

**Memory of plastic fluid  
and its application to control  
pattern formation  
of desiccation cracks**

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**Tottori Univ.**

**Shimane Univ.**

**Nara Women's Univ.**

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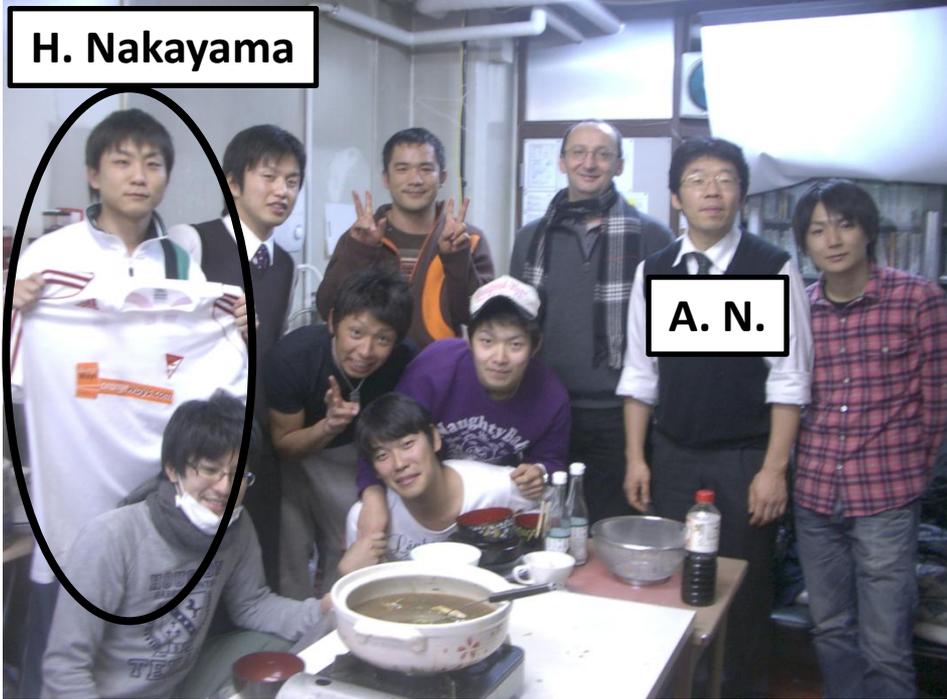
**Ooshida Takeshi**

**M. Otsuki**

**S. Kitsunozaki**



Y. Matsuo



H. Nakayama

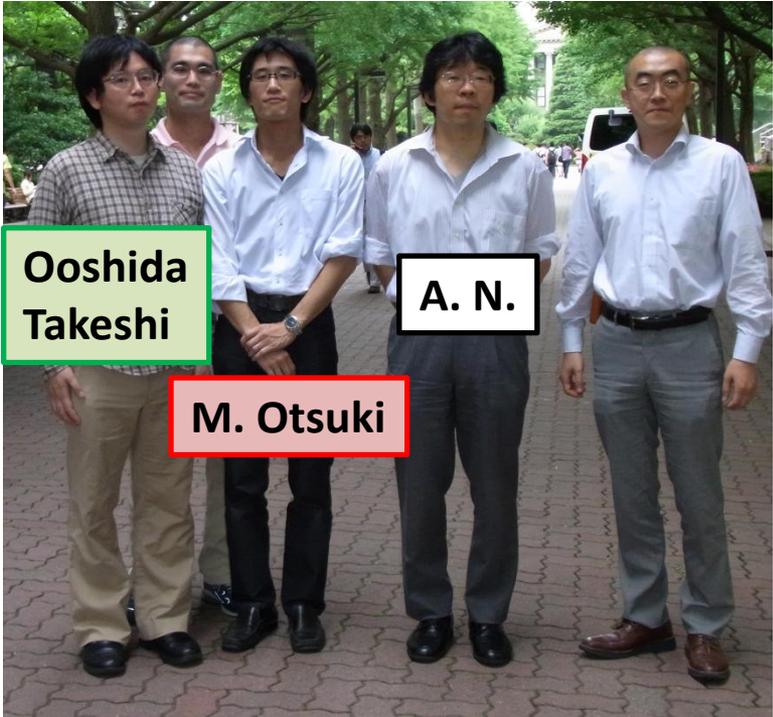
A. N.

Nihon Univ.

Tottori Univ.

Shimane Univ.

Nara Women's Univ.



Ooshida  
Takeshi

A. N.

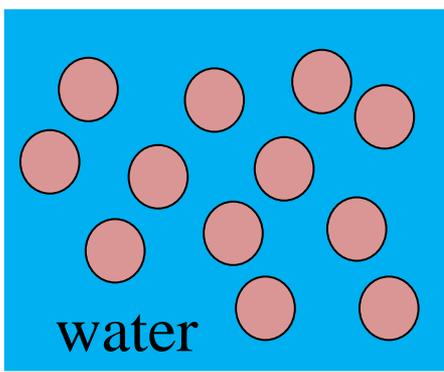
M. Otsuki



S. Kitsunezaki

# Desiccation cracks of colloidal suspension

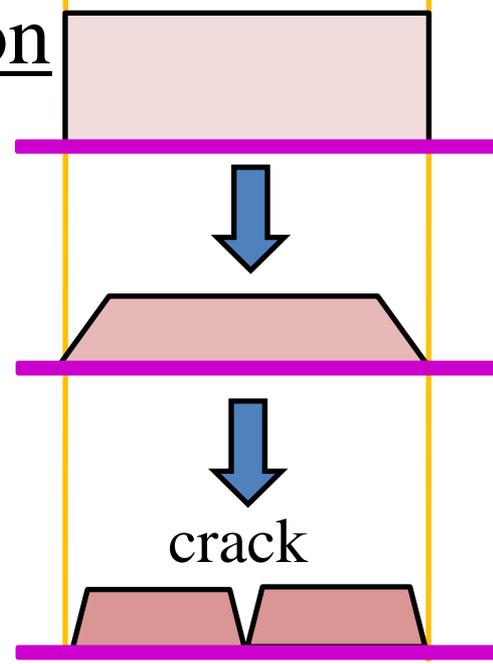
Colloidal particles  
( $\sim \mu\text{m}$ )



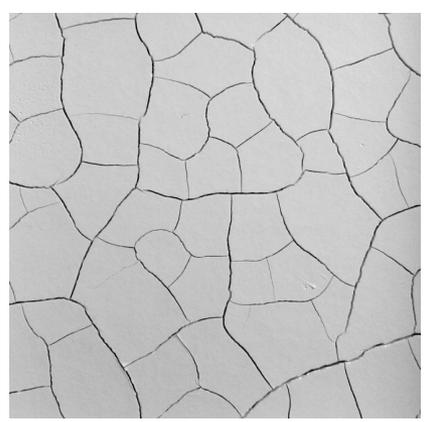
Contraction due to drying  
+  
Adhesion at the bottom

To release stress horizontally

Formation of cracks  
spacing  $\sim$  depth of paste

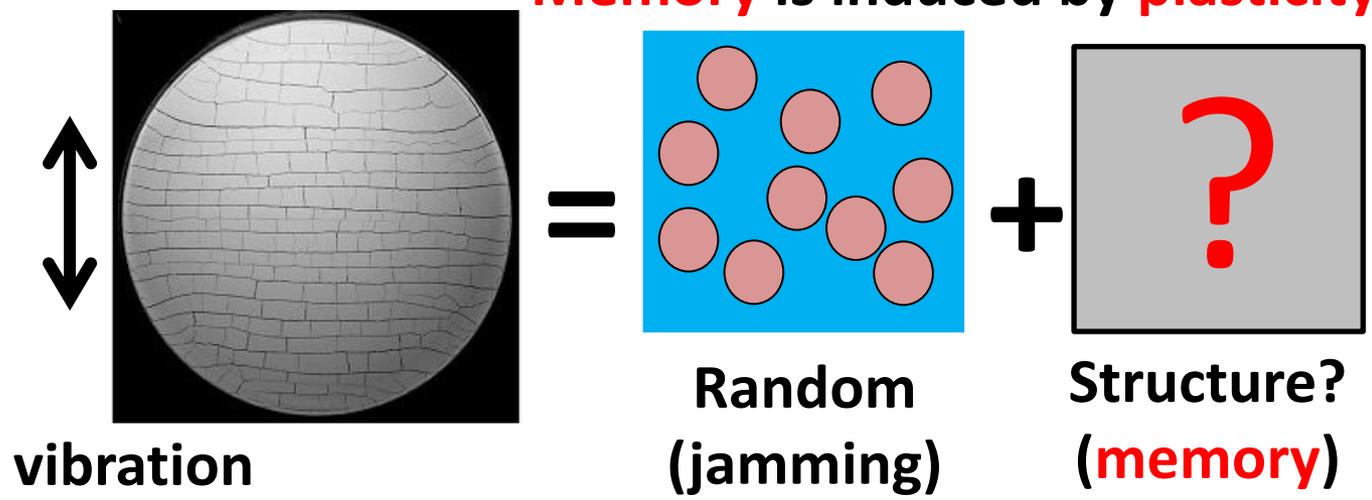


Usually cellular isotropic cracks

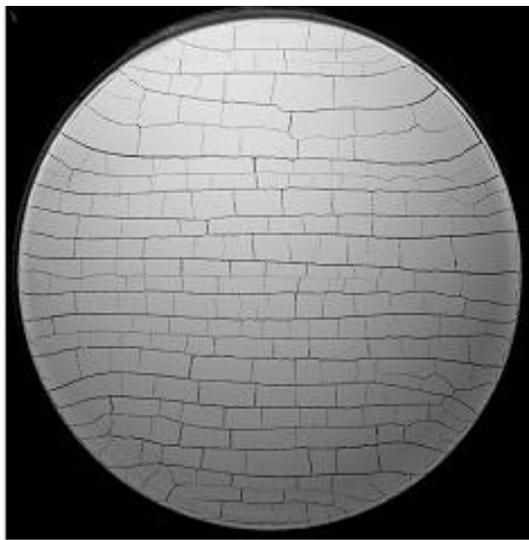


Crack patterns produced by memory effect

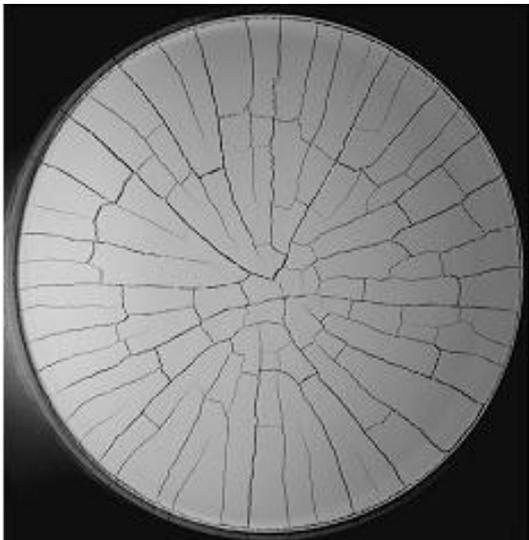
Memory is induced by plasticity



# Desiccation crack patterns produced by memory effect



Lamellar

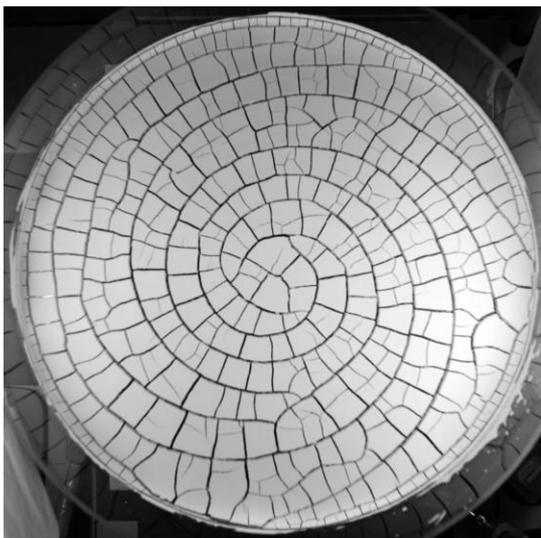


Radial

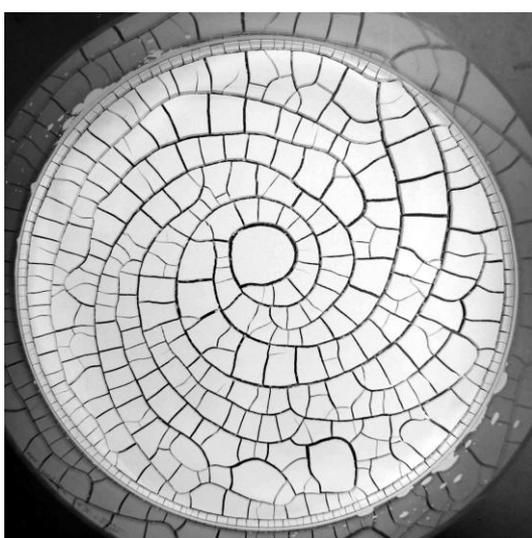


lattice

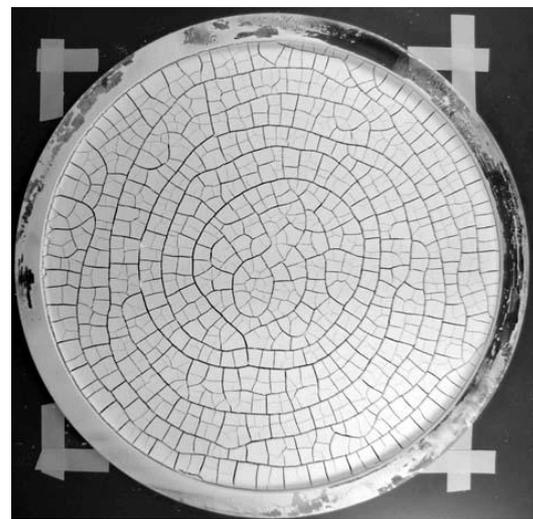
← 500mm →



Spiral



Spiral

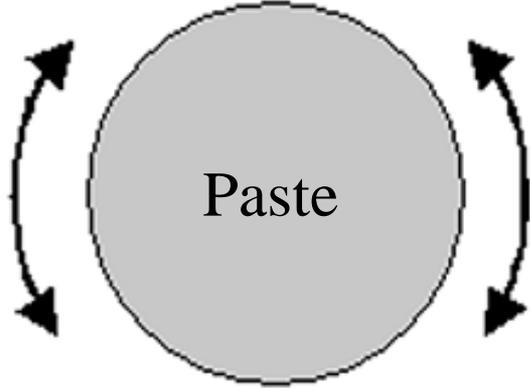


Ring

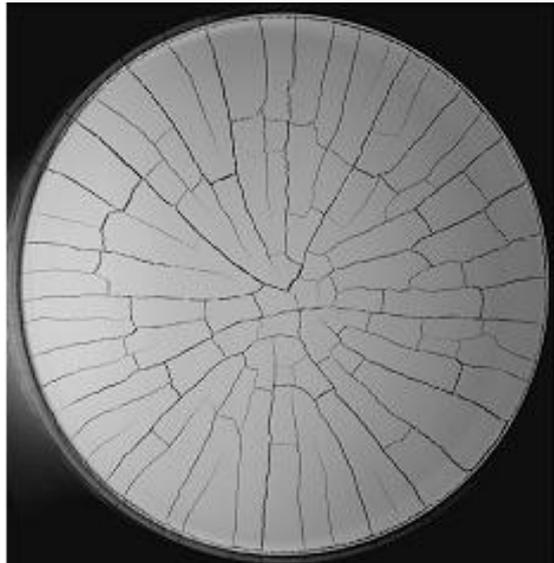
# Memory of vibration that water-poor $\text{CaCO}_3$ paste has

Water-poor  $\text{CaCO}_3$  paste

In angular direction

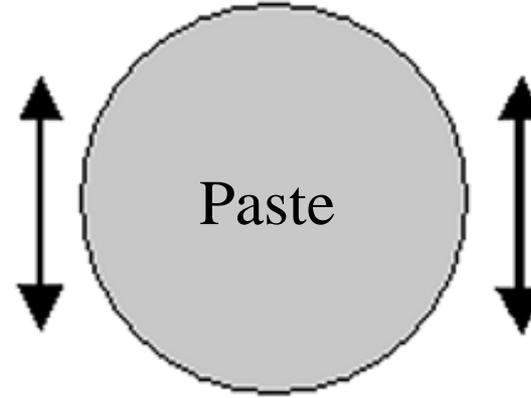


Stop vibration ↓ & dry

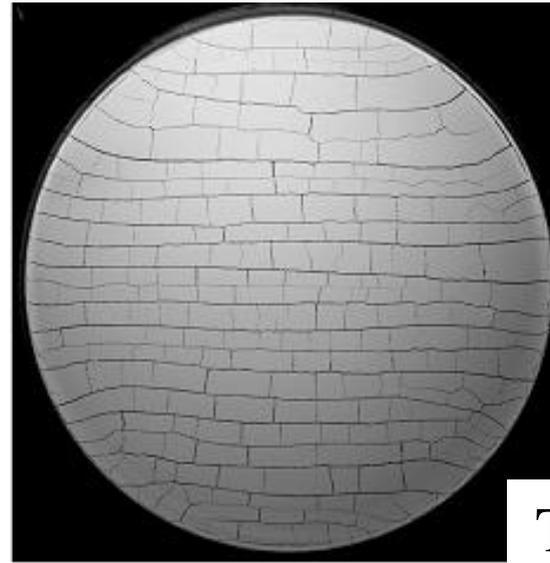


radial

In one direction



Stop vibration ↓ & dry



lamellar

Memory of vibration

Perpendicular to vibration

Not by boundary effect

Thickness 10mm  
Diameter 500mm

movie

movie

# Memory of vibration

A. Nakahara and Y. Matsuo  
J. Phys. Soc. Jpn. 74 (2005) 1362.

**Water-poor** CaCO<sub>3</sub> paste

⊕ charged

**Water-rich** CaCO<sub>3</sub> paste

Paste is  
**vibrated**



Initial  
vibration  
(horizontally)

Paste is  
**vibrated**  
and **fluidized**

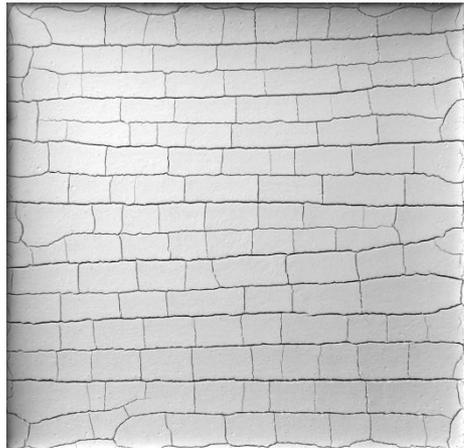


Stop vibration & dry

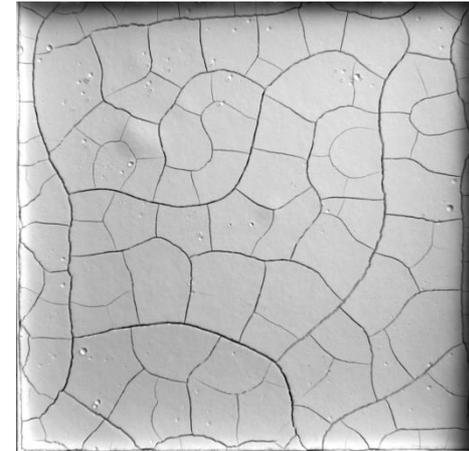


Lamellar cracks  
(**perpendicular to vibration**)

Cellular cracks  
(isotropic and homogeneous)



Crack  
formation  
(top view)



**Memory of vibration**

**No memory**

# Uncharged Pastes remember vibration and flow !

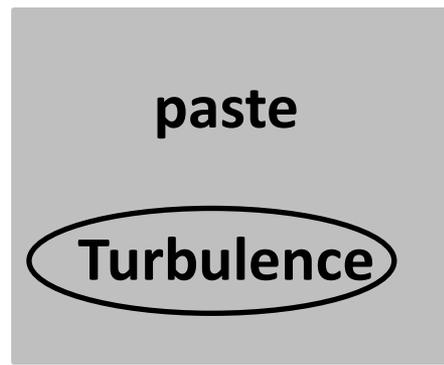
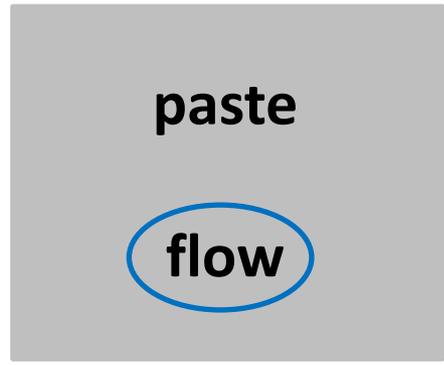
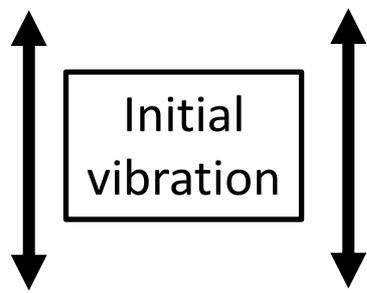
Magnesium Carbonate Hydroxide, Carbon, Kaolin, etc.

A. Nakahara and Y. Matsuo  
PRE. 74 (2006) 045102.

**Uncharged**  
**Water-rich**

**Water-poor**

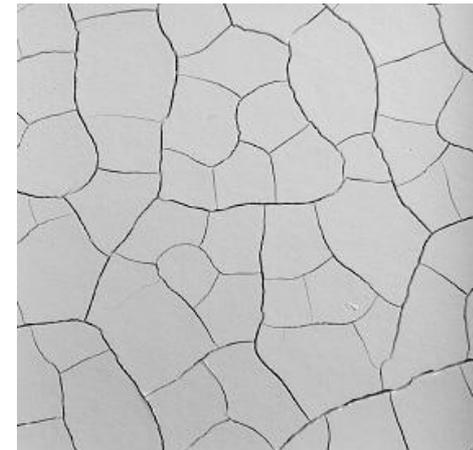
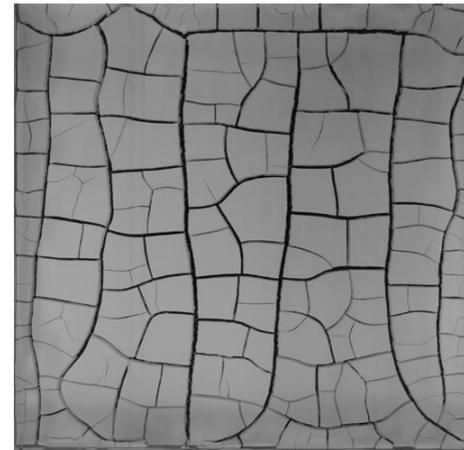
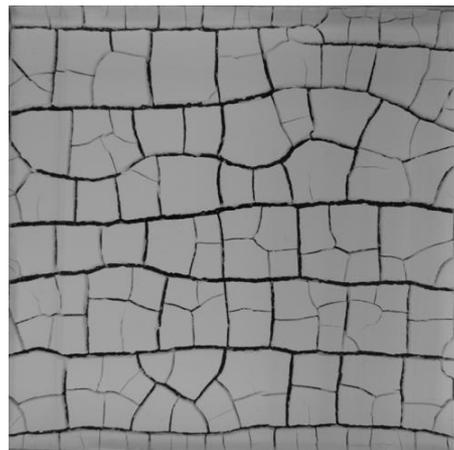
**Water-too-much**



Stop & dry



Crack formation



Memory of vibration

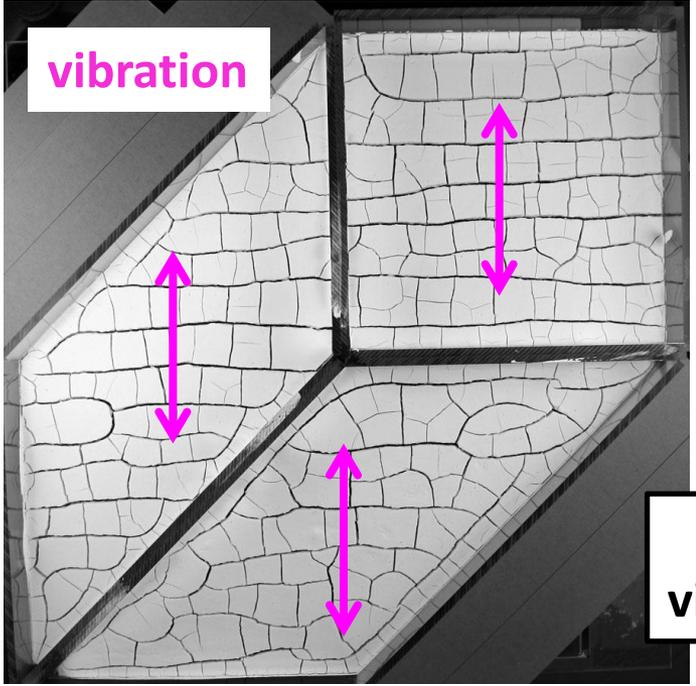
Memory of flow

No memory

# Difference between two memories

## Water-poor paste

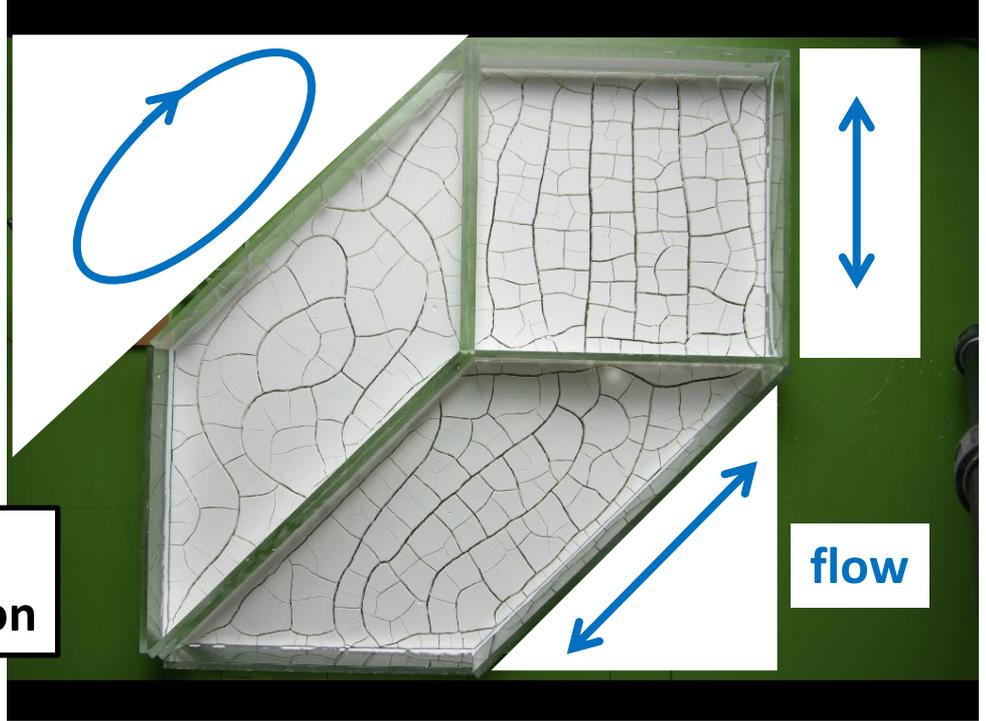
Lamellar cracks  
(perpendicular to vibration)  
Memory of vibration



all perpendicular to vibration

## Water-rich & uncharged paste

Lamellar cracks  
(parallel to flow)  
Memory of flow



parallel to local flow direction

movie

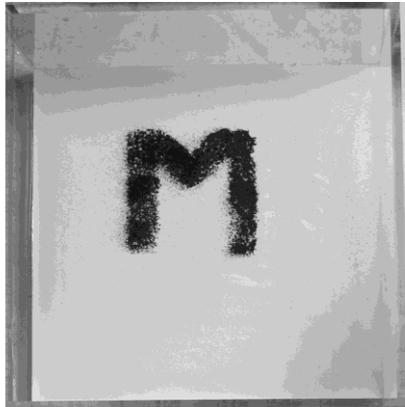
initial  
vibration

# Quantitative difference between **vibration** and **flow**

Water-poor paste



(a) before vibration

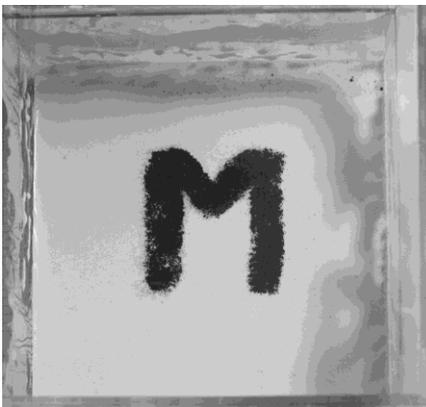


(b) under vibration

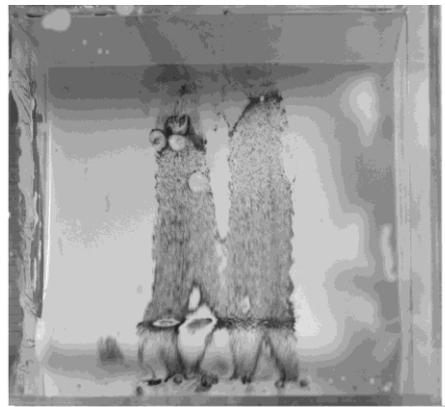
before vibration

under vibration

Water-rich paste



(a) before flow



(b) under flow

before flow

under flow

Check deformation of letter **M** (written by carbon powder)

Faint plastic deformation under vibration

Large scale deformation (elongation) under flow and shear

We need to control **Strain** and **Shear rate** in future experiments

# Only flow experiment (without vibration)

Magnesium Carbonate Hydroxide,  $\rho=7.7\%$

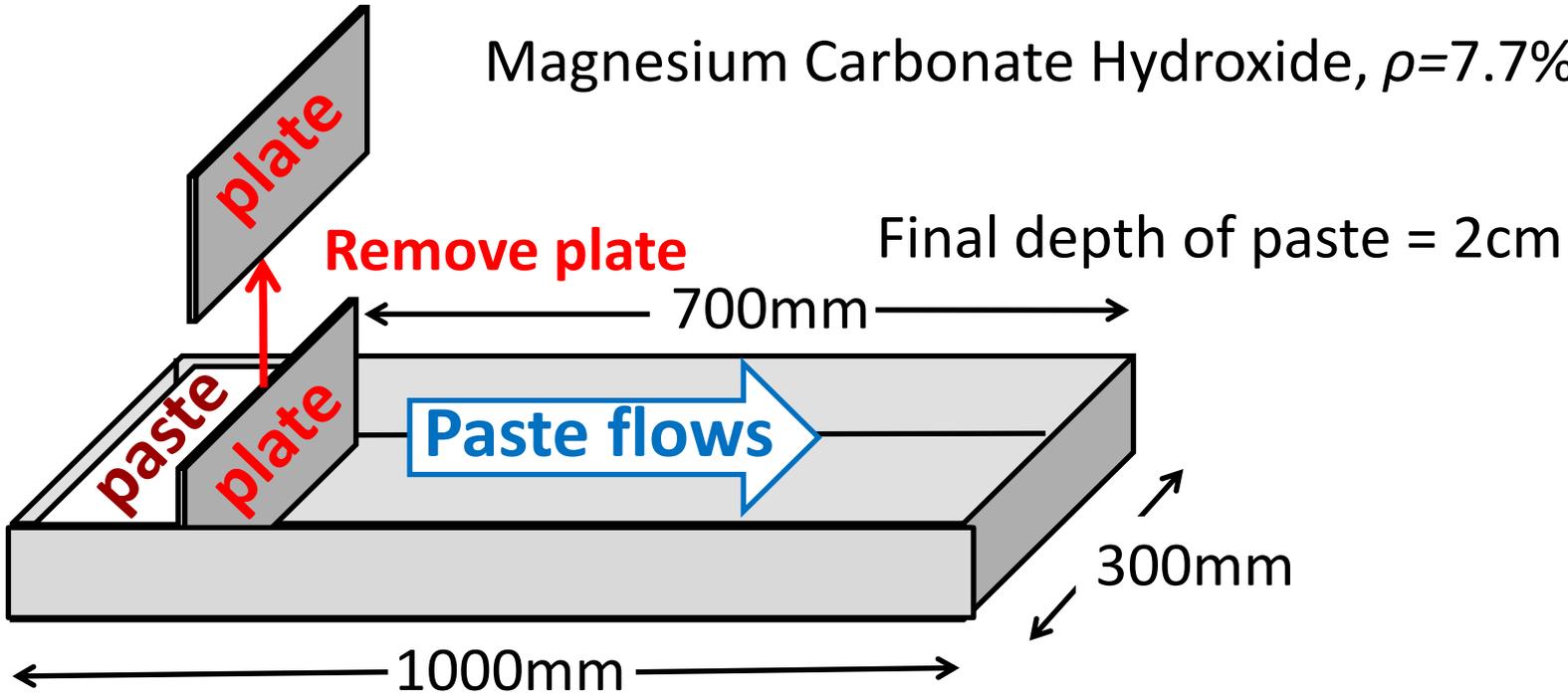
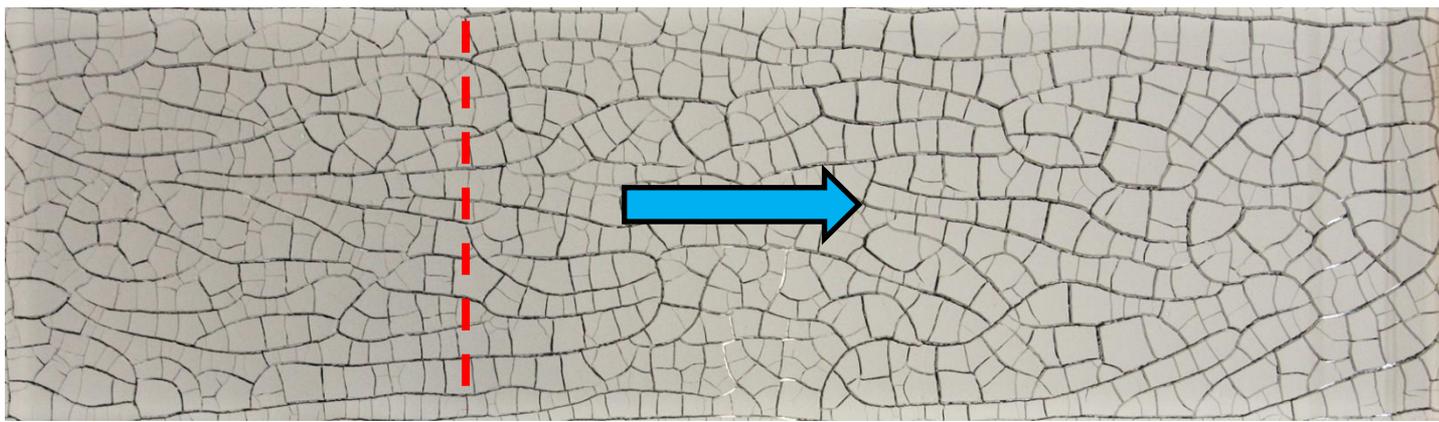


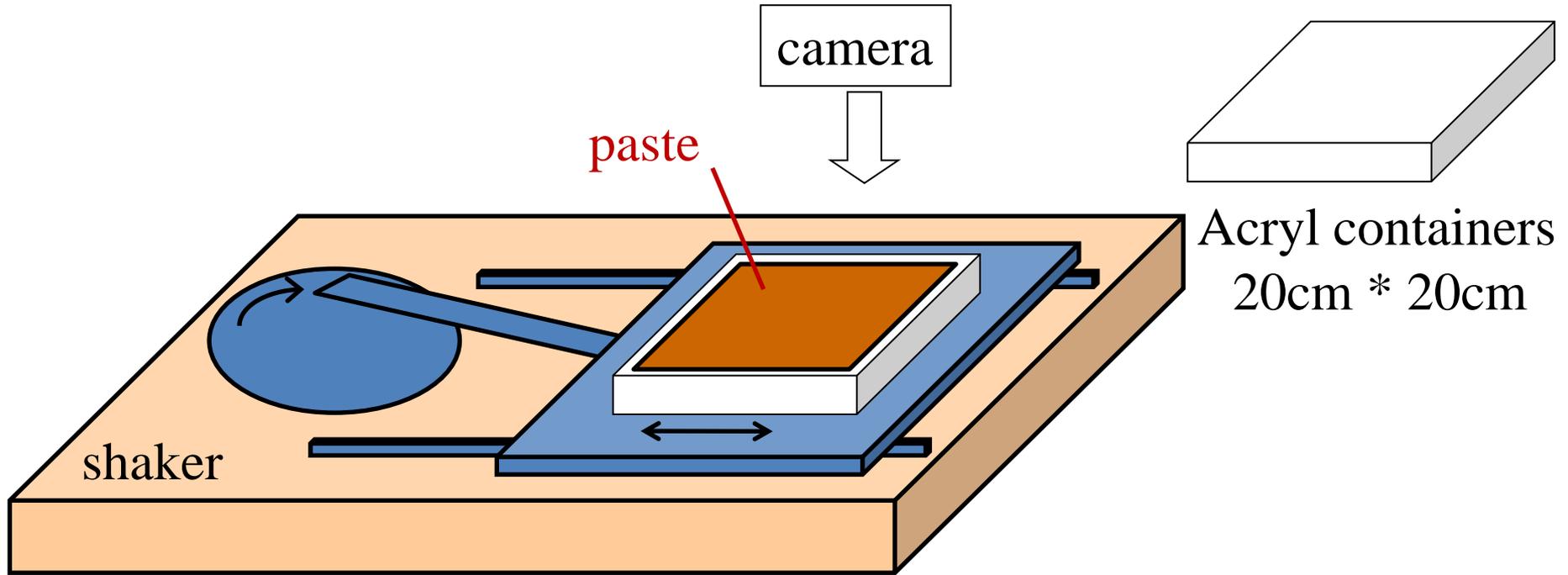
Plate was here.



Just a Flow

Memory of flow (cracks parallel to flow)

# Experiment to make morphological phase diagrams



Initial external vibration

for 60 sec, amplitude 15 mm, frequency 30~150 rpm

Controlling parameter in experiments

- solid volume fraction  $\rho$
  - strength of the initial vibration  
amplitude  $r$ , frequency  $f$
- Change systematically

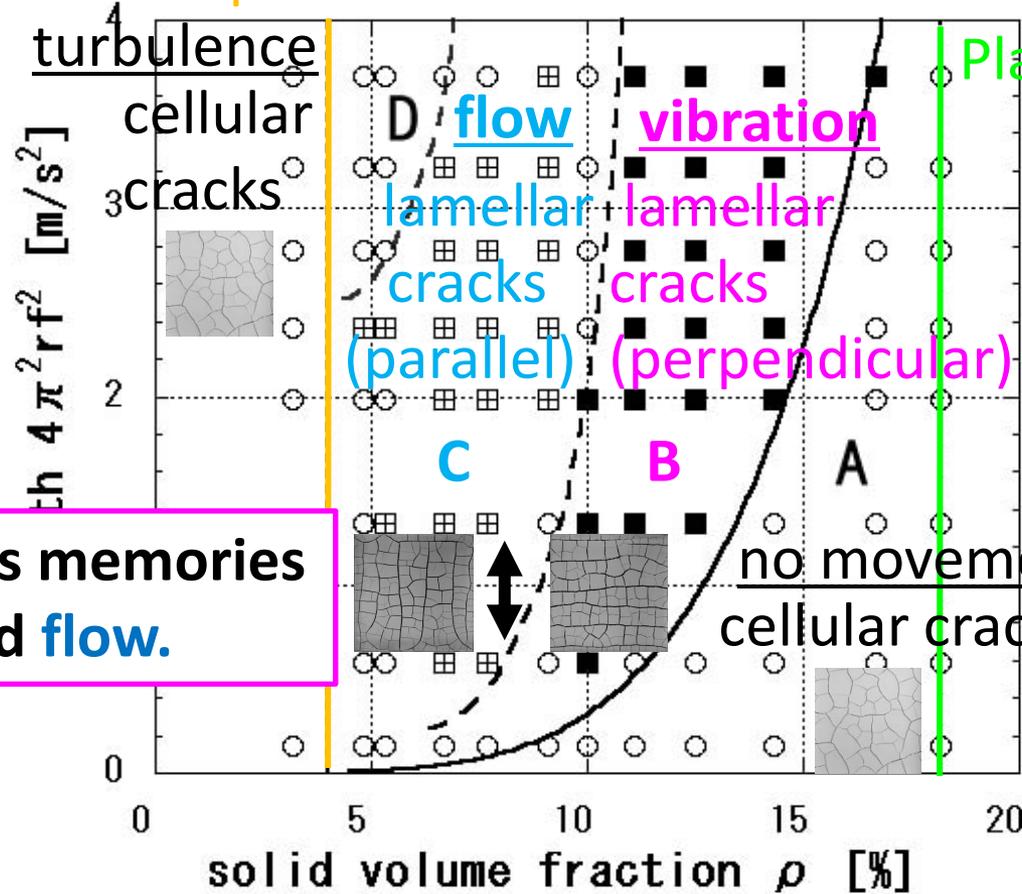
# Morphological Phase Diagram of **uncharged** paste

magnesium  
carbonate  
hydroxide

Liquid limit

Yield stress

Plastic limit

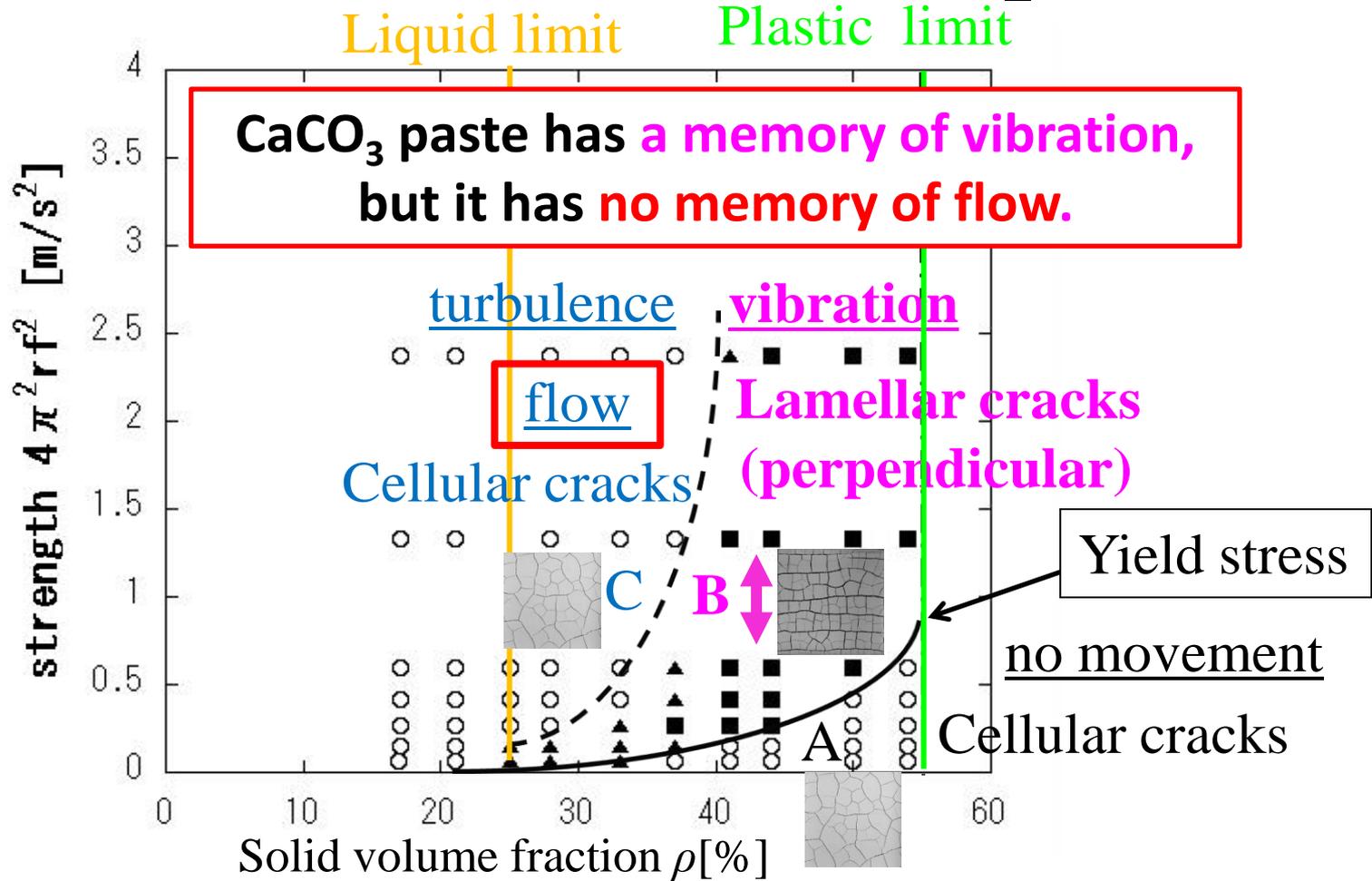


**Uncharged paste** has memories  
of **vibration** and **flow**.

- |   |                    |                 |                                   |
|---|--------------------|-----------------|-----------------------------------|
| A | <u>no movement</u> | → no experience | → cellular cracks (no movement)   |
| B | <u>vibration</u>   | → remember      | → Lamellar cracks (perpendicular) |
| C | <u>flow</u>        | → remember      | → Lamellar cracks (parallel)      |
| D | <u>turbulence</u>  | → forget        | → Cellular cracks (no memory)     |

# Morphological Phase Diagram of $\text{CaCO}_3$ paste

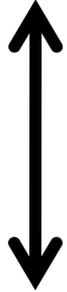
Movies of  
[no movement](#)  
[vibration](#)  
[flow](#)  
[turbulence](#)



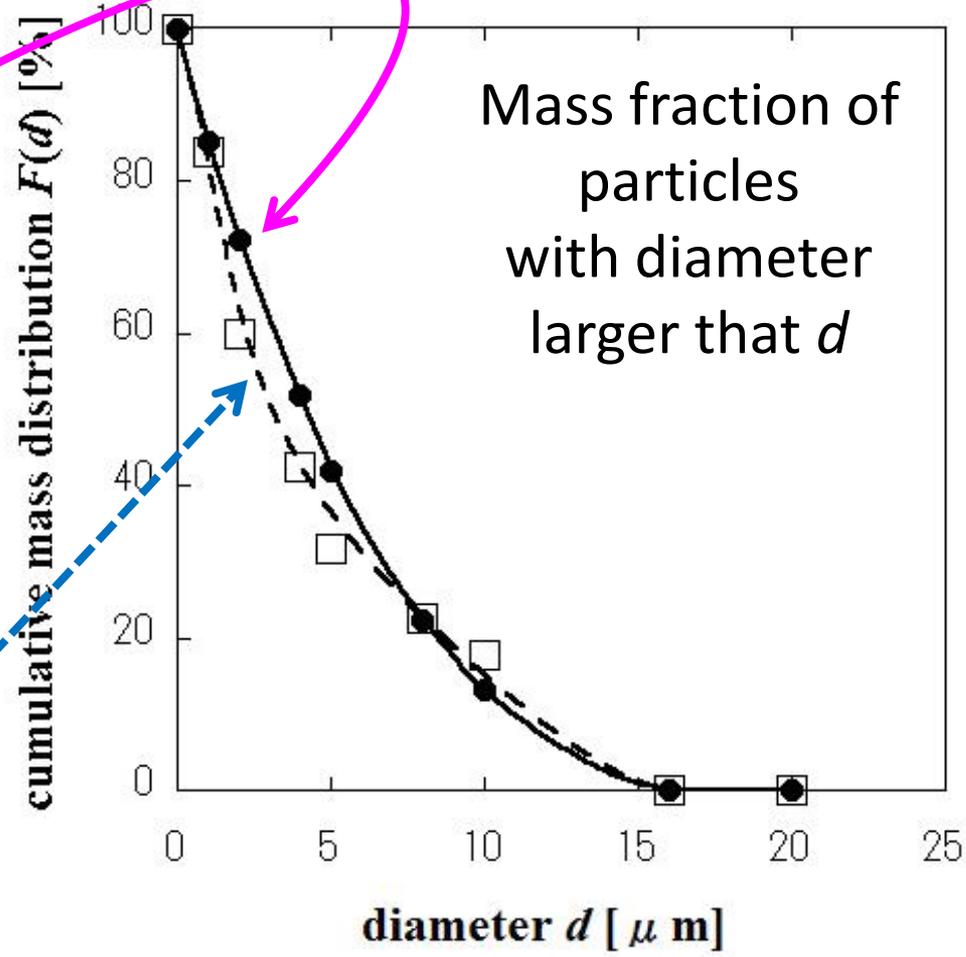
- A [no movement](#) → no experience → cellular cracks (no movement)
- B [vibration](#) → remember → Lamellar cracks (perpendicular)
- C [flow](#) → forget → Cellular cracks (no memory)
- C [turbulence](#) → forget → Cellular cracks (no memory)

# Why does $\text{CaCO}_3$ paste have **no memory of flow**?

$\text{CaCO}_3$   
Only memory of **vibration**



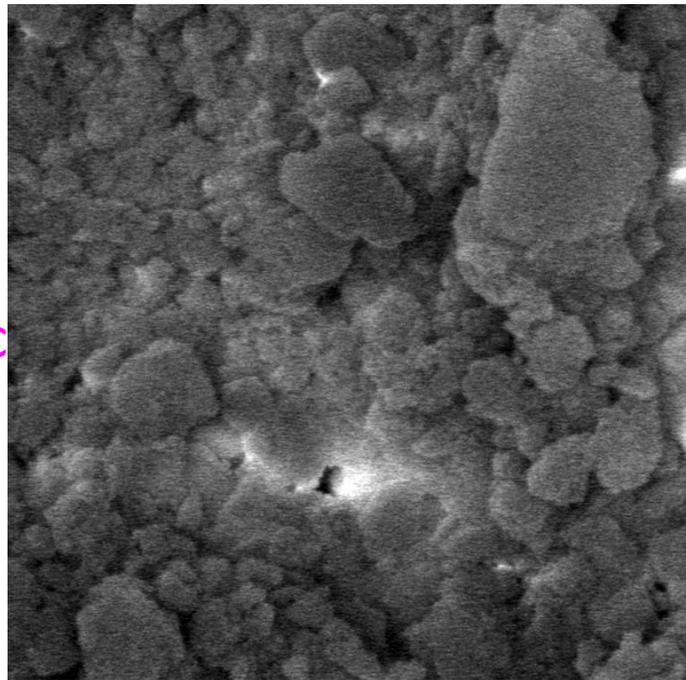
magnesium carbonate hydroxide  
Memory of **vibration** and **flow**



Similar size distribution

# Shape of particles

CaCO<sub>3</sub> paste has  
only memory of  
vibration



Rough

isotropic

1μm

magnesium carbonate hydroxide  
kaolin, carbon, ...  
who have  
memory of vibration  
+ memory of flow

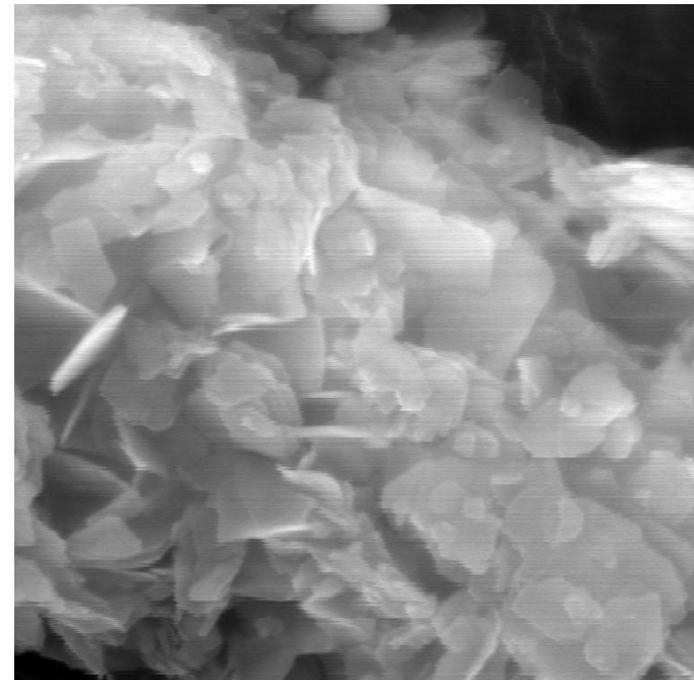


Plate-  
like

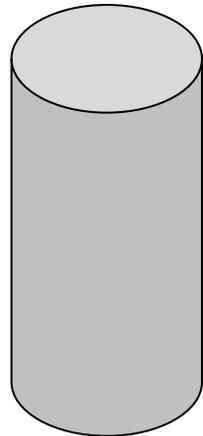
1μm

For memory of flow

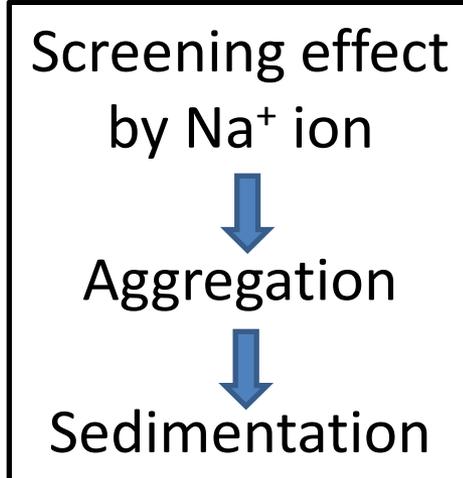
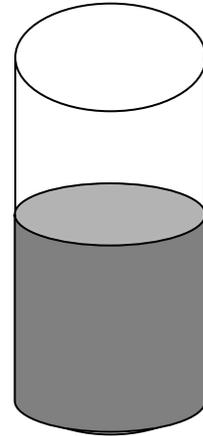
Shape dependence ?

# Coulombic repulsion between $\text{CaCO}_3$ particles

Dispersion of  $\text{CaCO}_3$  particles by Coulombic repulsion

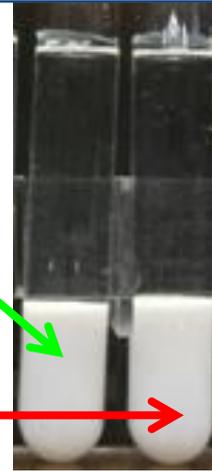
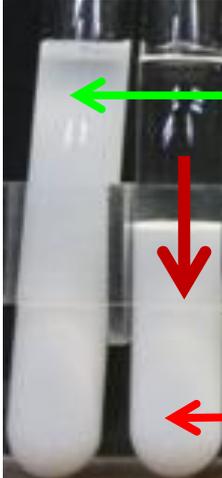


Add NaCl



$\text{CaCO}_3$   
(memory of only **vibration**)

magnesium carbonate hydroxide  
(memory of **vibration** and **flow**)



No NaCl

Add 0.1mol/l NaCl

**Charged**

Sedimentation any case

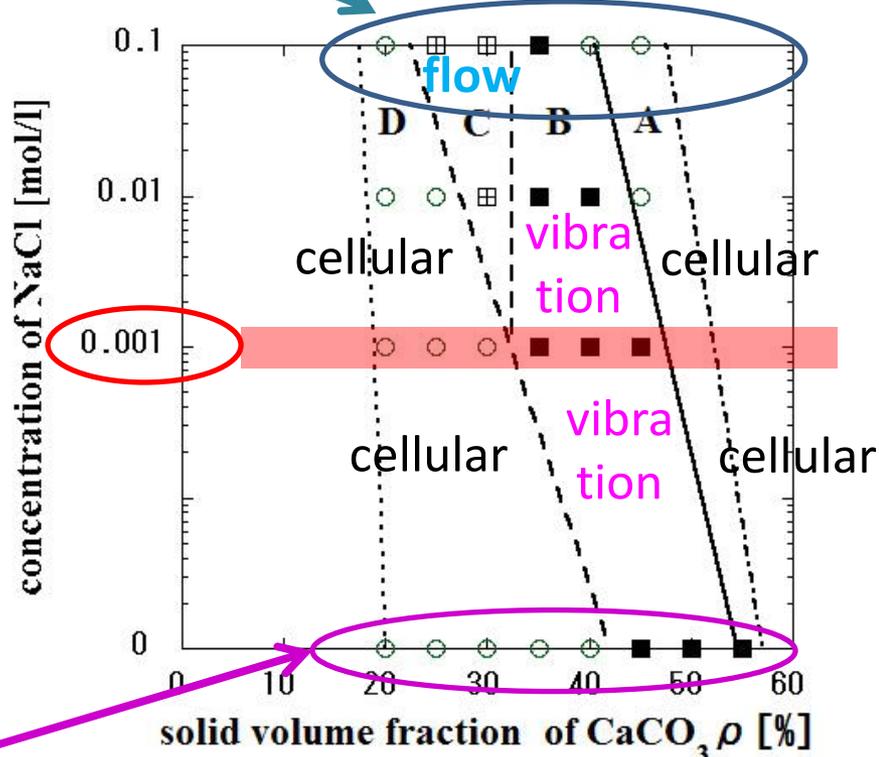
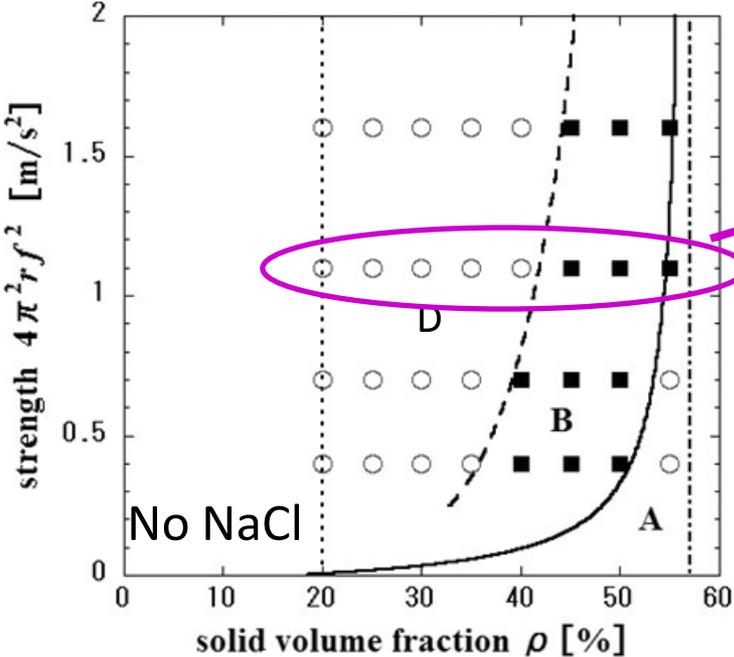
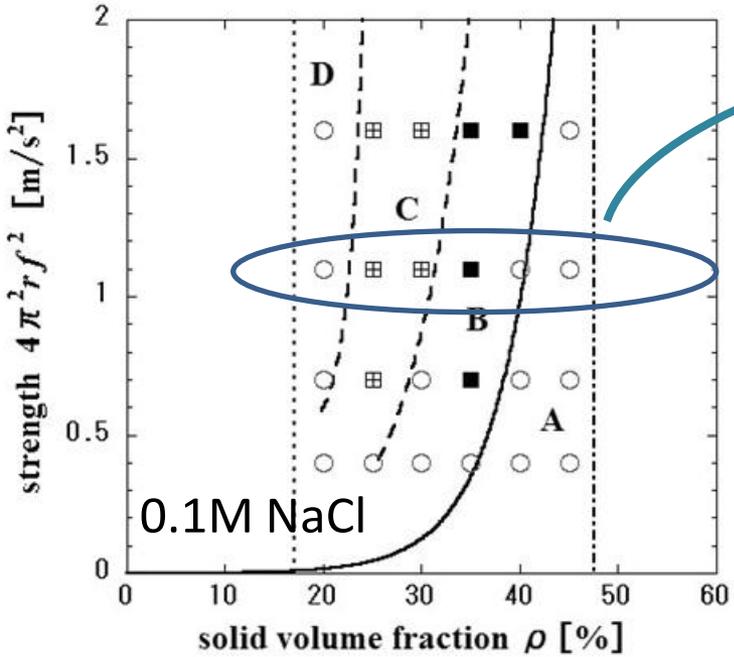
**Uncharged**

Particles interact with Coulombic repulsion

Particles interact without Coulombic repulsion

Sedimentation by adding NaCl

# Add NaCl and CaCO<sub>3</sub> paste can remember flow direction



What determines the value of this threshold?

- Debye length =  $0.304 / \sqrt{[\text{NaCl}]}$  (0.96nm for 0.1M)
- Mean particle radius =  $2\mu\text{m}$
- Mean interparticle distance =  $6\mu\text{m}$  (for  $\rho=30\%$ )
- Distance between surfaces of particles = ?
- Non-dimensional parameter ? Dynamics?

# Mechanism for memory of flow...Attractive interaction

Interaction surface charge	<b>van der Waals attraction</b>	<b>Repulsion No interaction</b>
charged		Coulombic repulsion $\times$ $\text{CaCO}_3$
<b>uncharged</b>	magnesium carbonate hydroxide 	$\times$

\* add NaCl

\*\*

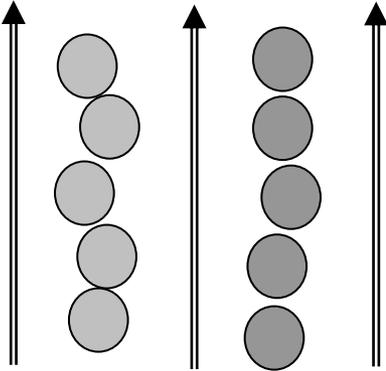
Dilute network is formed  
by van der Waals attraction

Attractive interaction is needed  
for the memory of flow.

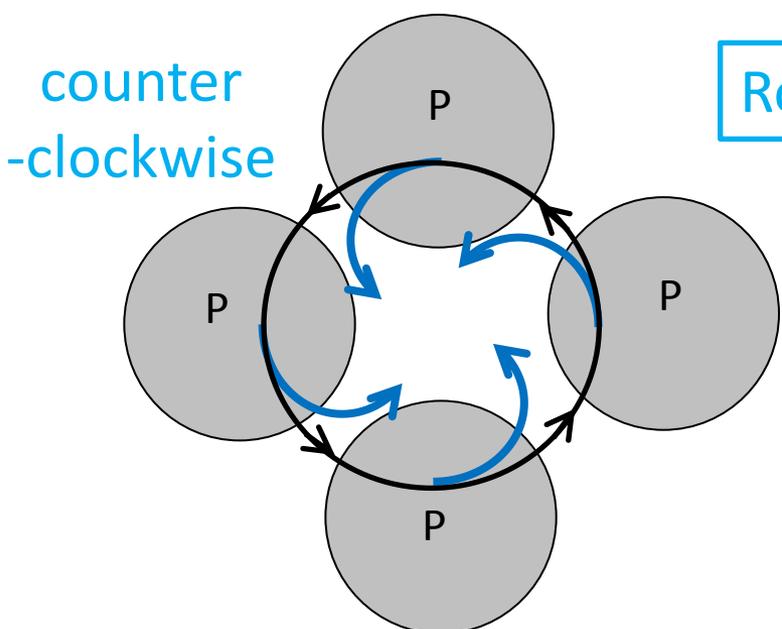
Flow → elongation of dilute network

\* Y. Matsuo and A. Nakahara  
JPSJ. 81 (2012) 024801.

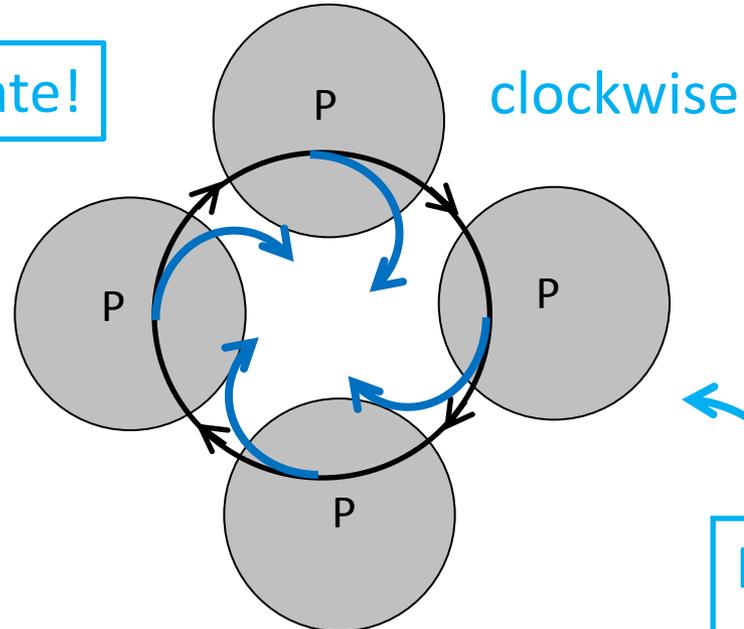
\*\* In preparation



# Spiral crack patterns made by memory of flow



Rotate!



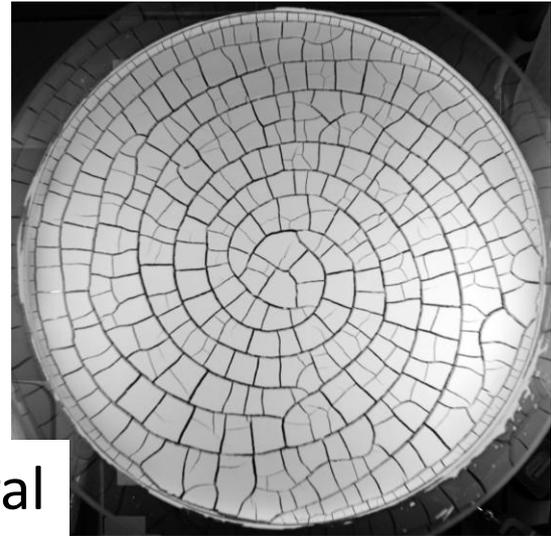
Stop rotation & dry

Stop rotation & dry

Parallel to final flow

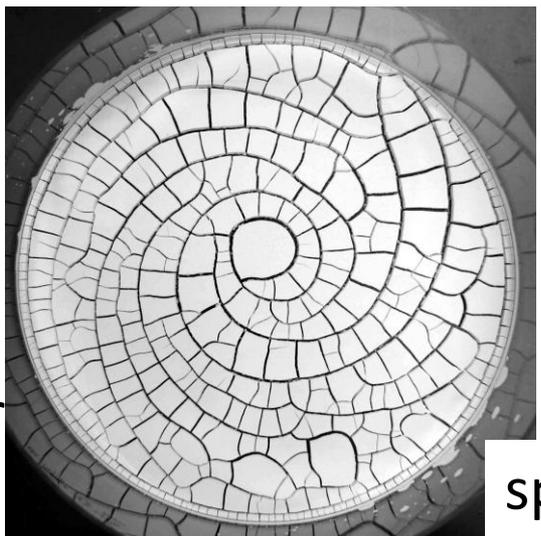
Memory of flow

movie



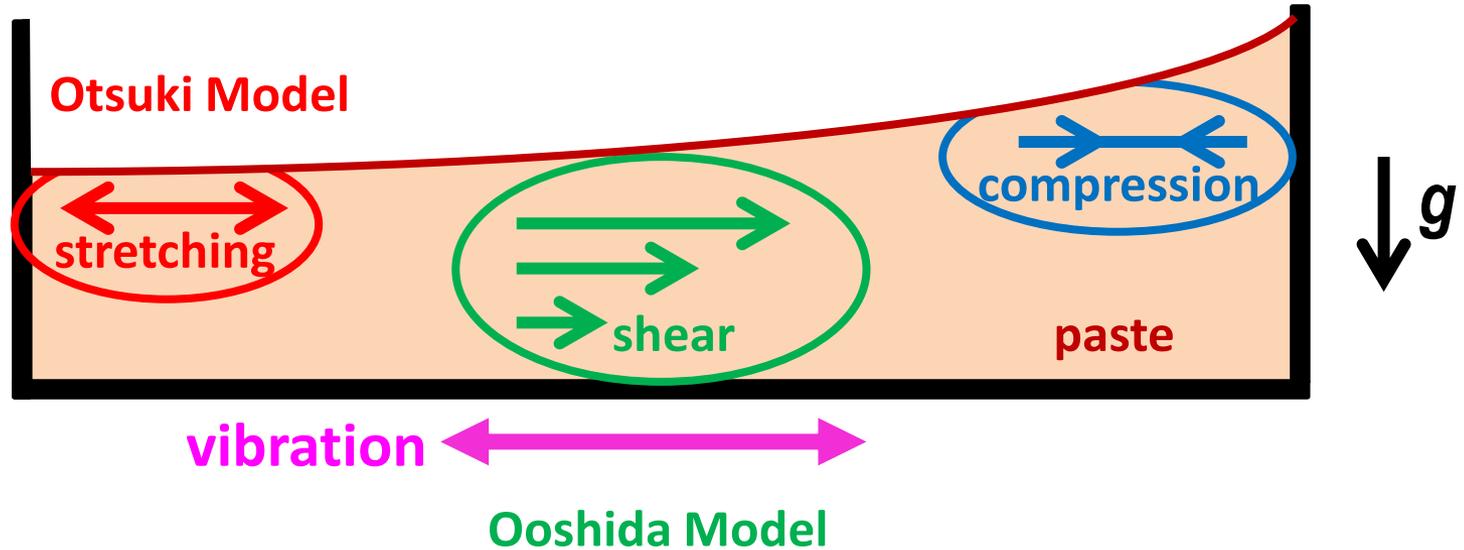
spiral

diameter 500mm



spiral

# Mechanism for memory of vibration...Residual tension



## Two Residual Tension Theories

**Model A: horizontal stretching mechanism**

**Non-uniform horizontal stretching, global model**

**M. Otsuki, PRE 72 (2005) 046114.**

**Model B: shear deformation mechanism**

**Longitudinal tension due to nonlinear elasticity,  
local model**

**Ooshida T., PRE77 (2008) 061501, JPSJ 78 (2009) 104801.**

# Model A: Otsuki Model on memory of vibration

Displacement in x direction,  $u(x, z, t)$

by M. Otsuki, PRE. 72 (2005) 046114.

$$\gamma \frac{\partial u(x, z, t)}{\partial t} = \frac{\partial \sigma_{xx}(x, z, t)}{\partial x} + \frac{\partial \sigma_{xz}(x, z, t)}{\partial z} + \alpha(t) \text{ vibration}$$

Plastic shear strain,  $s(x, z, t)$  (Bingham type)

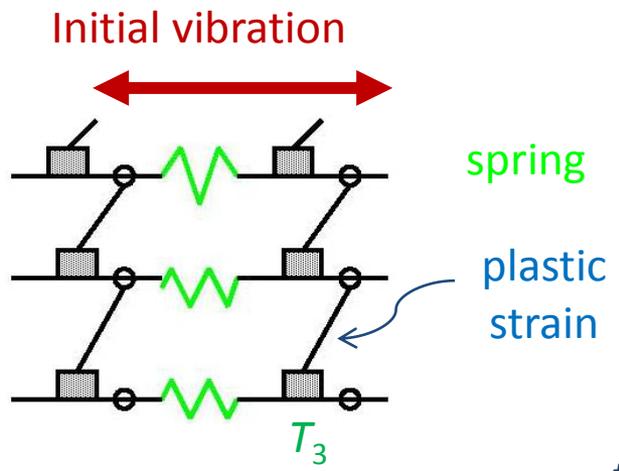
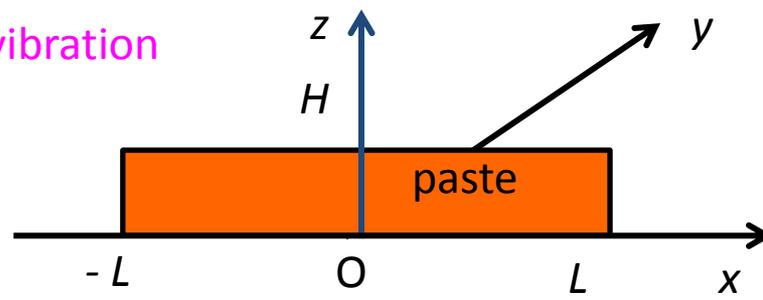
$$B \frac{\partial s(x, z, t)}{\partial t} = \begin{cases} 0 & |\sigma_{xz}| < \sigma_Y(t) \\ \left[ |\sigma_{xz}| - \sigma_Y(t) \right] \frac{\sigma_{xz}}{|\sigma_{xz}|} & |\sigma_{xz}| \geq \sigma_Y(t) \end{cases}$$

Normal stress,  $\sigma_{xx}(x, z, t)$  ( $\lambda, \mu$ ; Lamé coefficient)

$$\sigma_{xx}(x, z, t) = (\lambda + 2\mu) \left( \frac{\partial u(x, z, t)}{\partial x} + c(t) \right) \text{ shrinkage}$$

Shear stress,  $\sigma_{xz}(x, z, t)$

$$\sigma_{xz}(x, z, t) = \mu \left( \frac{\partial u(x, z, t)}{\partial z} - s(x, z, t) \right)$$



	0	$T_1$	$T_2$	$T_3$	$t$
External force	$\alpha(t) = \alpha_M \sin(\pi t / T_1)$	$\alpha(t) = 0$	$\alpha(t) = 0$	$\alpha(t) = 0$	$\alpha(t) = 0$
Yield stress	$\sigma_Y(t) = \sigma_{Y0}$	$\sigma_Y(t) = \sigma_{Y0}$	$\sigma_Y(t) = \infty$	$\sigma_Y(t) = \infty$	$\sigma_Y(t) = \infty$
Boundary cond.	$u(\pm L, z, t) = 0$	$u(\pm L, z, t) = 0$	$\sigma_{xx}(\pm L, z, t) = 0$	$\sigma_{xx}(\pm L, z, t) = 0$	$\sigma_{xx}(\pm L, z, t) = 0$
Reference strain rate	$c(t) = 0$	$c(t) = 0$	$c(t) = 0$	$c(t) = b(t - T_3)$	$c(t) = b(t - T_3)$
	vibration		relaxation	relaxation	dry
	plastic deformation			no plasticity	

# Model A: Otsuki Model on memory of vibration

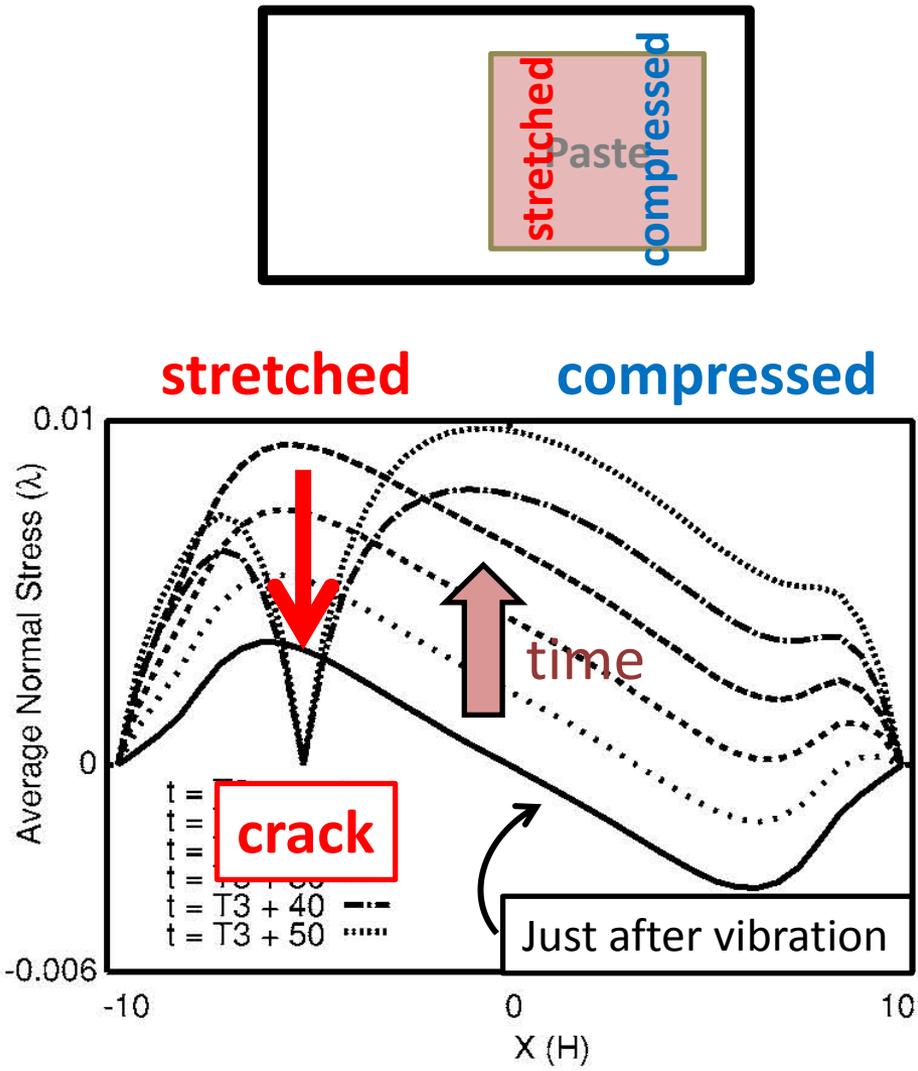
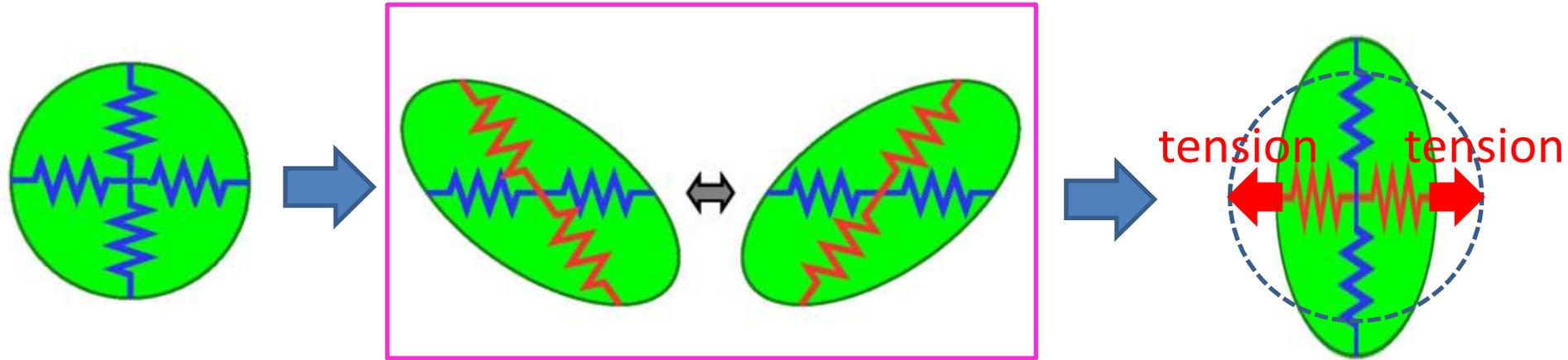
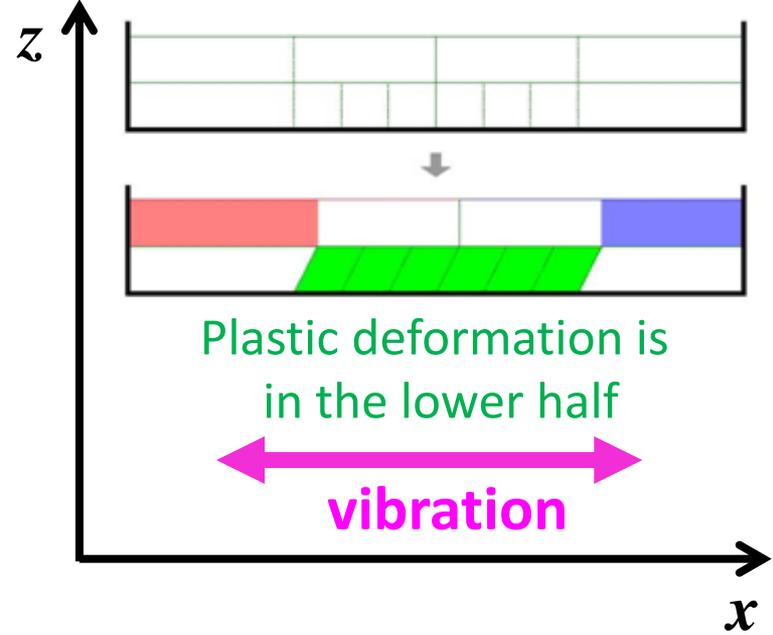


FIG. 6. The average normal stress  $\langle \sigma_{xx}(x,t) \rangle$  in the case  $\alpha_M=0.08$  as a function of  $x$ .

# Model B: Ooshida model

shear deformation mechanism

Longitudinal tension due to nonlinear elasticity



Vibration

||

Repeated shear  
(oblique stretching)

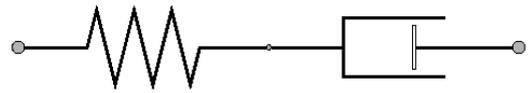
Horizontal  
*x*-directional  
tension remains

# Model B: Ooshida Model on memory of vibration

## Continuum model of a paste

Ooshida (2008) PRE 77; Ooshida (2009) JPSJ 78

- a kind of nonlinear Maxwell model of viscoelasticity

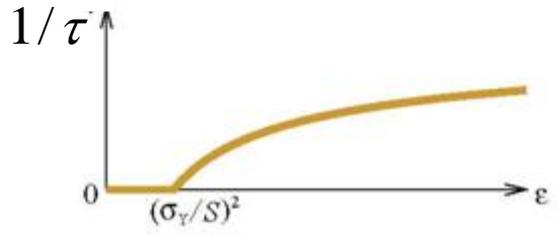


- formulation with label variable  $\xi = (\xi, \zeta)$  and natural metric  $g^{\natural} = (g^{\natural}_{ij}) = (g^{\natural ij})^{-1}$  (cf.  $T = \kappa(x - x^{\natural})$ )

$\mathbf{P} = P^{ij}(\partial_i \mathbf{r}) \otimes (\partial_j \mathbf{r}), \quad P^{ij} = \tilde{p}g^{ij} + S(g^{ij} - g^{\natural ij}) \quad (1)$ <p style="margin: 0;">stress <span style="margin-left: 100px;">convected basis</span></p> $\partial_t g^{\natural ij} = -\nu g^{\natural ij} + \nu_* g^{ij} \quad (2)$
---

$\mathbf{r} = \mathbf{r}(\xi, t), \quad g^{\natural ij} = g^{\natural ij}(\xi, t); \quad g_{\xi\zeta} = (\partial_{\xi} \mathbf{r}) \cdot (\partial_{\zeta} \mathbf{r})$  etc.;  $(g^{ij}) = (g_{ij})^{-1}$

- plasticity: relaxation time  $\tau$  can diverge depending on  $\varepsilon = g_{ij}g^{\natural ij} - n_d$   
elastic energy



# Model B: Ooshida Model on memory of vibration

## Equation of motion

For momentum  $\rho \mathbf{v}$  ( $i = \xi, \eta$ ;  $\mathbf{F} = F_x(t)\mathbf{e}_x - G\mathbf{e}_z$ )

$$\rho \left( \partial_t v^i + v^j \nabla_j v^i \right) = -\nabla_j \underbrace{P^{ij}}_{\text{stress}} + \underbrace{F^i}_{\text{forcing}} \quad (3)$$

All vectors/tensors in **convected coordinate system**

$$\mathbf{v} = \sum v^i \partial_i \mathbf{r} = \sum v_i \nabla \xi^i \quad \text{etc.} \quad \nabla_j v^i = \partial_j v^i + \Gamma_{jk}^i v^k$$

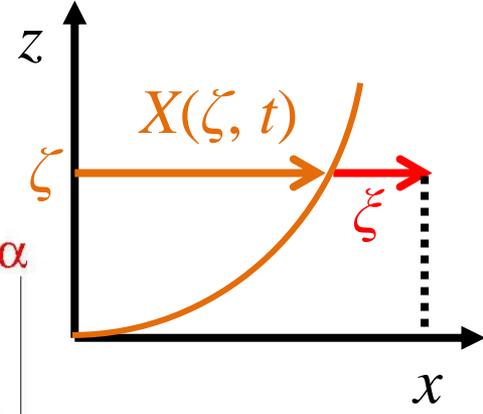
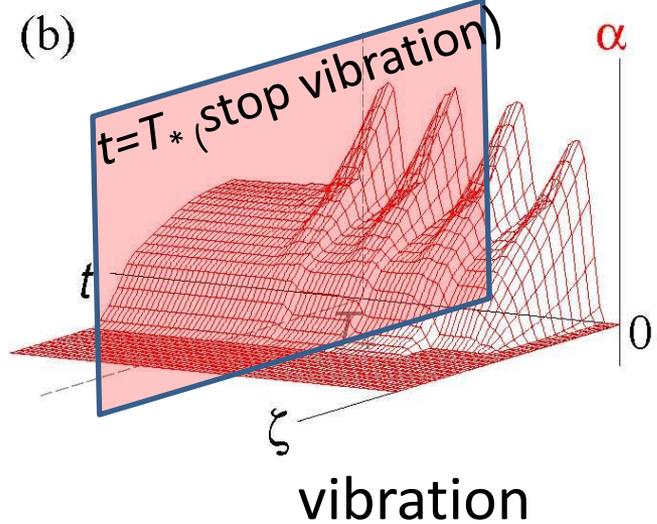
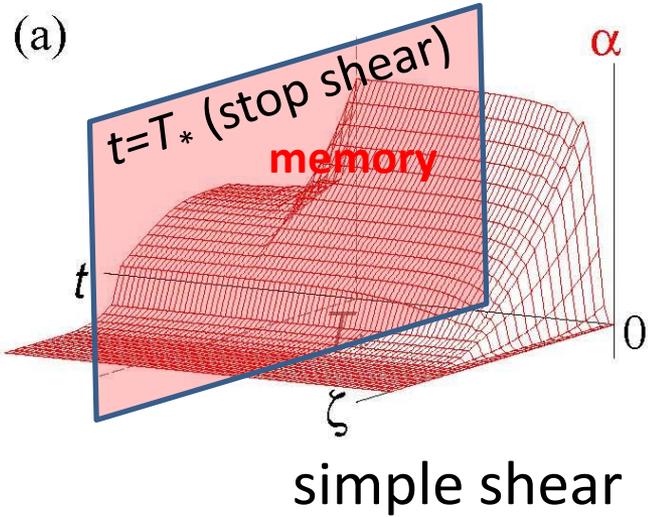
contravariant          covariant          covariant derivative

## Boundary conditions

- free surface at  $\zeta = \zeta_{\max}$
- no-slip condition at  $\zeta = 0$

# Model B: Ooshida Model on memory of vibration

## Residual tension theory in parallel flow setup



$\alpha = (\xi\text{-directional contraction of natural metric}) > 0$

- memory written ( $0 < t < T_*$ , flowing state)
- memory retained ( $t \rightarrow +\infty$ , solidified state)

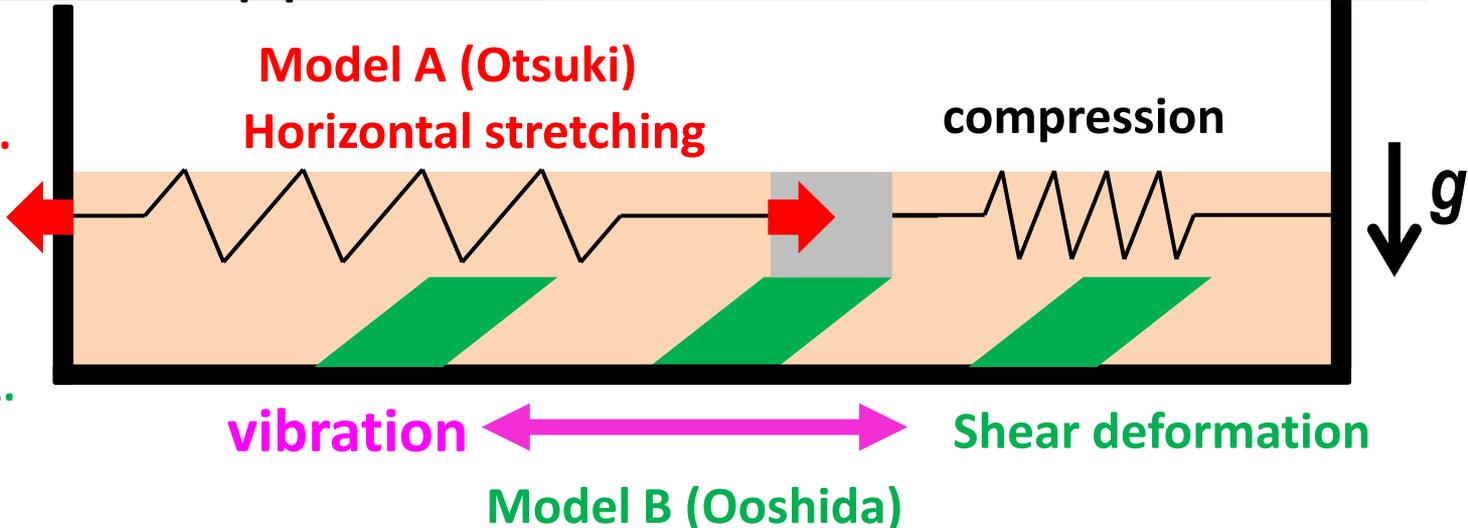
[Movie!](#)

$$\sigma_{ij} = \begin{pmatrix} e^{-\alpha} & \beta \\ \beta & (1+\beta^2) e^{\alpha} \end{pmatrix}$$

# Where will cracks appear in Residual tension models?

M. Otsuki,  
PRE 72 (2005) 046114.

Ooshida T.,  
PRE77 (2008) 061501,  
JPSJ 78 (2009) 104801.



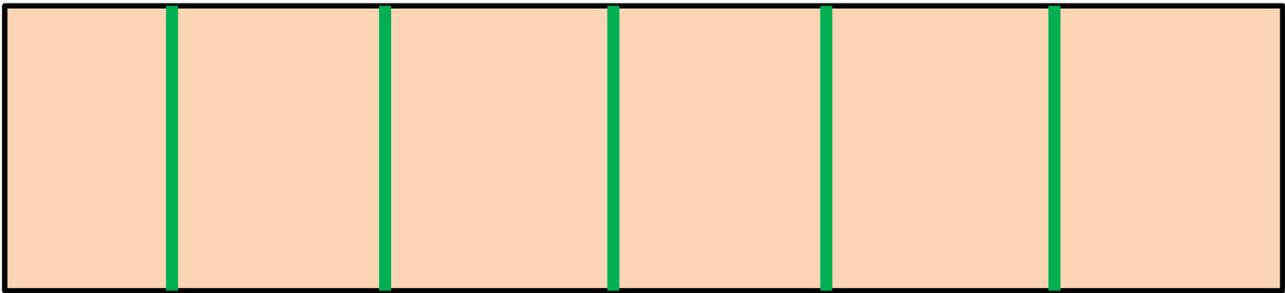
Crack here

Otsuki Model  
horizontal stretching



Cracks anywhere

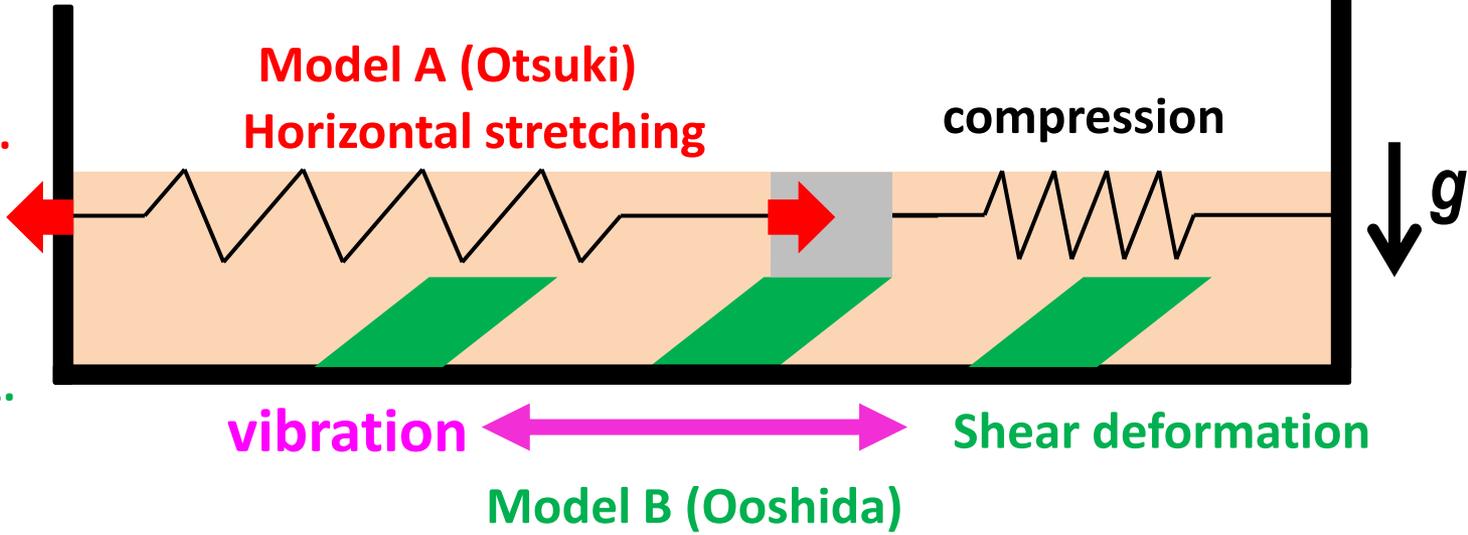
Ooshida Model  
shear deformation



# Experimental verification of two residual tension models

M. Otsuki,  
PRE 72 (2005) 046114.

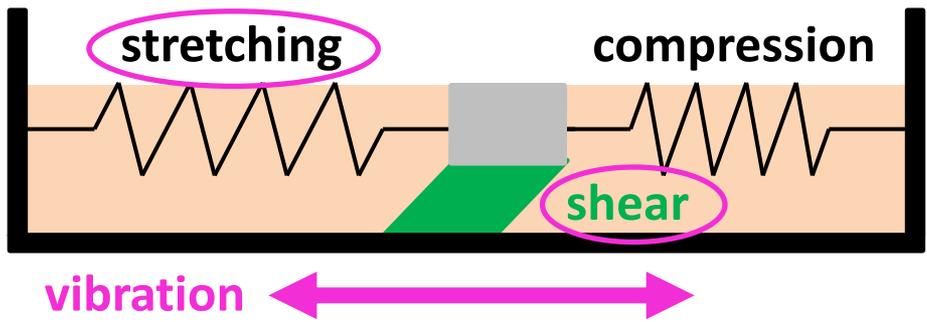
Ooshida T.,  
PRE77 (2008) 061501,  
JPSJ 78 (2009) 104801.



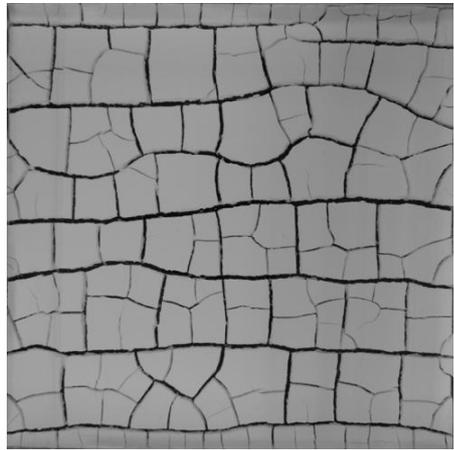
Overwriting	Where do the perpendicular cracks appear?
1/2 shake	Stretched part (Model A by Otsuki)
2 shakes	Sheared part (Model B by Ooshida)

# Conclusion 1

● Memory of vibration = residual tension

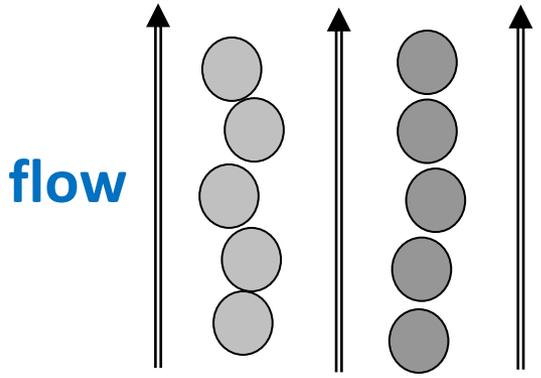


Water-poor

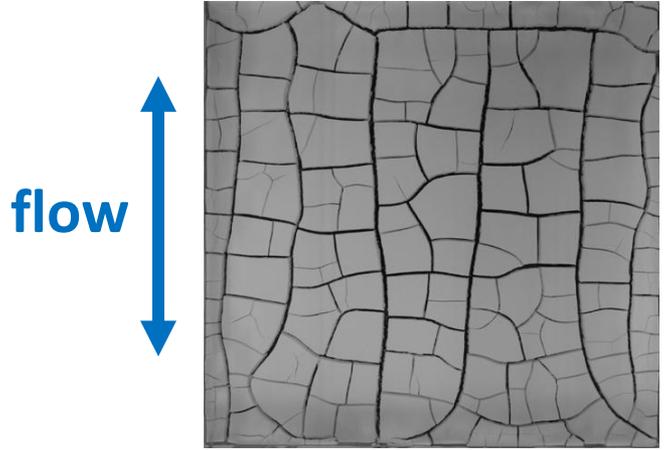


● Memory of flow = elongation of dilute network under flow

Attractive interaction is needed.



Uncharged  
Water-rich

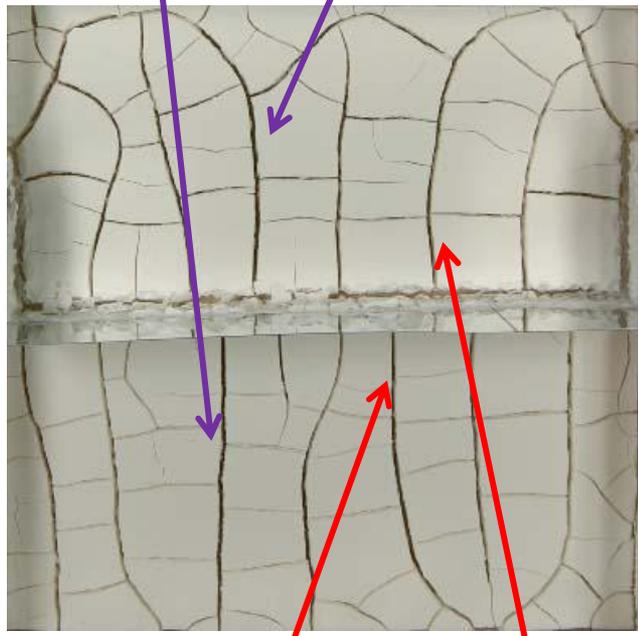
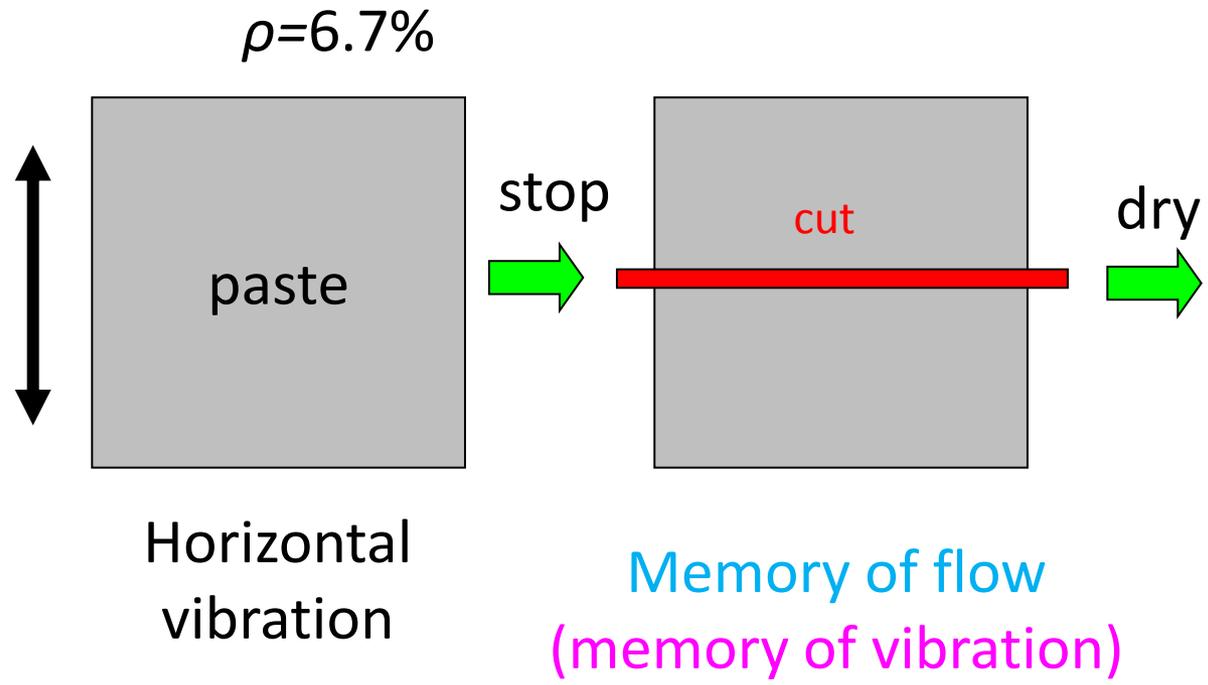


# Can we control position of cracks?

**By horizontal vibration... Only Direction!**

Direction: imprinted by memory effect  
Position : stochastically formed

direction . . . by same memory



position . . . stochastic

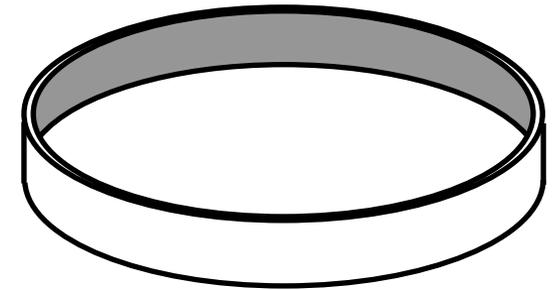
Memories are formed not by macroscopic hydrodynamic structure like convection.

# Position control of cracks using Faraday waves

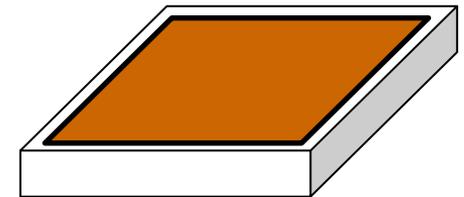
## Experimental setup for vertical vibration

Paste: Water-poor  $\text{CaCO}_3$  paste (only memory of vibration)

vertical vibration

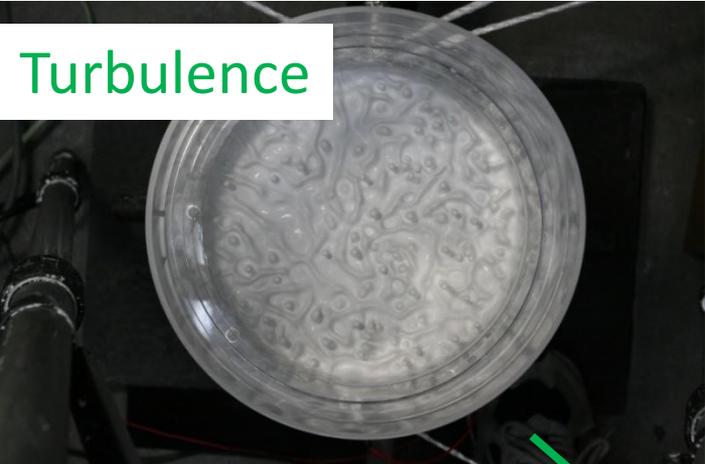
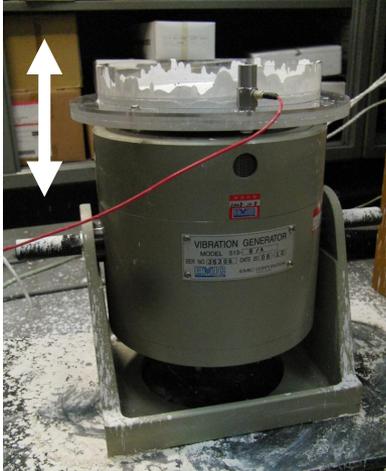


Circular container  
diameter  $d = 200$  mm



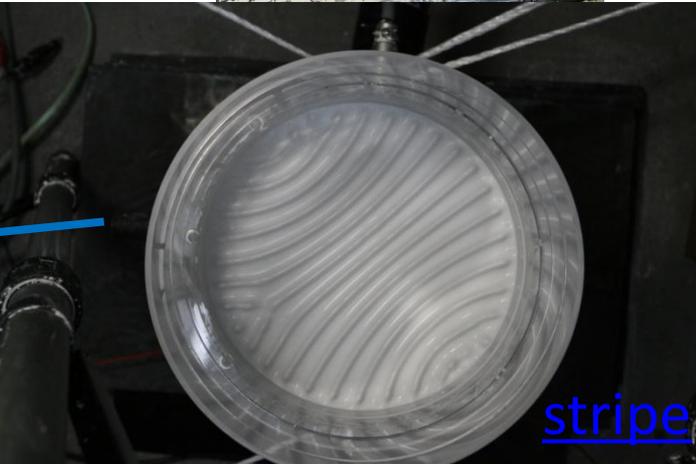
Square container  
150mm side

# Position control by vertical vibration



Turbulence

Vertical vibration  
(Faraday waves)  
↓  
Stop and dry!



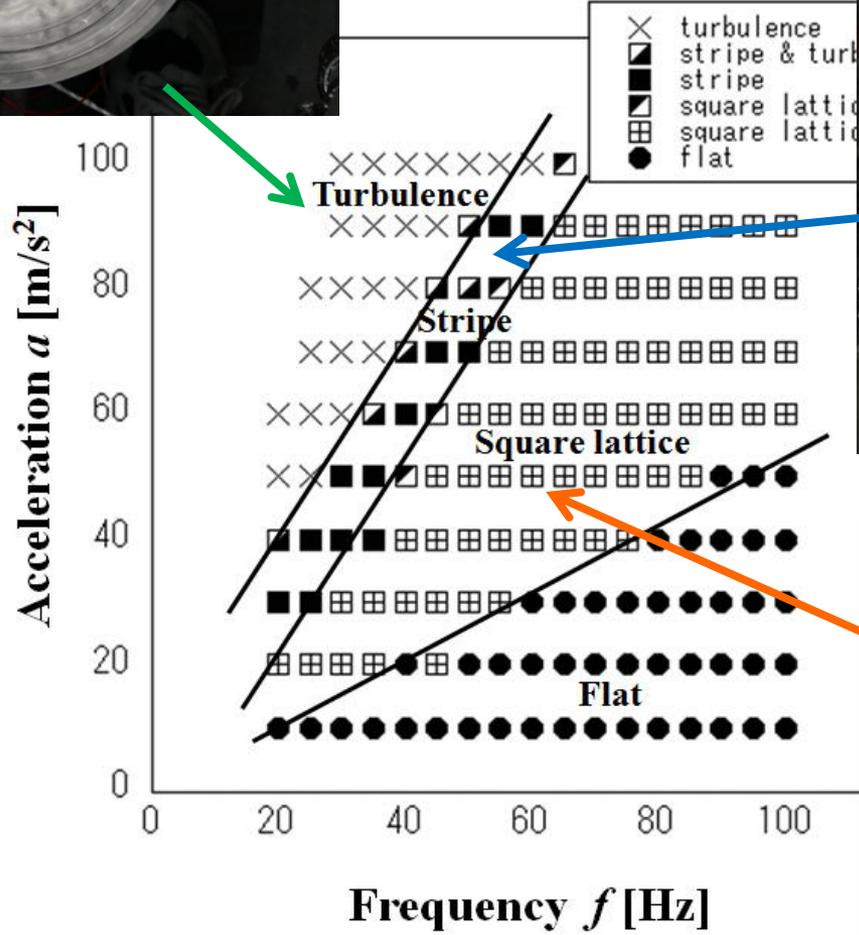
stripe



Square lattice

diameter  
 $d = 200$  cm

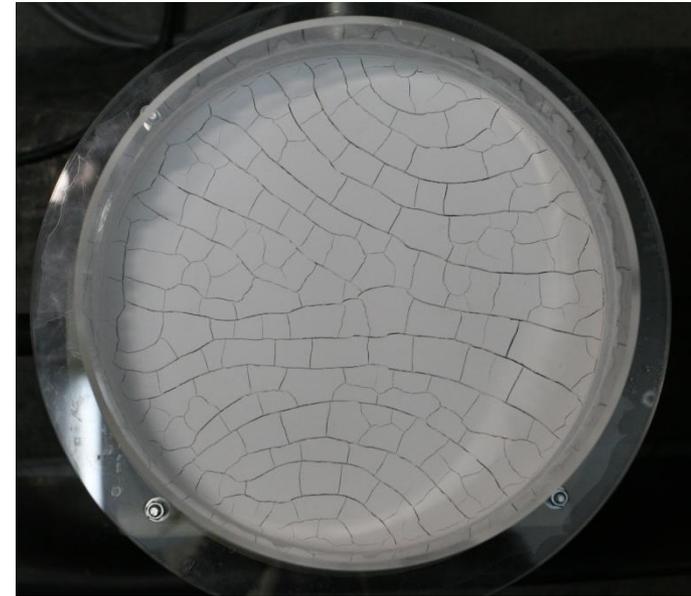
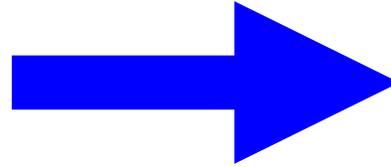
Water-poor  
 $\text{CaCO}_3$  paste  
which has  
only  
memory of  
vibration



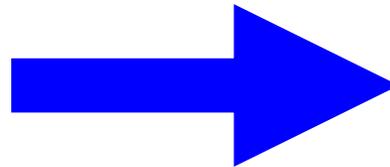
# Stripe and ring cracks produced **by vertical vibration**

Vertical vibration → Faraday waves

desiccation crack patterns



Stop vibration  
& dry

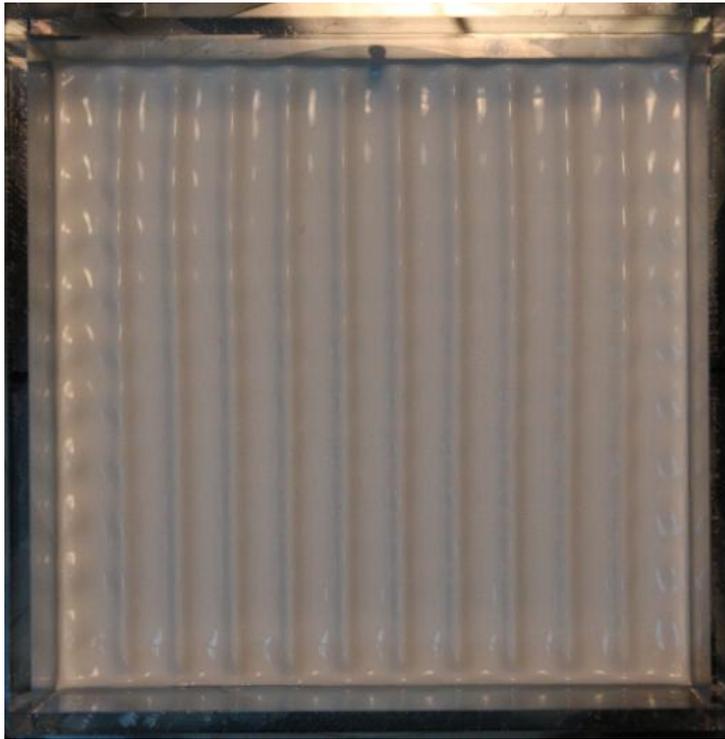


# Spatial correlation between Faraday waves and cracks

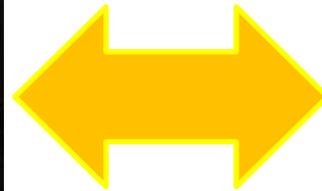
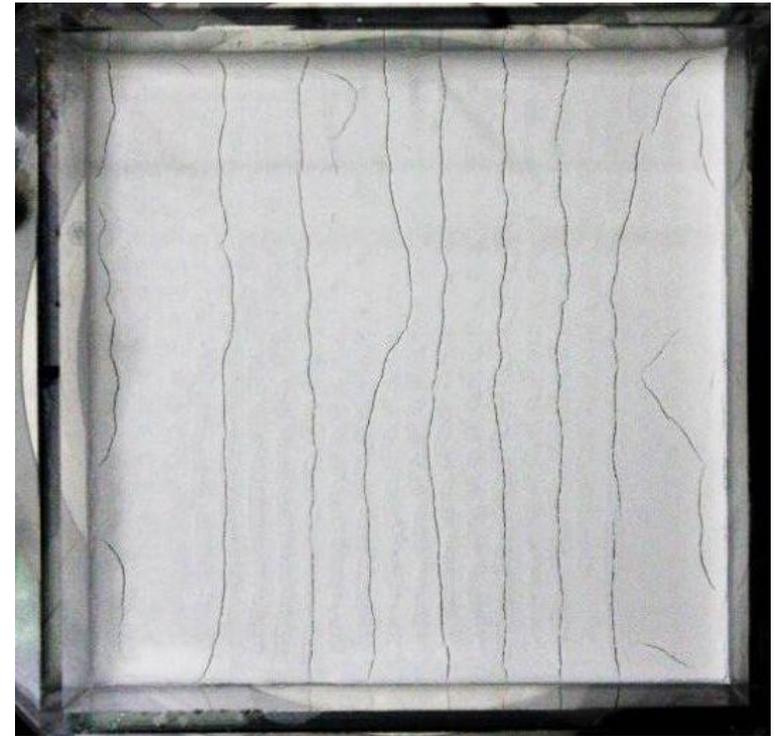
Where do cracks appear?

At Anti-Node regions or **at Node regions?**

Faraday waves

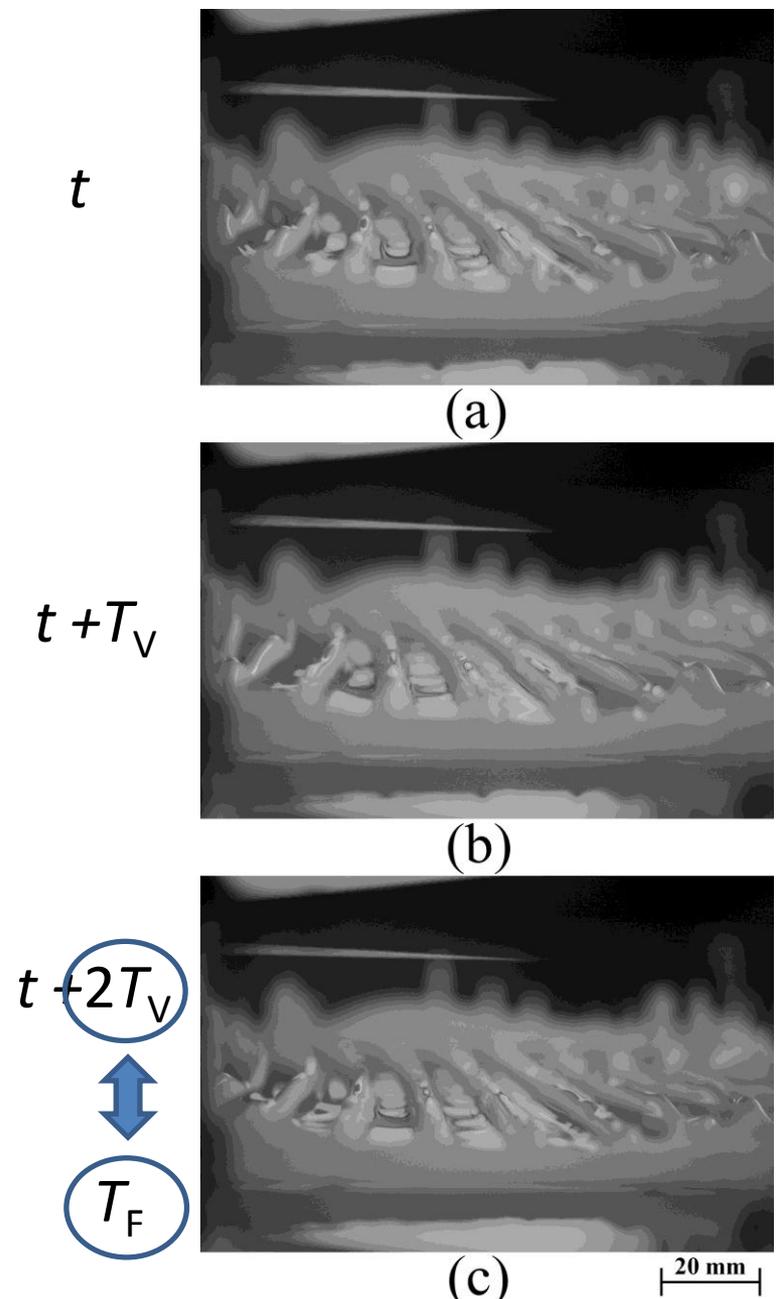


Desiccation crack pattern

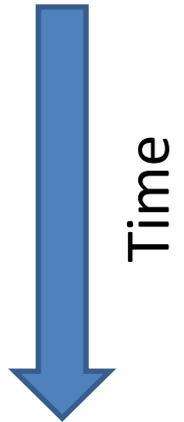


overlap

# Structure of Faraday waves of $\text{CaCO}_3$ paste



Faraday waves  
at each snapshot

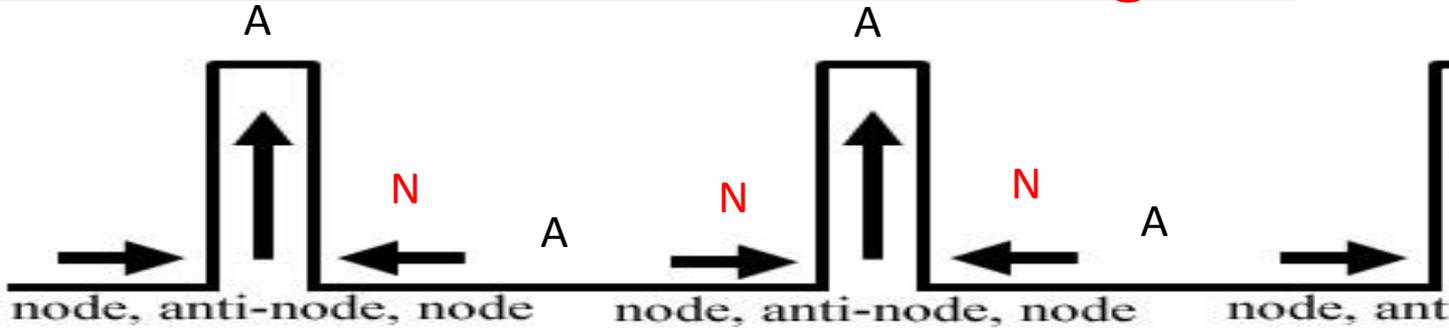


Period of Faraday waves  $T_F$   
 $= 2 \times$  Period of vertical vibration  $T_V$   
Side view : Square waves

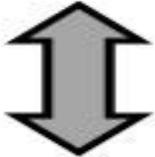
# Prediction of Horizontal oscillation in node regions

Schematic

Faraday waves at each snapshot

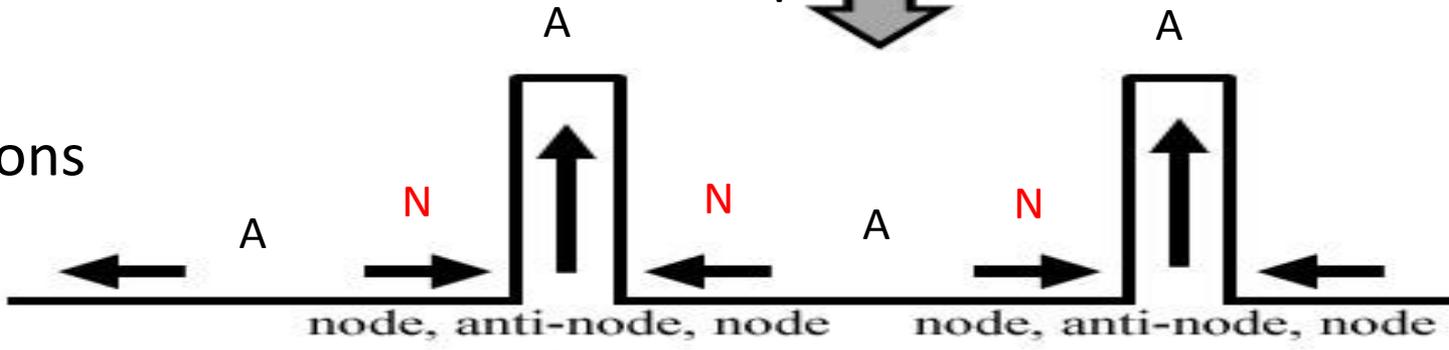


Period of vertical vibration  $T_V$

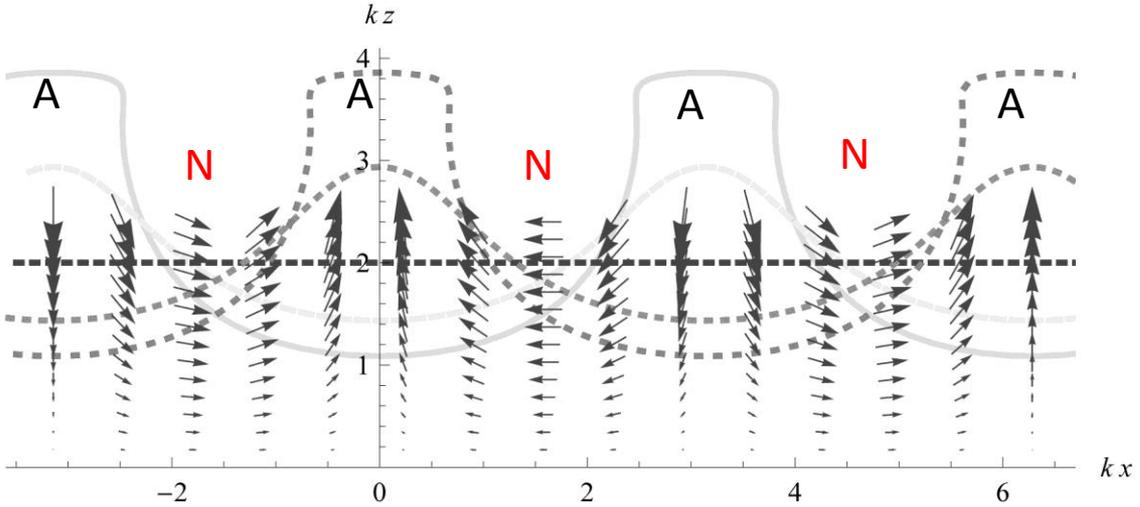


N : Node regions

A : Anti-node regions

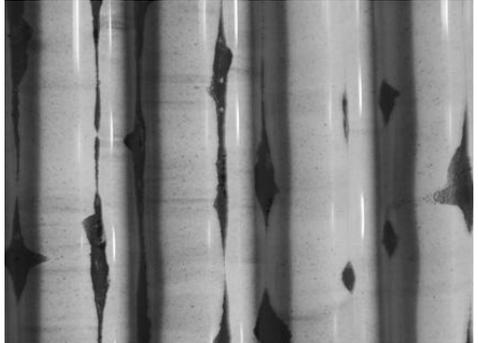


Analytic

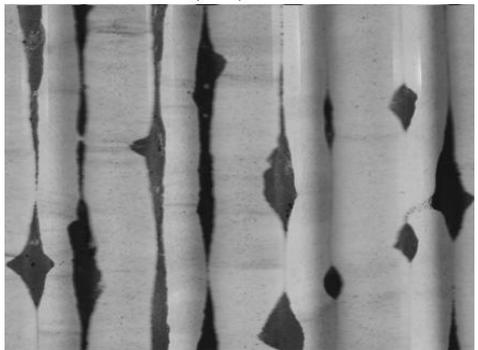


# Experimental observation of horizontal oscillation in node regions

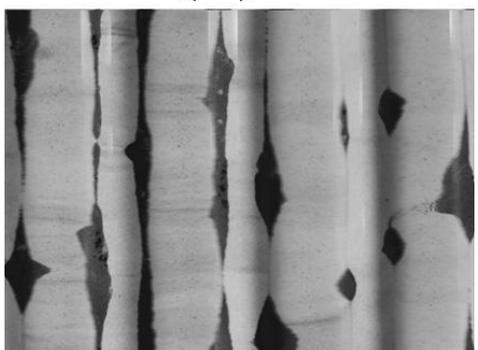
Faraday waves  
at each snapshot



(a)

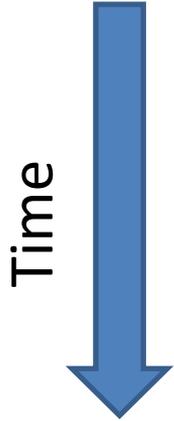


(b)



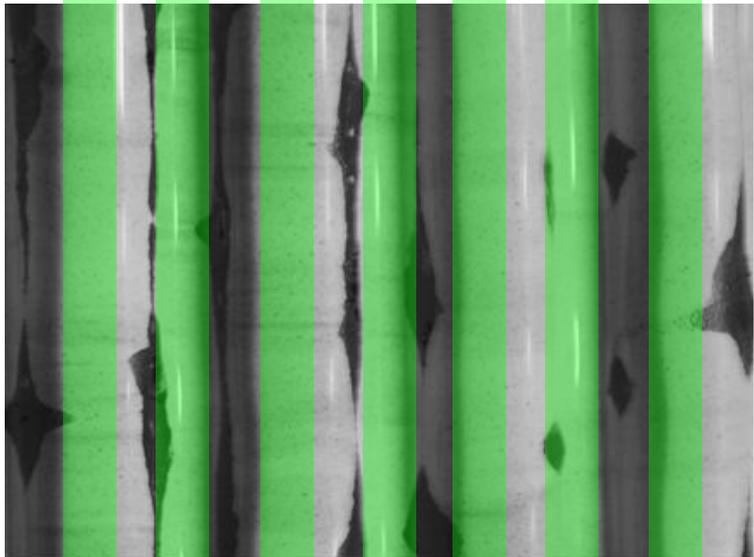
(c)

10 mm



$t$   
 $t + T_V$   
 $t + 2T_V$   
 $T_F$

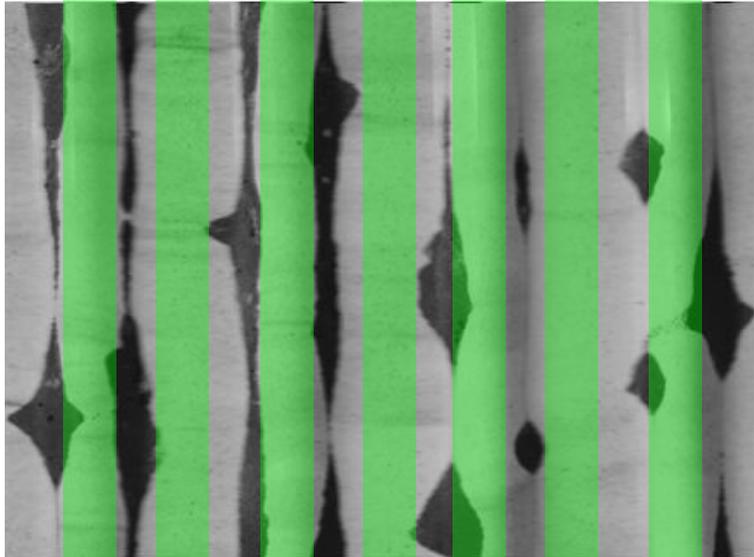
ANANANANANAN



Black carbon powder on  $\text{CaCO}_3$  paste

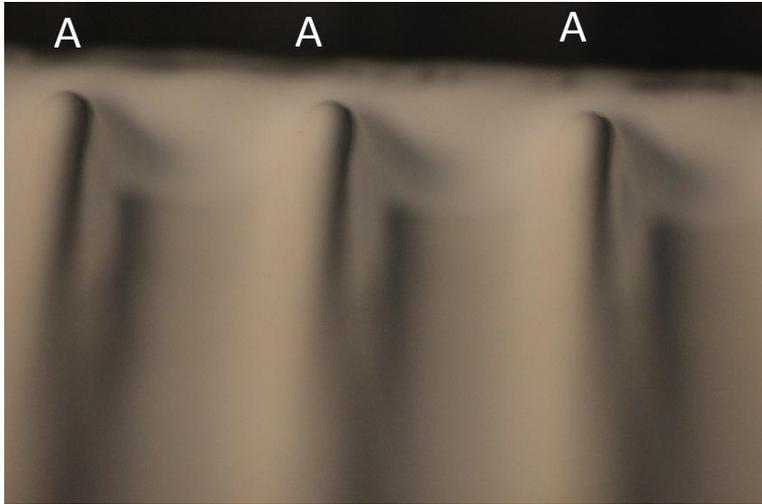


Horizontal vibration

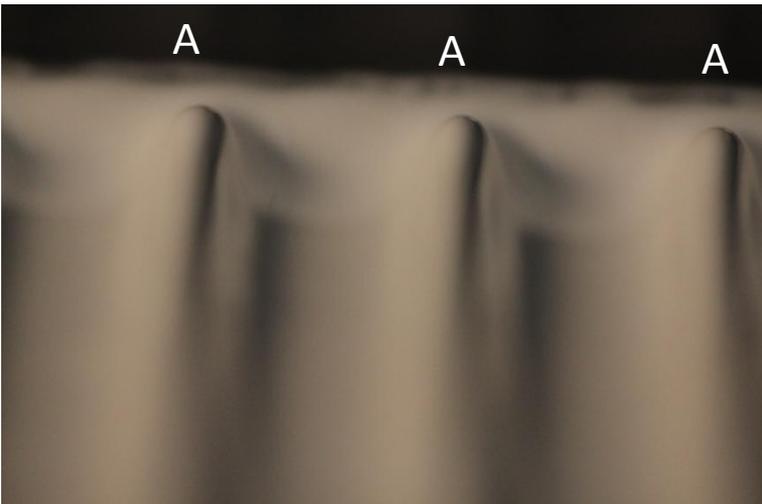


# Visualization of **node regions (N)** of Faraday waves

## Faraday waves at each snapshot

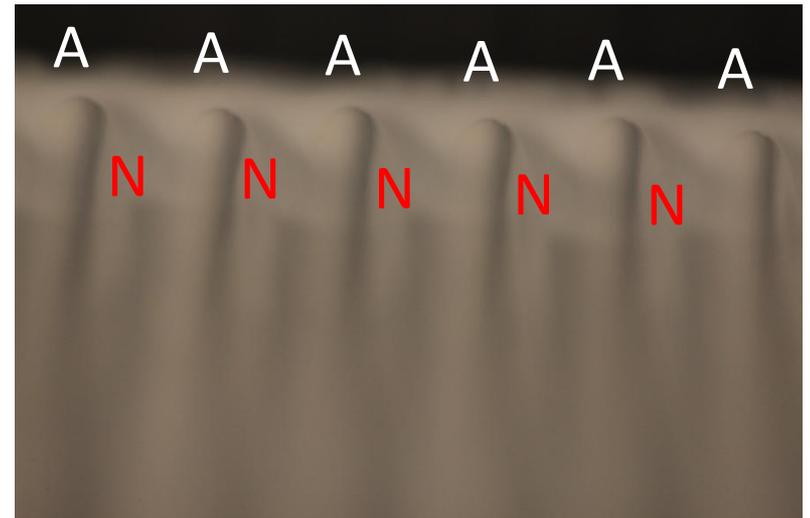


↕ Period of vertical vibration  $T_v$



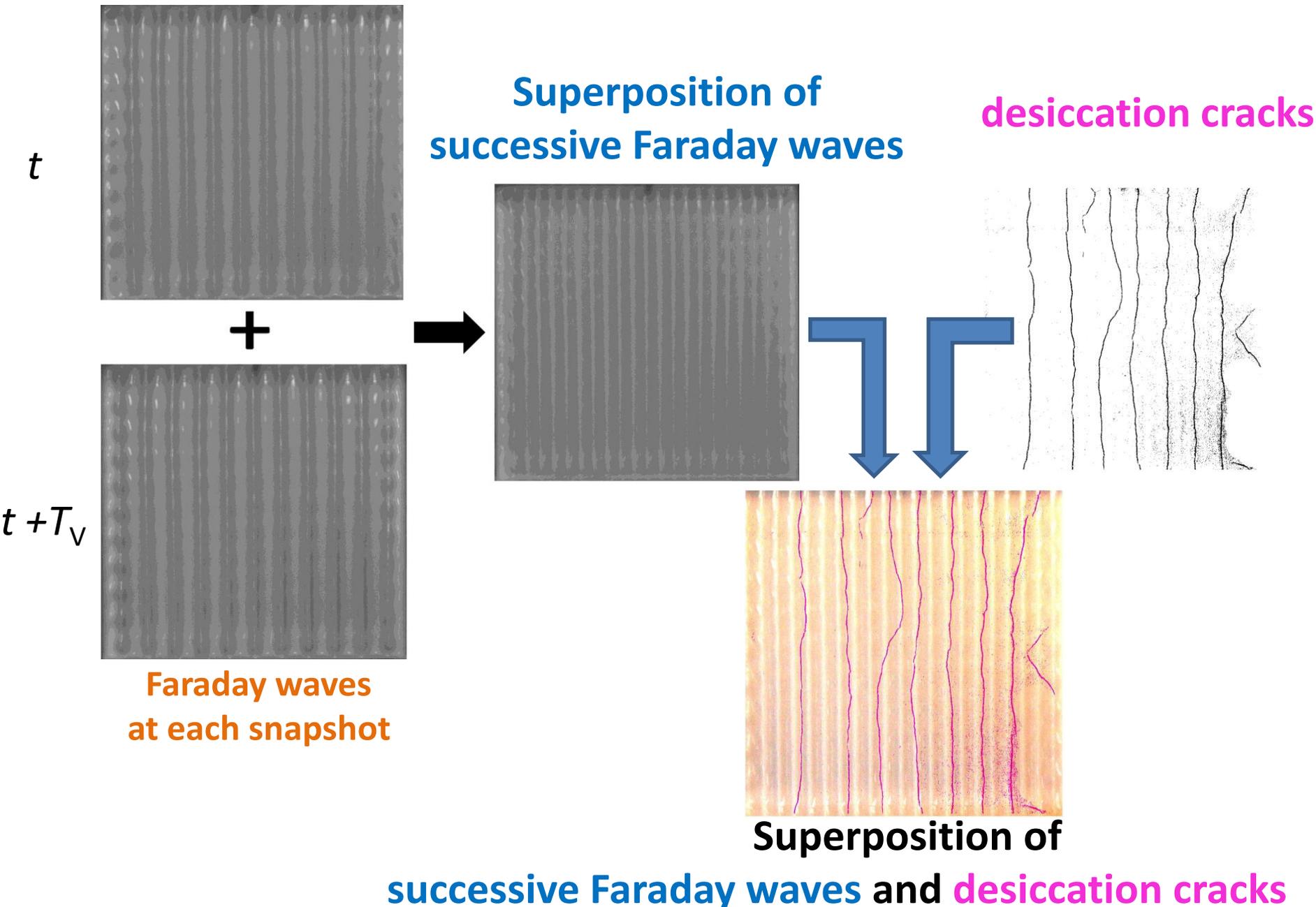
A : Anti-node regions  
(Regions where anti-nodes stand up alternatively)

N : Node regions  
(Always flat regions)



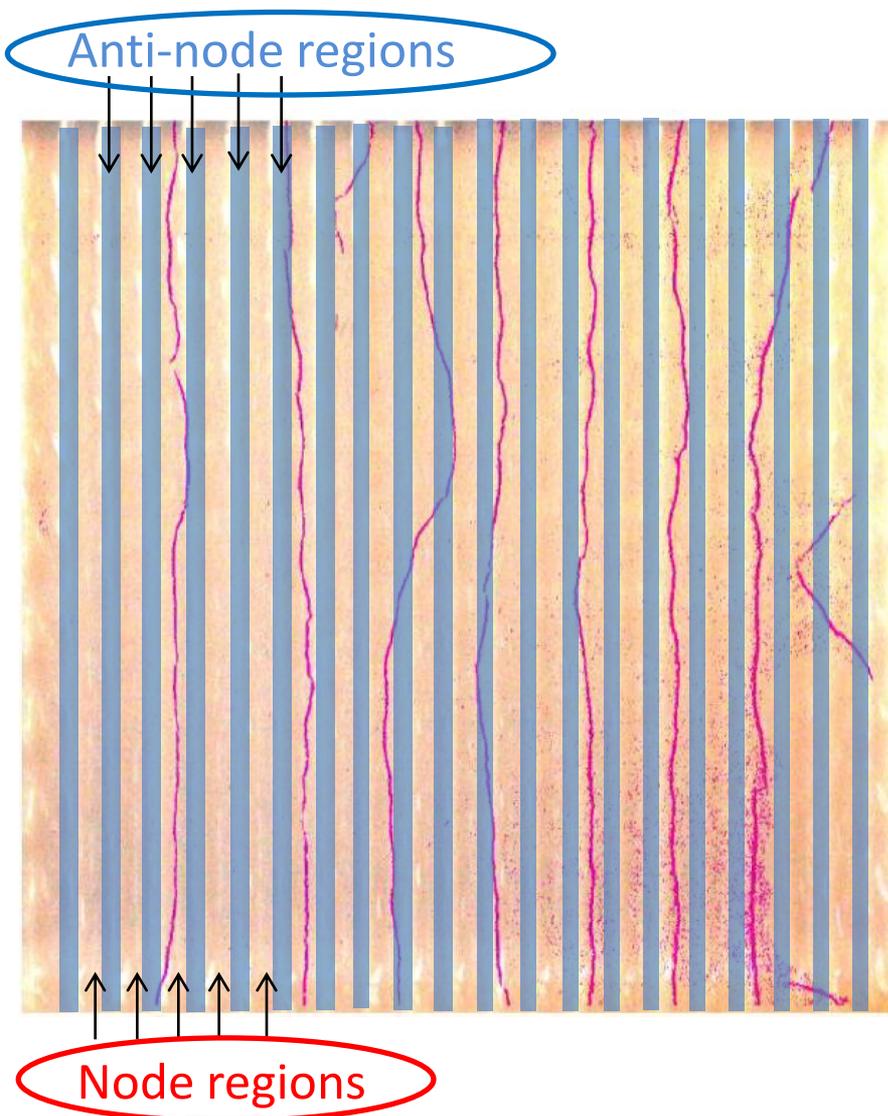
**Superposition of successive Faraday waves**

# Superposition of successive Faraday waves and desiccation cracks



# Statistical analysis

Ratio of observing cracks **at node regions** to that of anti-node regions



$$P = \log_e \frac{L_n / S_n}{L_a / S_a}$$

node regions

anti-node regions

$L_n$  : length of cracks at **node regions**

$L_a$  : length of cracks at **anti-node regions**

$S_n$  : Area of **node regions**

$S_a$  : Area of **anti-node regions**

From 10 experiments

$$P = 0.76 \pm 0.40$$

**At node regions**, cracks are formed

$$e^P = e^{0.76} = 2.1 \text{ times}$$

more frequently than in anti-node regions.

Cracks emerge **at node regions**.

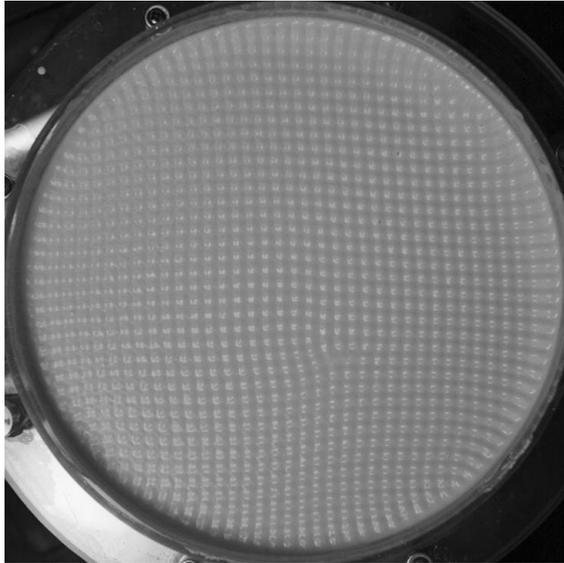
# Square lattice cracks produced **by vertical vibration**

Vertical vibration

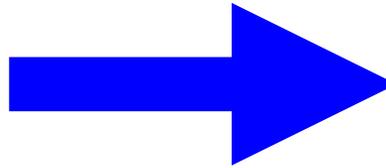
→ Faraday waves

desiccation crack patterns

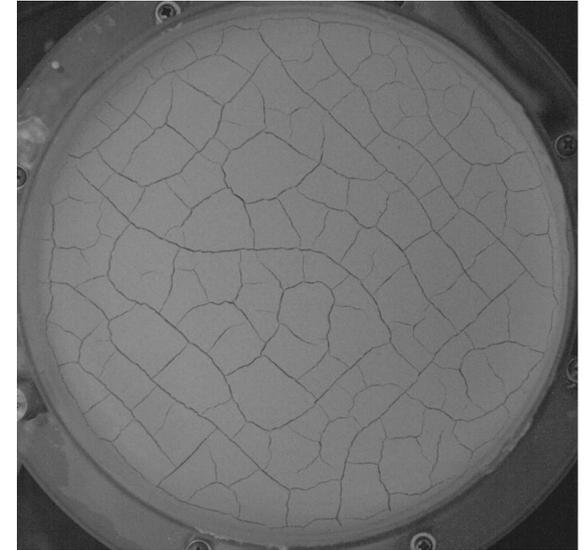
Square  
lattice



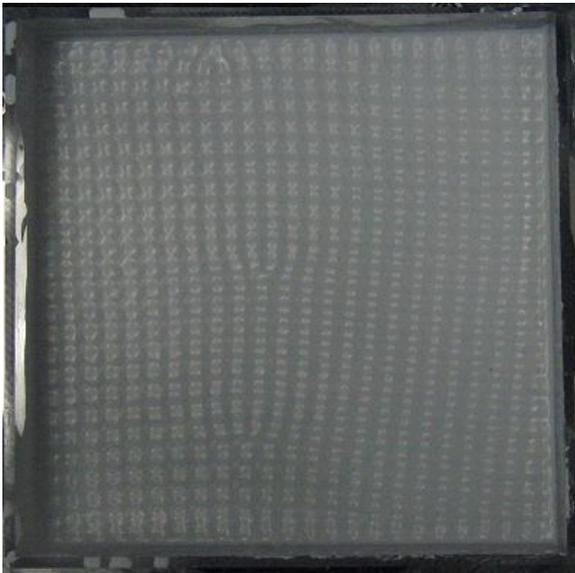
Stop & dry



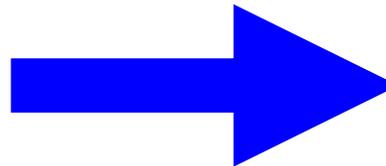
Why tilt by 45° ?



Square  
lattice



Stop & dry



Why tilt by 45° ?



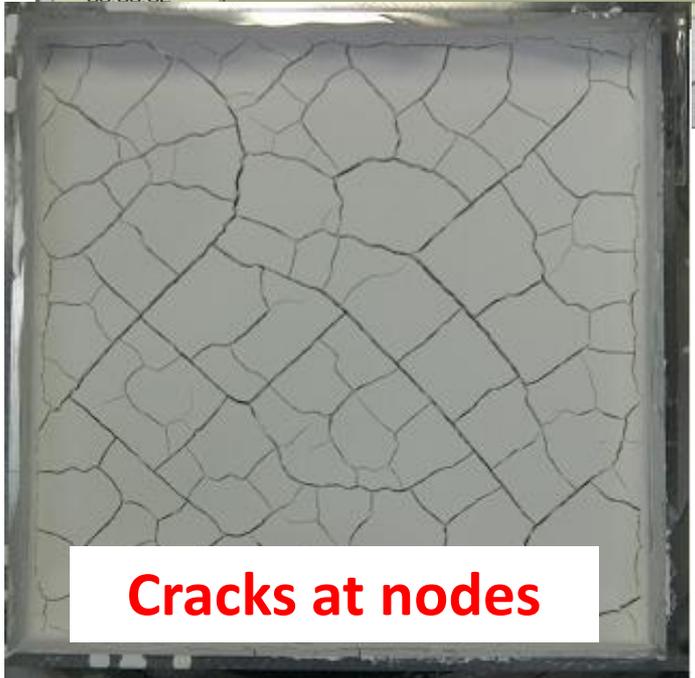
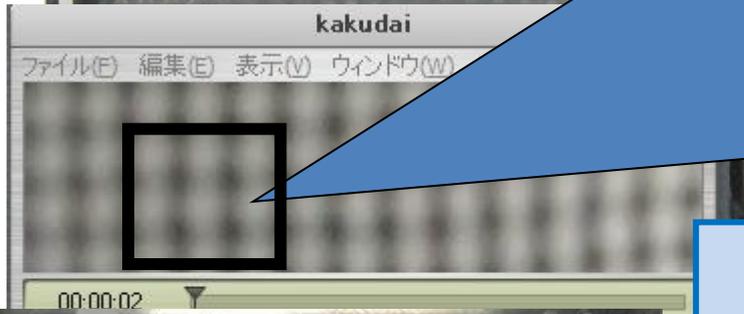
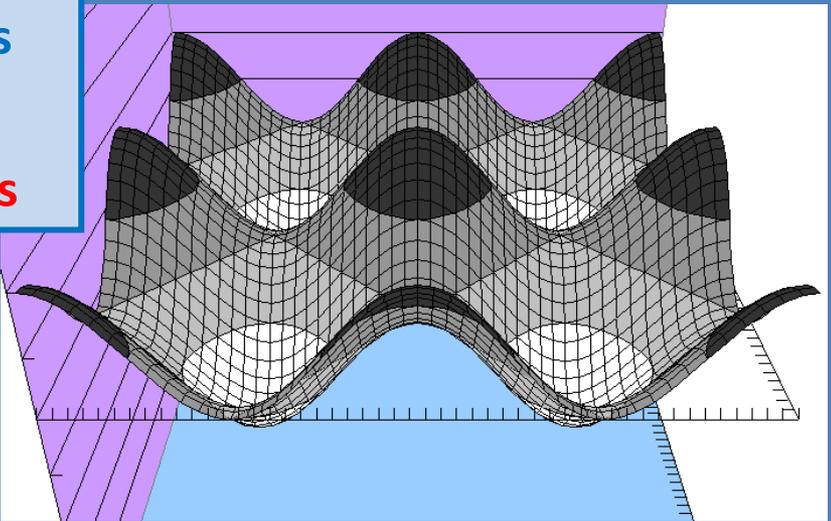
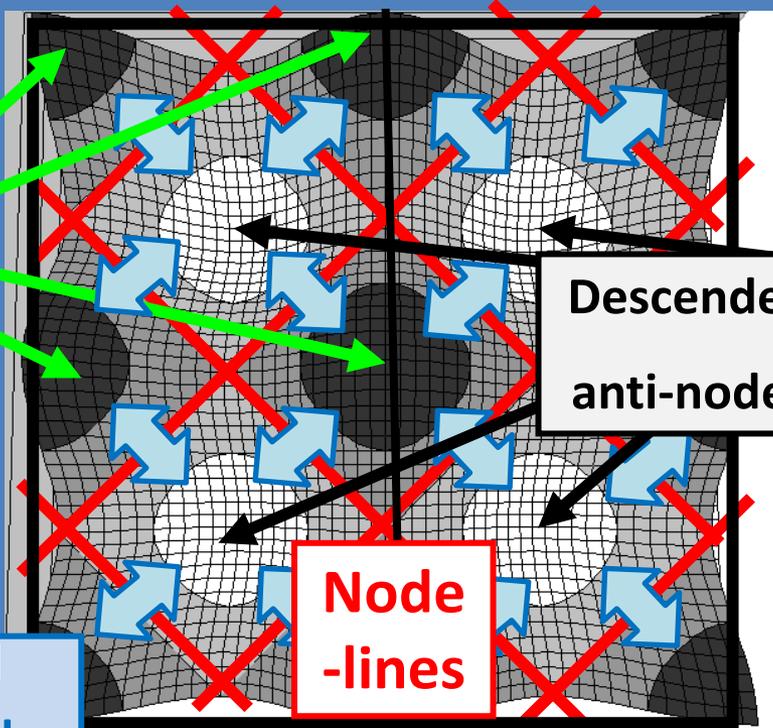
# Reason for tilt by 45°

Excited anti-nodes

Descended anti-nodes

Node-lines

Localized horizontal vibrations across node-lines

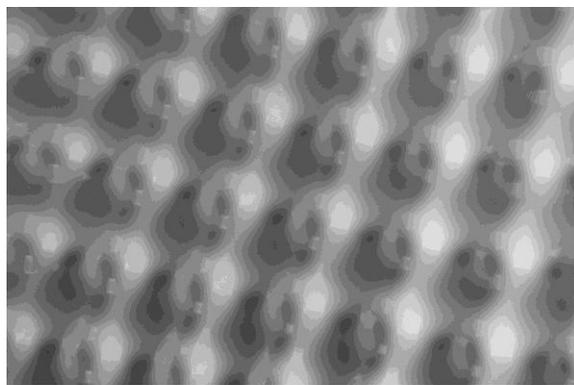


Cracks at nodes

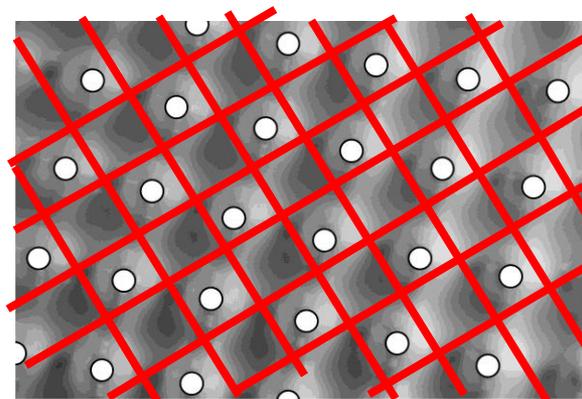
# Experimental check: superposition of successive Faraday waves

Faraday waves  
at one snapshot

$t$



(a)



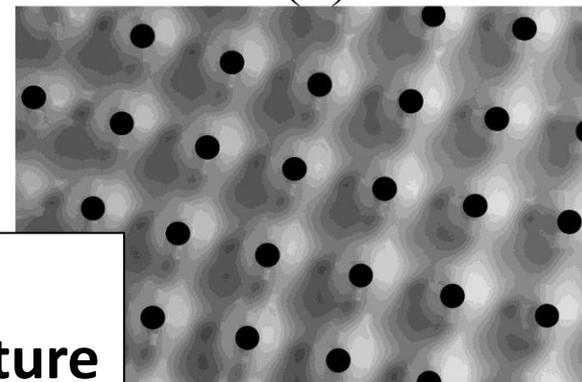
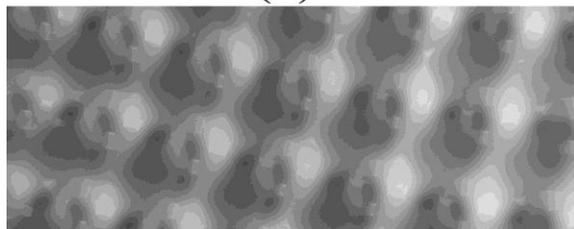
(b)

Snapshot

Tilt by  $45^\circ$  !

Faraday waves  
at next snapshot

$t + T_V$

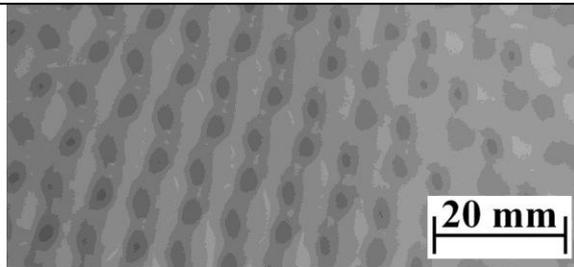


(d)

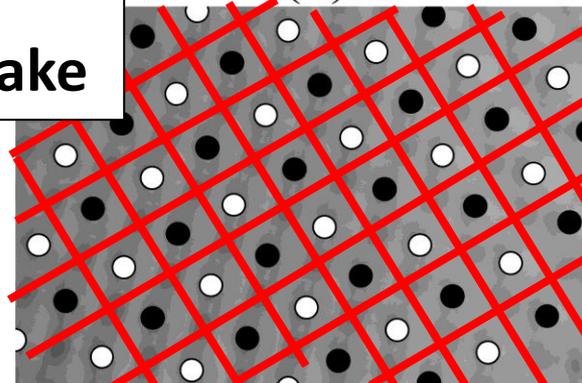
Top of excited  
anti-nodes  
are marked  
as circles

**Node lines** are regarded  
as a direction of lattice structure  
that the superposition of  
successive Faraday waves make

Superposition  
of successive  
Faraday waves  
 $t$  and  $t + T_V$



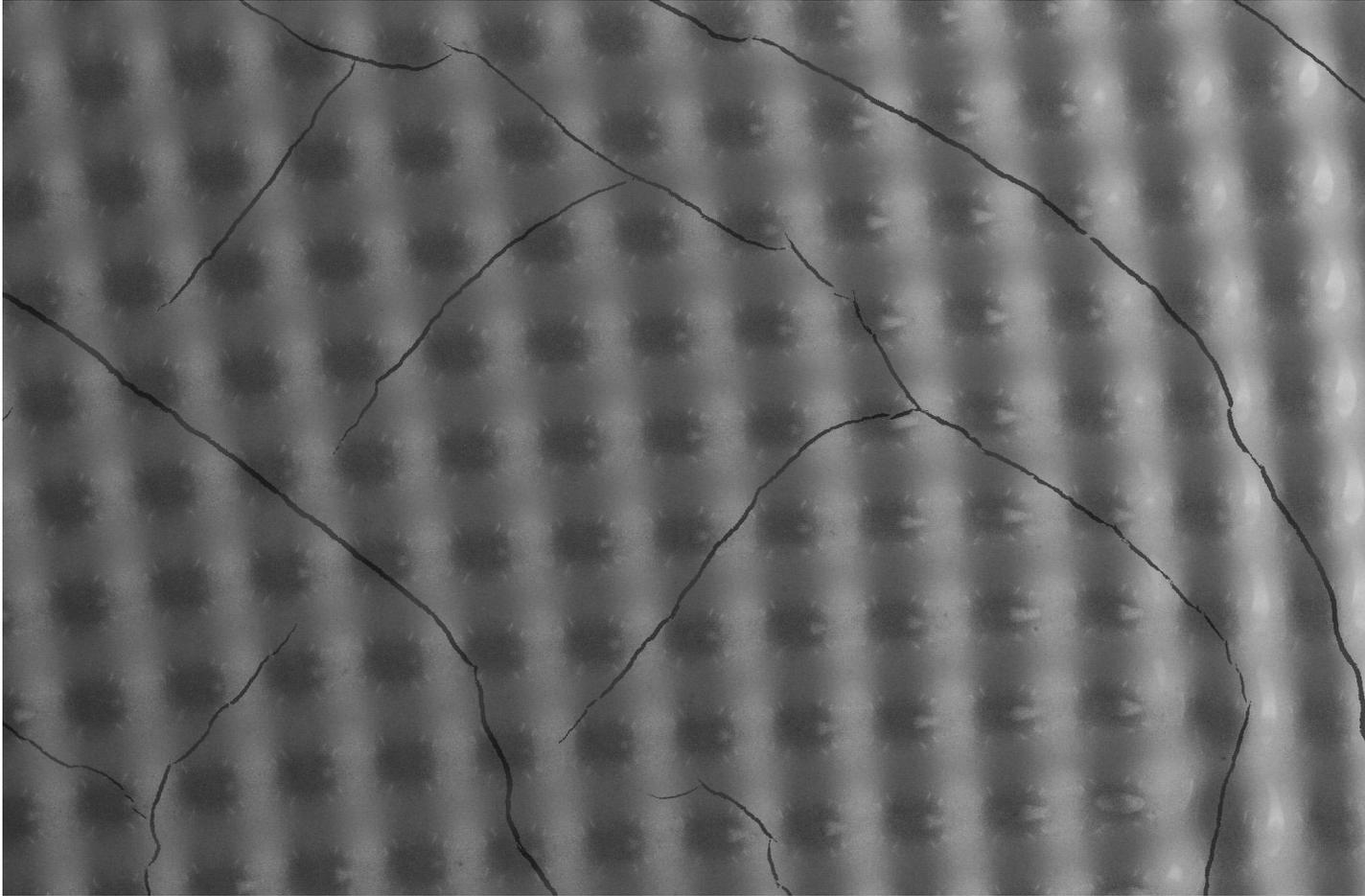
(e)



(f)

Node lines

Experimental check: Spatial correlation between  
Faraday waves at one snapshot and desiccation cracks



**Structures : Tilt by 45° !**

## Conclusion 2

- Using **vertical vibration**, we can control not only the direction but also the **position** of cracks.
- Cracks with **lattice** structure can be created !



**Square lattice**

H. Nakayama, Y. Matsuo, Ooshida Takeshi and A. Nakahara  
European Physical Journal E **36** (2013) 1.

# Further applications

## 1. What will happen **in future** (Technology)

→ Control microstructure of materials,  
anisotropy in conductivity and elastic moduli,  
and macroscopic crack patterns

## 2. What happened **before** (Geoscience)

→ Know past by memories and cracks in rocks

# Future plans

Flow ··· large deformation

- Definition and quantification of **Memories in pastes**  
including **Memory of flow**
- Theoretical model which explains **Memory of flow**
- Transition from **Memory of vibration** to **Memory of flow**

# Future plans 2

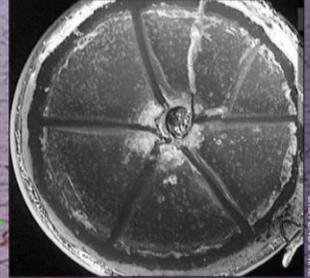
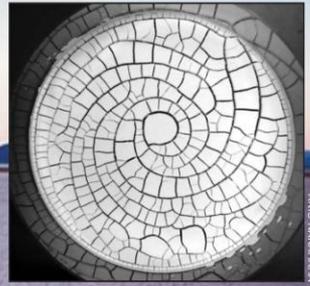
back scatter

Combine various external fields to control crack patterns

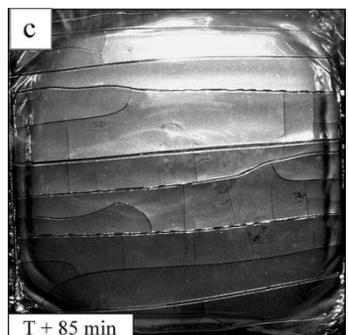
Mechanical method (vibration, flow)  
A. Nakahara & Y. Matsuo  
JPSJ (2005), PRE (2006)

Electrical method  
D. Mal et. al., Physica A (2007)

Magnetic method  
L. Pauchard et. al., PRE, (2008)  
A. T. Ngo et. el., Nano Letters (2008)



**Desiccation cracks**  
Crack patterns in layers of mud in dried river beds or in the glaze on old porcelain crockery are familiar to everyone. Desiccation crack patterns share some typical characteristics, which make them an active topic of research. Details such as the width distribution of cracks, the area they cover, the angles at which they meet, and the number of sides and the size distribution of the polygonal pieces depend on numerous factors, including the material, substrate, drying rate, humidity, and temperature. Realistic crack patterns can be generated by computer simulation using models that take into account the details of the physics and chemistry of the process. Numerous studies are under way to better understand the cracking process and, in some cases, how to control it, since crack patterns are sometimes intentionally produced as an artistic effect for decorative objects. Shown above are the spiral crack patterns produced when a clay sample is moved in a circular path before drying, and radial crack patterns formed when a synthetic clay is dried in a radially symmetric static electric field. More about desiccation crack patterns can be found in D. Mal et al., *J. Phys. Soc. Jpn.* **76**, 014801 (2007); D. Mal et al., *J. Appl. Clay Sci.* doi:10.1016/j.clay.2007.05.005 [in press]; A. Nakahara, Y. Matsuo, *Phys. Rev. E* **74**, 045102 (2006).



Desiccation cracks

xx September 2007 Physics Today

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