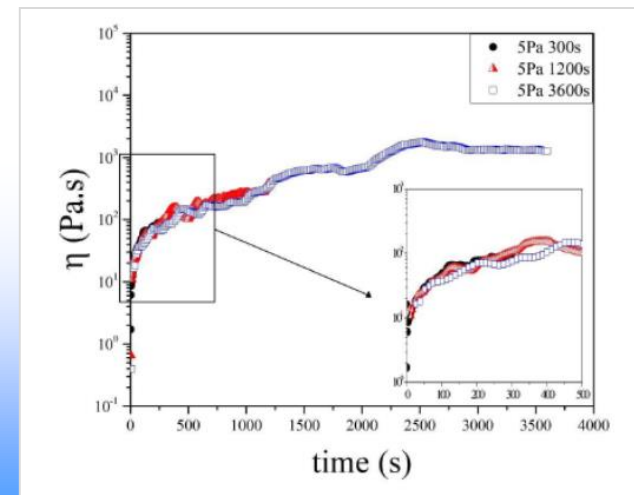
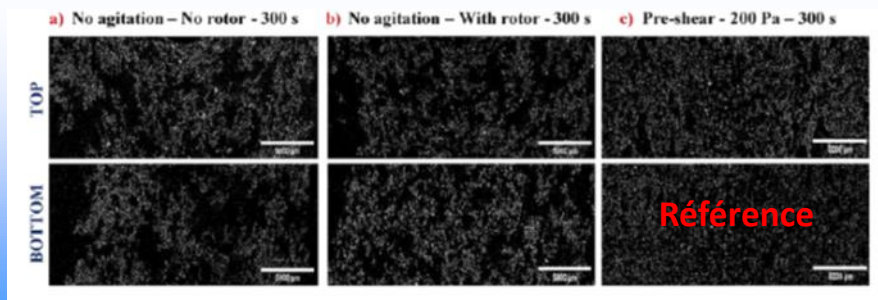
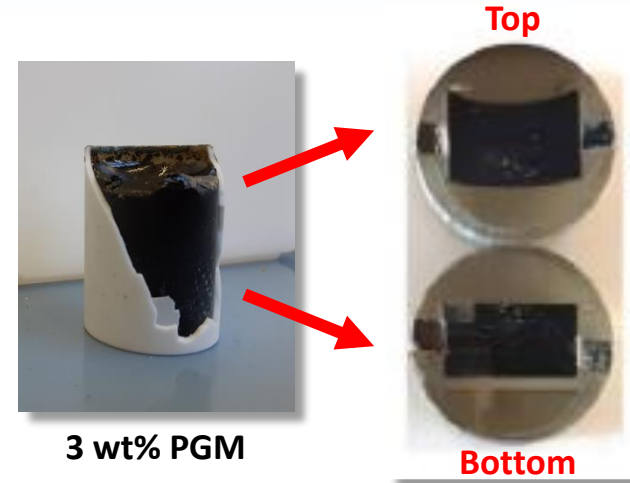
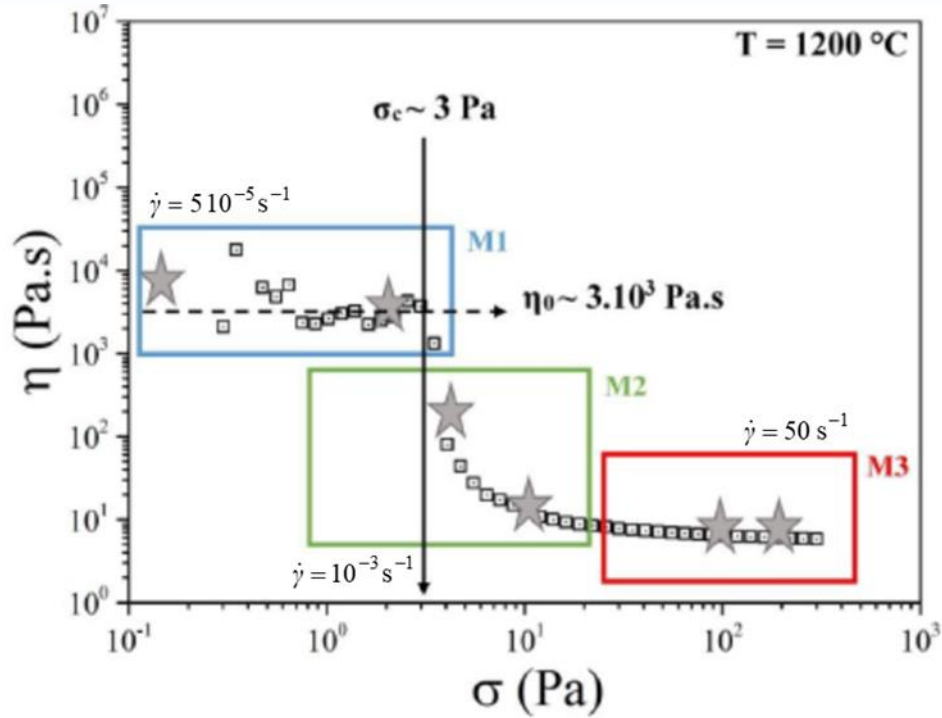
Agrégation - Protocole d'analyse



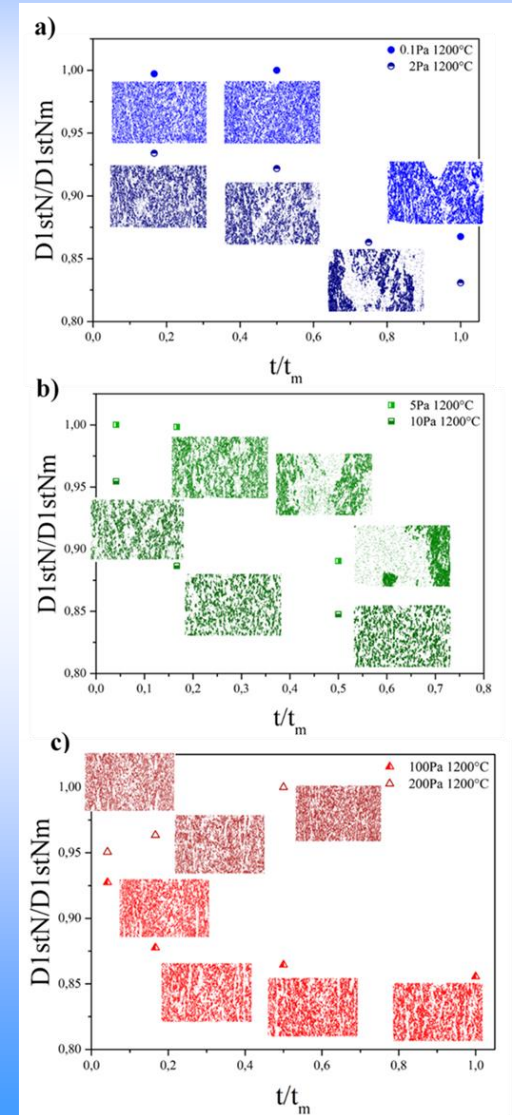
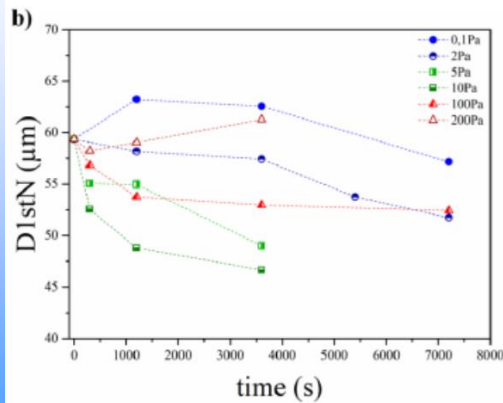
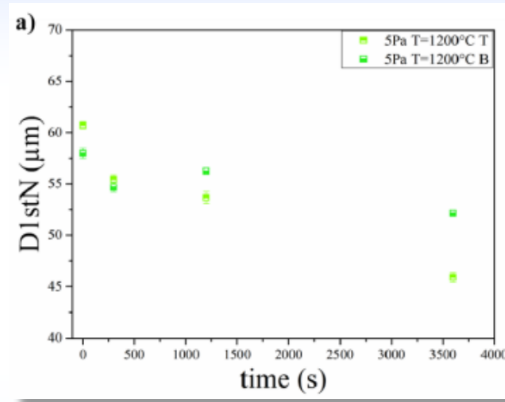
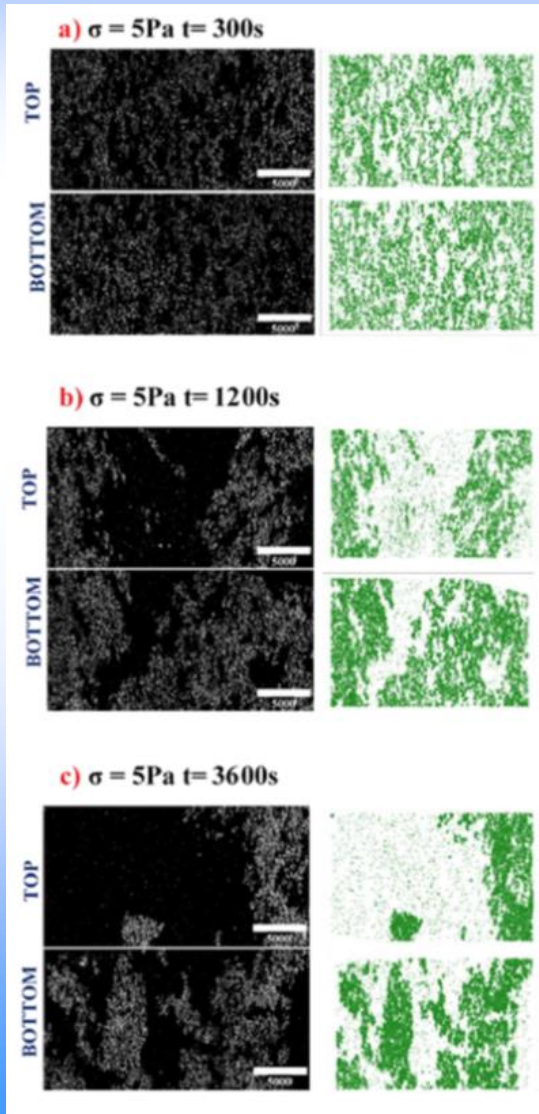
Analyse
d'image



Agrégation - Effet du cisaillement



Agrégation - Résultats expérimentaux

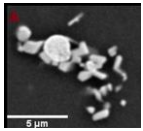


Agrégation - Modélisation

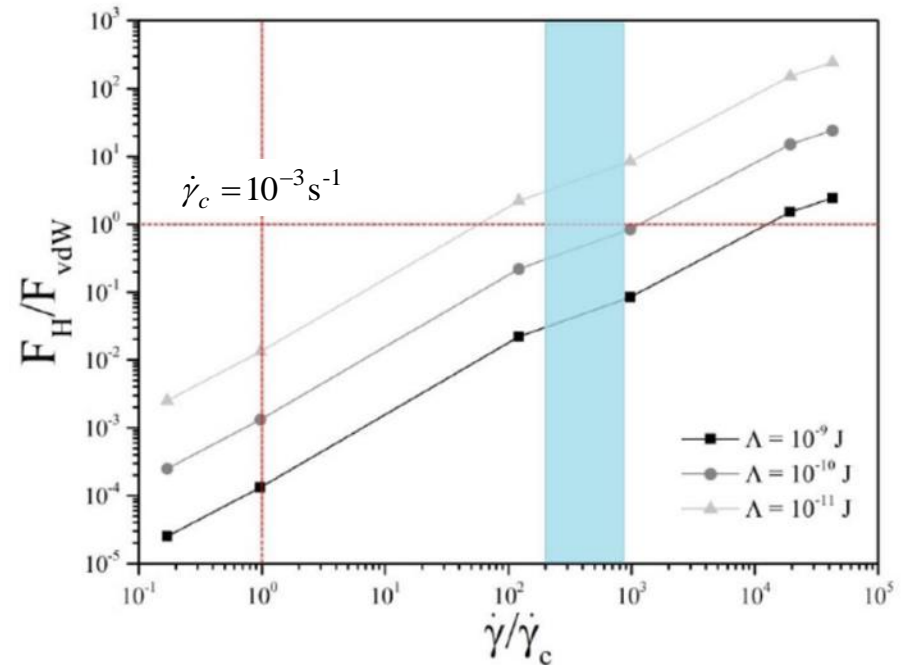
Modèle de Allain Taille maximale des amas

$$R \cong a \left(\frac{\Lambda}{a 12 h^2 \eta_f \dot{\gamma}} \right)^{1/2}$$

Λ ← Constante d'Hamaker
 $\dot{\gamma}$ ← Taux de cisaillement
 a ← Taille des particules primaires ($\approx 5 \mu\text{m}$)
 h ← Distance entre particules
 η_f ← Viscosité phase continue



$$F_H = \eta_f \dot{\gamma} R^2 \qquad F_{vdW} = \frac{\Lambda a}{12 h^2}$$



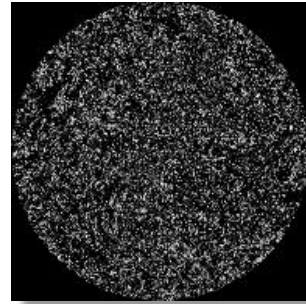
$1000^\circ\text{C} \rightarrow R \approx 180 \mu\text{m} \Rightarrow D \approx 360 \mu\text{m}$
 $1200^\circ\text{C} \rightarrow R \approx 500 \mu\text{m} \Rightarrow D \approx 1000 \mu\text{m}$



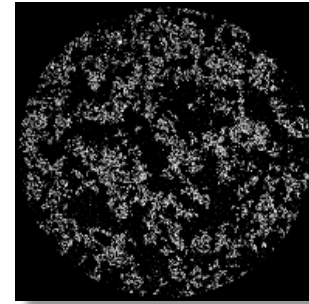
$1000^\circ\text{C} \rightarrow D \approx 20 \text{ à } 350 \mu\text{m}$
 $1200^\circ\text{C} \rightarrow D \approx 20 \text{ à } 750 \mu\text{m}$

Sédimentation

1000°C

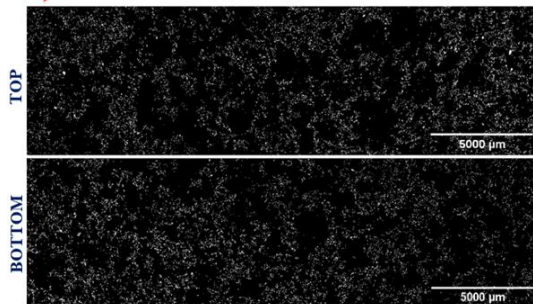


1200°C

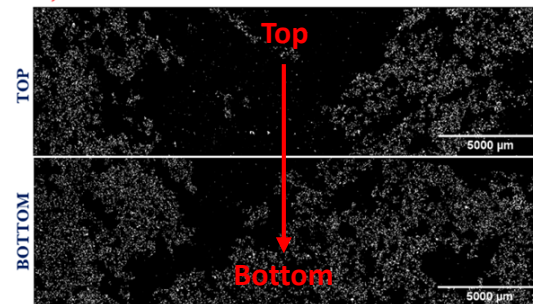


Diffusion \Rightarrow *Agrégation* \Rightarrow *Sédimentation*

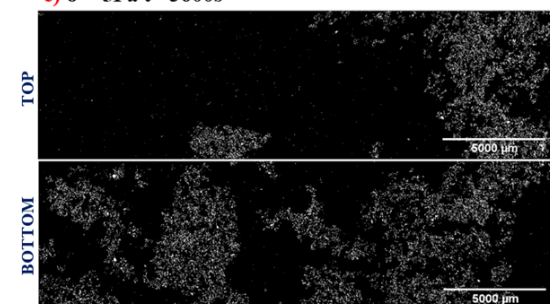
a) $\sigma = 5\text{Pa}$ $t = 300\text{s}$



b) $\sigma = 5\text{Pa}$ $t = 1200\text{s}$

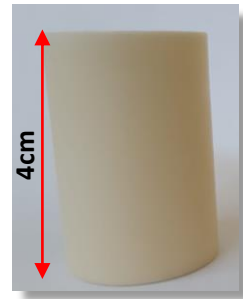
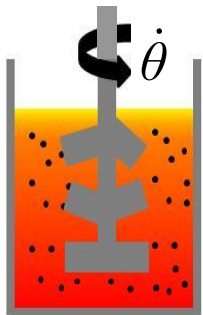


c) $\sigma = 5\text{Pa}$ $t = 3600\text{s}$

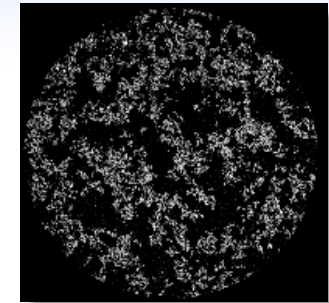


temps

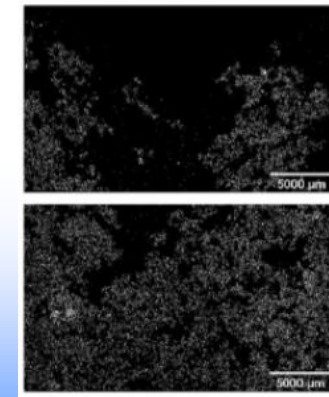
Sédimentation - Protocole d'analyse



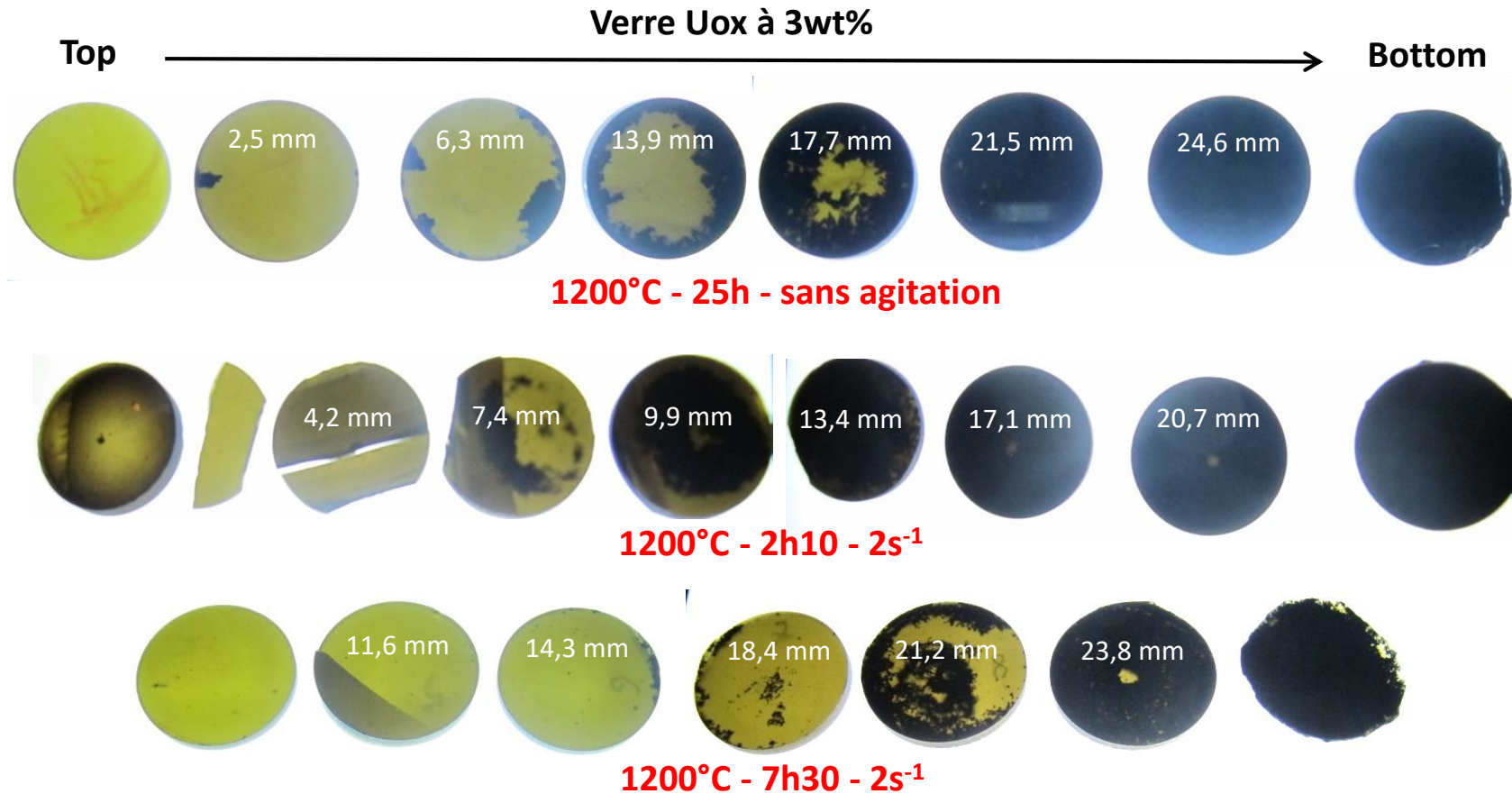
Carottage et
tronçonnage



Prises et analyses
d'images horizontales
et verticales

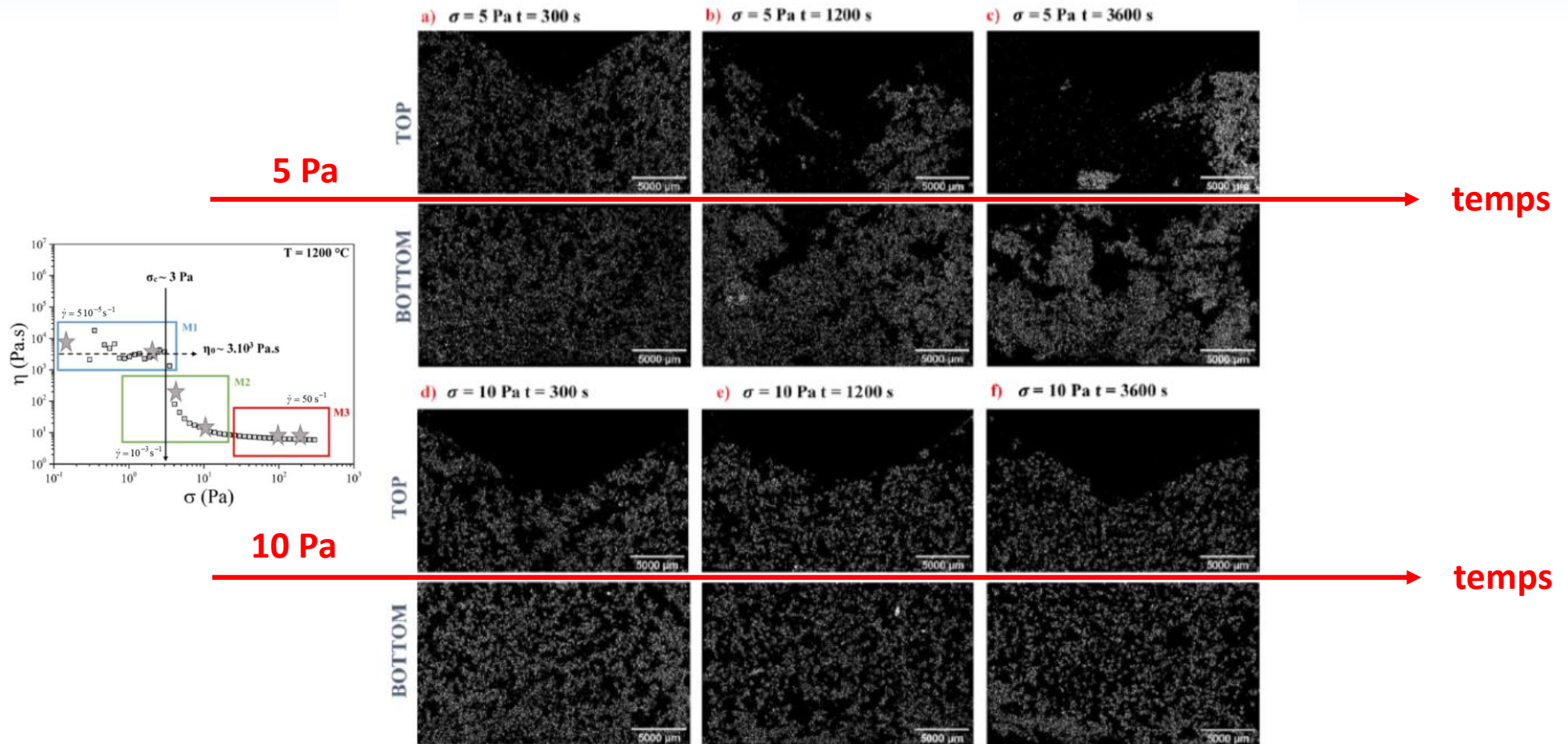


Sédimentation - Résultats expérimentaux

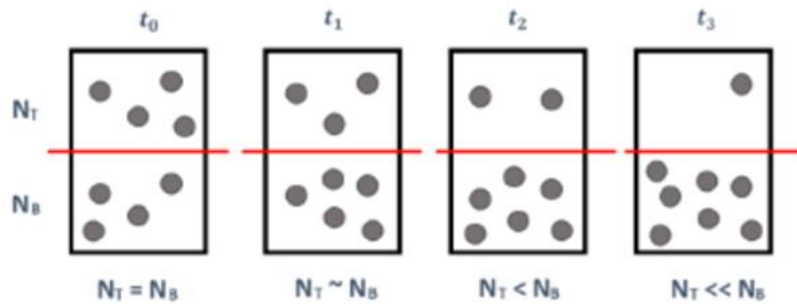


→ Le cisaillement accélère la sédimentation

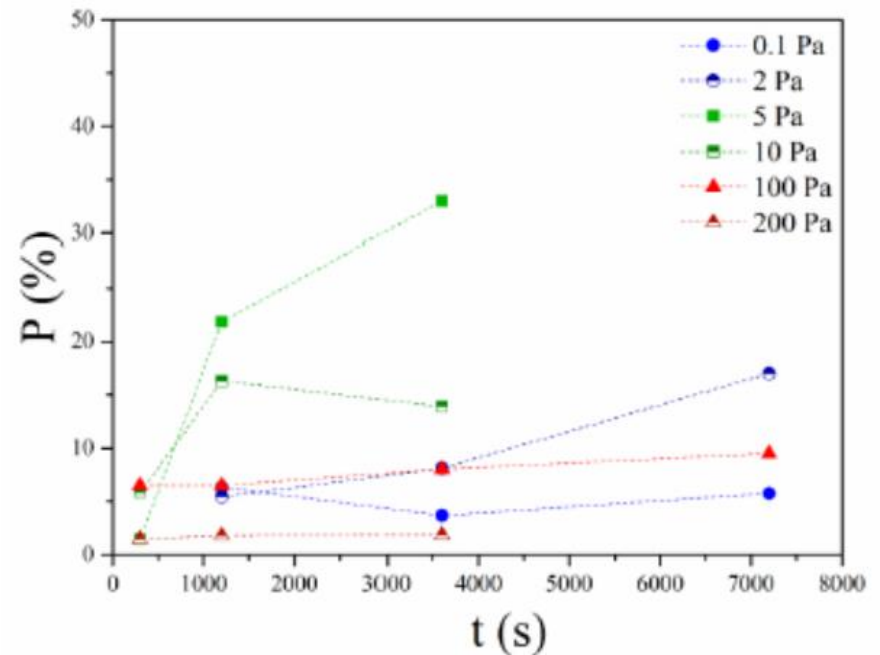
Sédimentation



Sédimentation - Analyse des résultats



$$P = 50 - \left(\left(\frac{N_T}{N_T + N_B} \right) \cdot 100 \right)$$



Modèle de Allain

Vitesse de sédimentation avec
prise en compte de l'agglomération

$$U = \frac{2}{9} \frac{(\rho_p - \rho_f) g a^2}{\eta_f} \left(\frac{R}{a} \right)^{D_f - 1}$$

Rayon des amas \rightarrow R

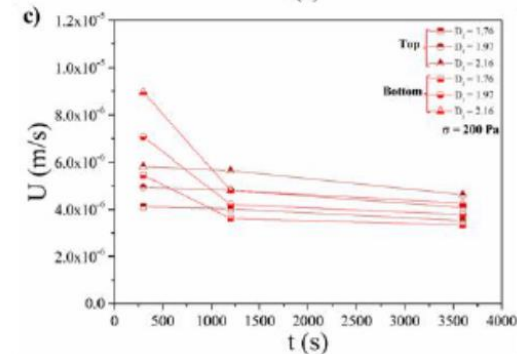
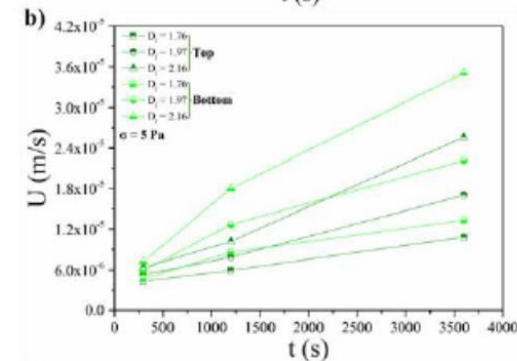
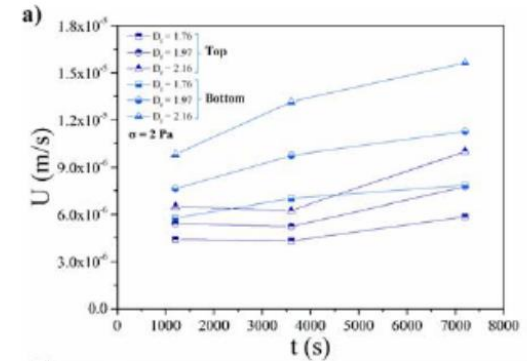
Taille des particules primaires \rightarrow a

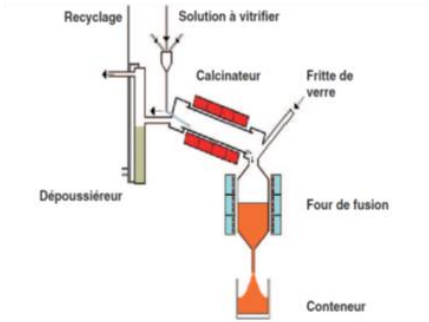
Dimension fractale des amas \rightarrow D_f

$$\rho_p = 8212 \text{ kg.m}^{-3} \quad \rho_f = 2736 \text{ kg.m}^{-3} \quad a = 25 \text{ }\mu\text{m}$$

$$R = R_{\text{max}} = 500 \text{ }\mu\text{m} \quad \eta_f = 3.6 \text{ Pa.s} \quad D_f = 1.7 - 2.2$$

→ $U \sim 20 \text{ }\mu\text{m.s}^{-1}$ ($U_{\text{ox}}/1200^\circ\text{C}$)

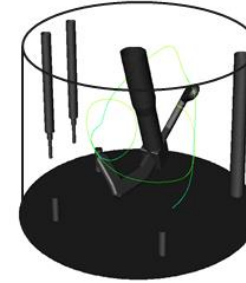




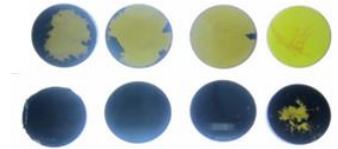
Équations de transport (conservation de la masse et de la quantité de mouvement), CFD, sédimentation, ...

$$\frac{\partial \rho}{\partial t} = -\vec{\nabla} \cdot (\rho \vec{v}) \quad \frac{\partial \rho \vec{v}}{\partial t} = -\vec{\nabla} \cdot (\rho \vec{v} \vec{v}) - \vec{\nabla} p - \vec{\nabla} \cdot \underline{\underline{\tau}} + \rho \vec{f}$$

$$\underline{\underline{\dot{\gamma}}} = (\vec{\nabla} \vec{v})^t + \vec{\nabla} \vec{v} \quad \underline{\underline{\tau}} = -\eta \underline{\underline{\dot{\gamma}}} \quad \eta = \eta_{\infty} + \frac{\eta_o - \eta_{\infty}}{1 + \dot{\gamma} / \dot{\gamma}_c}$$



$$U_{\phi} = \frac{2(\rho_p - \rho_f)ga^2}{9\eta_f} \left(\frac{R}{a}\right)^{D_f - 1}$$



macro (milieu continu)
Mécanique des fluides



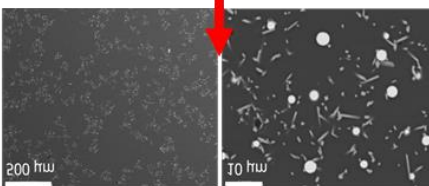
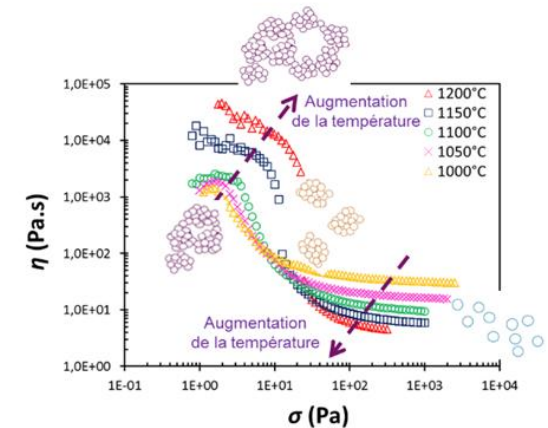
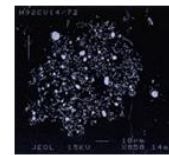
méso (milieu moyenné)
Rhéologie

Coefficients de transport (viscosité, module élastique,...) vs paramètres structuraux (forme des particules, taille des agrégats,, dimension fractale, ...)

$$\eta_f = A \exp\left(\frac{B}{T - T_o}\right) \quad R \cong a \left(\frac{A}{a 12 h^2 \eta_f \dot{\gamma}}\right)^{1/2}$$

$$\eta_o(T, \varphi_o) = \eta_f(T) \exp\left[\frac{\varphi_o}{\varphi_c} \left(\frac{R}{a}\right)^{3-D_f}\right]$$

$$\eta_{\infty} = \eta_f \left(1 - \frac{\alpha \varphi_o}{\varphi_m}\right)^{-2} = \eta_f \left(1 - \frac{\varphi_o}{\varphi_{meff}}\right)^{-2}$$



micro (milieu discret)
Physicochimie

Paramètres caractéristiques $\eta_f, \eta_o, \eta_{\infty}, a, A...$ vs composition chimique et structure moléculaire. Interactions de van der Waals, constante d'Hamaker, processus de cristallisation, constante de solubilité, ...

	%Ma. Min.	%Ma. Max.
SiO ₂	42.4	51.7
Al ₂ O ₃	3.6	6.6
B ₂ O ₃	12.4	16.5
Na ₂ O	8.1	11.0
FP* + Act* + Ptds+ ZrO ₂	4.2	18.5
RuO ₂ + Pd + Rh	0	3.0

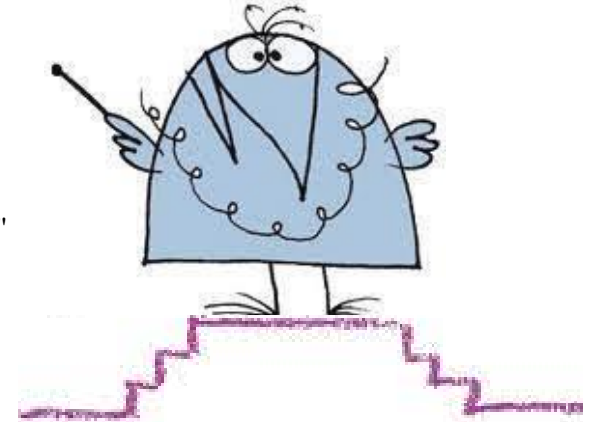
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Questions ?

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C'EST
TOUT
POUR
AUJOURD'
HUI.

