

## Role of steric repulsions on the precipitation kinetics and structure of calcium–silicate–hydrate gels

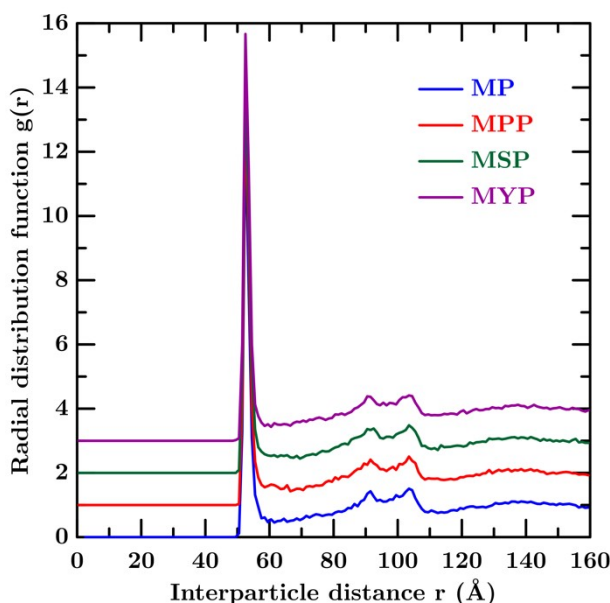
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### Supplementary Material



**Figure S1. Radial distribution function.** Radial distribution function at simulation step  $10^8$  for MP, MPP, MSP, and MYP potential with the interparticle distance.

Figure S1 shows the radial distribution function for calcium–silicate–hydrate (C–S–H) structure at  $10^8$  steps. The first peak, and the secondary peaks do not show the effect of interaction potential on the radial distribution function.

**Table S1.** The stiffness matrix of calcium–silicate–hydrate (C–S–H) structure at  $10^8$  steps

Potential	Stiffness matrix ( $C_{ij}$ )					
MP	26.300	15.000	15.100	-0.200	0.100	-0.200
	14.700	27.200	14.600	-0.500	0.000	-0.100
	14.800	14.700	26.400	-0.700	0.200	-0.400
	-0.300	-0.600	-0.800	5.200	0.100	-0.200
	0.100	-0.100	-0.100	0.000	5.700	-0.200

	-0.400	-0.100	-0.400	-0.200	-0.100	5.500
MPP	21.100	11.100	12.500	0.100	-0.100	-0.300
	11.500	21.200	12.500	-0.400	0.100	-0.200
	12.200	12.600	20.200	0.300	-0.300	0.200
	0.000	-0.700	0.400	4.400	-0.200	0.600
	-0.100	-0.100	-0.200	-0.200	4.700	0.200
	-0.300	-0.100	-0.100	0.600	0.300	4.900
MSP	22.200	13.200	11.400	0.700	0.400	-0.300
	13.100	22.700	12.300	0.200	-0.200	-0.100
	11.400	12.200	20.900	0.100	0.400	-0.400
	0.800	0.200	0.200	4.400	-0.100	0.500
	0.500	-0.300	0.400	-0.100	4.700	-0.100
	-0.200	-0.100	-0.600	0.500	0.000	4.400
MYP	20.200	9.600	10.700	-0.700	0.300	0.300
	9.600	21.200	10.800	-0.300	0.200	0.500
	10.700	10.800	22.200	-0.300	-0.300	0.200
	-0.800	-0.400	-0.200	5.100	0.300	0.100
	0.300	-0.300	-0.300	0.300	5.300	-0.200
	0.300	0.400	0.200	0.300	-0.300	5.100

The LAMMPS code used to compute the elastic modulus following the tapping and relaxation process for clarity and reproducibility.

### Initialization program

# NOTE: This script can be modified for different atomic structures,

# units, etc. See in.elastic for more info.

# variable to verify that results do not depend on it.

variable up equal 5.0e-4

# real units, elastic constants in GPa

units real

variable cfac equal 1.01325e-4

variable cunits string GPa

```

# Define minimization parameters
variable      etol equal 0
variable      ftol equal 0.000000001
variable      maxiter equal 100
variable      maxeval equal 1000
variable      dmax equal 1.0e-2

# atom positions
dimension     3
processors    * * *
boundary      p p p
atom_style    full # changed from charge

box           tilt large
read_data     input.dat # put the datafile name
change_box    all triclinic
#thermo_style custom v_dir v_eps pe pxx pyy pzz pxy pxz pyz lx ly lz xy xz yz

```

### **Tapping and relaxation process with the stiffness matrix program**

```

include init.mod
variable dir equal 0
variable eps equal 0.0
include potential.mod
variable      d loop 30
label         loop
print         "Loop step = $d"
fix           3 all box/relax tri 0.0
min_style     cg
minimize      ${etol} ${ftol} ${maxiter} ${maxeval}
unfix         3
fix           3 all box/relax tri 0.0
min_style     cg
minimize      ${etol} ${ftol} ${maxiter} ${maxeval}

```

unfix 3

variable a equal lx

variable b equal (\$a\*0.002)

variable c equal (-1.0\*\$a\*0.002)

change\_box all x delta 0.0 \$b remap units box

minimize 1.0e-8 1.0e-8 100000 100000

change\_box all x delta 0.0 \$c remap units box

minimize 1.0e-8 1.0e-8 100000 100000

change\_box all y delta 0.0 \$b remap units box

minimize 1.0e-8 1.0e-8 100000 100000

change\_box all y delta 0.0 \$c remap units box

minimize 1.0e-8 1.0e-8 100000 100000

change\_box all z delta 0.0 \$b remap units box

minimize 1.0e-8 1.0e-8 100000 100000

change\_box all z delta 0.0 \$c remap units box

minimize 1.0e-8 1.0e-8 100000 100000

change\_box all xy delta \$b remap units box

minimize 1.0e-8 1.0e-8 100000 100000

change\_box all xy delta \$c remap units box

minimize 1.0e-8 1.0e-8 100000 100000

change\_box all xz delta \$b remap units box

minimize 1.0e-8 1.0e-8 100000 100000

change\_box all xz delta \$c remap units box

minimize 1.0e-8 1.0e-8 100000 100000

change\_box all yz delta \$b remap units box

minimize 1.0e-8 1.0e-8 100000 100000

change\_box all yz delta \$c remap units box

minimize 1.0e-8 1.0e-8 100000 100000

fix 3 all box/relax tri 0.0

min\_style cg

minimize \${etol} \${ftol} \${maxiter} \${maxeval}

unfix 3

```

fix          3 all box/relax tri 0.0
min_style    cg
minimize    ${etol} ${ftol} ${maxiter} ${maxeval}
unfix       3

next        d
jump        in.elastic loop
variable tmp equal pxx
variable pxx0 equal ${tmp}
variable tmp equal pyy
variable pyy0 equal ${tmp}
variable tmp equal pzz
variable pzz0 equal ${tmp}
variable tmp equal pyz
variable pyz0 equal ${tmp}
variable tmp equal pxz
variable pxz0 equal ${tmp}
variable tmp equal pxy
variable pxy0 equal ${tmp}
variable tmp equal lx
variable lx0 equal ${tmp}
variable tmp equal ly
variable ly0 equal ${tmp}
variable tmp equal lz
variable lz0 equal ${tmp}
# These formulas define the derivatives w.r.t. strain components
# Constants uses $, variables use v_
variable d1 equal -(v_pxx1-${pxx0})/(v_delta/v_len0)*${cfac}
variable d2 equal -(v_pyy1-${pyy0})/(v_delta/v_len0)*${cfac}
variable d3 equal -(v_pzz1-${pzz0})/(v_delta/v_len0)*${cfac}
variable d4 equal -(v_pyz1-${pyz0})/(v_delta/v_len0)*${cfac}
variable d5 equal -(v_pxz1-${pxz0})/(v_delta/v_len0)*${cfac}
variable d6 equal -(v_pxy1-${pxy0})/(v_delta/v_len0)*${cfac}

```

```
# Write restart  
write_restart restart.equil
```

```
# uxx Perturbation
```

```
variable dir equal 1  
include displace.mod
```

```
# uyy Perturbation
```

```
variable dir equal 2  
include displace.mod
```

```
# uzz Perturbation
```

```
variable dir equal 3  
include displace.mod
```

```
# uyz Perturbation
```

```
variable dir equal 4  
include displace.mod
```

```
# uxz Perturbation
```

```
variable dir equal 5  
include displace.mod
```

```
# uxy Perturbation
```

```
variable dir equal 6  
include displace.mod
```

**Displacement program for stiffness matrix**

```

# NOTE: This script should not need to be
# modified. See in.elastic for more info.
#
# Find which reference length to use

if "${dir} == 1" then &
  "variable len0 equal ${lx0}"
if "${dir} == 2" then &
  "variable len0 equal ${ly0}"
if "${dir} == 3" then &
  "variable len0 equal ${lz0}"
if "${dir} == 4" then &
  "variable len0 equal ${lz0}"
if "${dir} == 5" then &
  "variable len0 equal ${lz0}"
if "${dir} == 6" then &
  "variable len0 equal ${ly0}"

# Reset box and simulation parameters

clear
box      tilt large
read_restart restart.equil
include potential.mod

# Negative deformation

variable delta equal -${up}*${len0}
variable eps equal 0.0

variable eps equal ${eps}-${up}
if "${dir} == 1" then &
  "change_box all x delta 0 ${delta} units box"
if "${dir} == 2" then &

```

```

"change_box all y delta 0 ${delta} units box"
if "${dir} == 3" then &
  "change_box all z delta 0 ${delta} units box"
if "${dir} == 4" then &
  "change_box all yz delta ${delta} units box"
if "${dir} == 5" then &
  "change_box all xz delta ${delta} units box"
if "${dir} == 6" then &
  "change_box all xy delta ${delta} units box"
minimize ${etol} ${ftol} ${maxiter} ${maxeval}

```

```

variable eps equal ${eps}-${up}
if "${dir} == 1" then &
  "change_box all x delta 0 ${delta} units box"
if "${dir} == 2" then &
  "change_box all y delta 0 ${delta} units box"
if "${dir} == 3" then &
  "change_box all z delta 0 ${delta} units box"
if "${dir} == 4" then &
  "change_box all yz delta ${delta} units box"
if "${dir} == 5" then &
  "change_box all xz delta ${delta} units box"
if "${dir} == 6" then &
  "change_box all xy delta ${delta} units box"
minimize ${etol} ${ftol} ${maxiter} ${maxeval}

```

```

variable eps equal ${eps}-${up}
if "${dir} == 1" then &
  "change_box all x delta 0 ${delta} units box"
if "${dir} == 2" then &
  "change_box all y delta 0 ${delta} units box"
if "${dir} == 3" then &
  "change_box all z delta 0 ${delta} units box"
if "${dir} == 4" then &

```



```
"change_box all yz delta ${delta} units box"
if "${dir} == 5" then &
  "change_box all xz delta ${delta} units box"
if "${dir} == 6" then &
  "change_box all xy delta ${delta} units box"
minimize ${etol} ${ftol} ${maxiter} ${maxeval}
```

```
variable eps equal ${eps}-${up}
if "${dir} == 1" then &
  "change_box all x delta 0 ${delta} units box"
if "${dir} == 2" then &
  "change_box all y delta 0 ${delta} units box"
if "${dir} == 3" then &
  "change_box all z delta 0 ${delta} units box"
if "${dir} == 4" then &
  "change_box all yz delta ${delta} units box"
if "${dir} == 5" then &
  "change_box all xz delta ${delta} units box"
if "${dir} == 6" then &
  "change_box all xy delta ${delta} units box"
minimize ${etol} ${ftol} ${maxiter} ${maxeval}
```

```
variable eps equal ${eps}-${up}
if "${dir} == 1" then &
  "change_box all x delta 0 ${delta} units box"
if "${dir} == 2" then &
  "change_box all y delta 0 ${delta} units box"
if "${dir} == 3" then &
  "change_box all z delta 0 ${delta} units box"
if "${dir} == 4" then &
  "change_box all yz delta ${delta} units box"
if "${dir} == 5" then &
  "change_box all xz delta ${delta} units box"
if "${dir} == 6" then &
```

```
"change_box all xy delta ${delta} units box"
minimize ${etol} ${ftol} ${maxiter} ${maxeval}
```

```
variable eps equal ${eps}-${up}
if "${dir} == 1" then &
  "change_box all x delta 0 ${delta} units box"
if "${dir} == 2" then &
  "change_box all y delta 0 ${delta} units box"
if "${dir} == 3" then &
  "change_box all z delta 0 ${delta} units box"
if "${dir} == 4" then &
  "change_box all yz delta ${delta} units box"
if "${dir} == 5" then &
  "change_box all xz delta ${delta} units box"
if "${dir} == 6" then &
  "change_box all xy delta ${delta} units box"
minimize ${etol} ${ftol} ${maxiter} ${maxeval}
```

```
variable eps equal ${eps}-${up}
if "${dir} == 1" then &
  "change_box all x delta 0 ${delta} units box"
if "${dir} == 2" then &
  "change_box all y delta 0 ${delta} units box"
if "${dir} == 3" then &
  "change_box all z delta 0 ${delta} units box"
if "${dir} == 4" then &
  "change_box all yz delta ${delta} units box"
if "${dir} == 5" then &
  "change_box all xz delta ${delta} units box"
if "${dir} == 6" then &
  "change_box all xy delta ${delta} units box"
minimize ${etol} ${ftol} ${maxiter} ${maxeval}
```

```
variable eps equal ${eps}-${up}
```

```

if "${dir} == 1" then &
  "change_box all x delta 0 ${delta} units box"
if "${dir} == 2" then &
  "change_box all y delta 0 ${delta} units box"
if "${dir} == 3" then &
  "change_box all z delta 0 ${delta} units box"
if "${dir} == 4" then &
  "change_box all yz delta ${delta} units box"
if "${dir} == 5" then &
  "change_box all xz delta ${delta} units box"
if "${dir} == 6" then &
  "change_box all xy delta ${delta} units box"
minimize ${etol} ${ftol} ${maxiter} ${maxeval}

```

```

variable eps equal ${eps}-${up}
if "${dir} == 1" then &
  "change_box all x delta 0 ${delta} units box"
if "${dir} == 2" then &
  "change_box all y delta 0 ${delta} units box"
if "${dir} == 3" then &
  "change_box all z delta 0 ${delta} units box"
if "${dir} == 4" then &
  "change_box all yz delta ${delta} units box"
if "${dir} == 5" then &
  "change_box all xz delta ${delta} units box"
if "${dir} == 6" then &
  "change_box all xy delta ${delta} units box"
minimize ${etol} ${ftol} ${maxiter} ${maxeval}

```

```

variable eps equal ${eps}-${up}
if "${dir} == 1" then &
  "change_box all x delta 0 ${delta} units box"
if "${dir} == 2" then &
  "change_box all y delta 0 ${delta} units box"

```

```
if "${dir} == 3" then &
  "change_box all z delta 0 ${delta} units box"
if "${dir} == 4" then &
  "change_box all yz delta ${delta} units box"
if "${dir} == 5" then &
  "change_box all xz delta ${delta} units box"
if "${dir} == 6" then &
  "change_box all xy delta ${delta} units box"
minimize ${etol} ${ftol} ${maxiter} ${maxeval}
```

```
# Reset box and simulation parameters
```

```
clear
```

```
box      tilt large
```

```
read_restart restart.equil
```

```
include potential.mod
```

```
# Positive deformation
```

```
variable eps equal 0.0
```

```
variable delta equal ${up}*${len0}
```

```
variable eps equal ${eps}+${up}
```

```
if "${dir} == 1" then &
```

```
  "change_box all x delta 0 ${delta} units box"
```

```
if "${dir} == 2" then &
```

```
  "change_box all y delta 0 ${delta} units box"
```

```
if "${dir} == 3" then &
```

```
  "change_box all z delta 0 ${delta} units box"
```

```
if "${dir} == 4" then &
```

```
  "change_box all yz delta ${delta} units box"
```

```
if "${dir} == 5" then &
```

```
  "change_box all xz delta ${delta} units box"
```

```
if "${dir} == 6" then &
```

```
"change_box all xy delta ${delta} units box"  
minimize ${etol} ${ftol} ${maxiter} ${maxeval}
```

```
variable eps equal ${eps}+${up}  
if "${dir} == 1" then &  
  "change_box all x delta 0 ${delta} units box"  
if "${dir} == 2" then &  
  "change_box all y delta 0 ${delta} units box"  
if "${dir} == 3" then &  
  "change_box all z delta 0 ${delta} units box"  
if "${dir} == 4" then &  
  "change_box all yz delta ${delta} units box"  
if "${dir} == 5" then &  
  "change_box all xz delta ${delta} units box"  
if "${dir} == 6" then &  
  "change_box all xy delta ${delta} units box"  
minimize ${etol} ${ftol} ${maxiter} ${maxeval}
```

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variable eps equal ${eps}+${up}  
if "${dir} == 1" then &  
  "change_box all x delta 0 ${delta} units box"  
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  "change_box all y delta 0 ${delta} units box"  
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  "change_box all yz delta ${delta} units box"  
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minimize ${etol} ${ftol} ${maxiter} ${maxeval}
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variable eps equal ${eps}+${up}
```

```

if "${dir} == 1" then &
  "change_box all x delta 0 ${delta} units box"
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if "${dir} == 6" then &
  "change_box all xy delta ${delta} units box"
minimize ${etol} ${ftol} ${maxiter} ${maxeval}

```

```

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if "${dir} == 1" then &
  "change_box all x delta 0 ${delta} units box"
if "${dir} == 2" then &
  "change_box all y delta 0 ${delta} units box"

```

```
if "${dir} == 3" then &
  "change_box all z delta 0 ${delta} units box"
if "${dir} == 4" then &
  "change_box all yz delta ${delta} units box"
if "${dir} == 5" then &
  "change_box all xz delta ${delta} units box"
if "${dir} == 6" then &
  "change_box all xy delta ${delta} units box"
minimize ${etol} ${ftol} ${maxiter} ${maxeval}
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variable eps equal ${eps}+${up}
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if "${dir} == 5" then &
  "change_box all xz delta ${delta} units box"
if "${dir} == 6" then &
  "change_box all xy delta ${delta} units box"
minimize ${etol} ${ftol} ${maxiter} ${maxeval}
```

```
variable eps equal ${eps}+${up}
if "${dir} == 1" then &
  "change_box all x delta 0 ${delta} units box"
if "${dir} == 2" then &
  "change_box all y delta 0 ${delta} units box"
if "${dir} == 3" then &
  "change_box all z delta 0 ${delta} units box"
if "${dir} == 4" then &
  "change_box all yz delta ${delta} units box"
```

```
if "${dir} == 5" then &
  "change_box all xz delta ${delta} units box"
if "${dir} == 6" then &
  "change_box all xy delta ${delta} units box"
minimize ${etol} ${ftol} ${maxiter} ${maxeval}
```

```
variable eps equal ${eps}+${up}
if "${dir} == 1" then &
  "change_box all x delta 0 ${delta} units box"
if "${dir} == 2" then &
  "change_box all y delta 0 ${delta} units box"
if "${dir} == 3" then &
  "change_box all z delta 0 ${delta} units box"
if "${dir} == 4" then &
  "change_box all yz delta ${delta} units box"
if "${dir} == 5" then &
  "change_box all xz delta ${delta} units box"
if "${dir} == 6" then &
  "change_box all xy delta ${delta} units box"
minimize ${etol} ${ftol} ${maxiter} ${maxeval}
```

```
variable eps equal ${eps}+${up}
if "${dir} == 1" then &
  "change_box all x delta 0 ${delta} units box"
if "${dir} == 2" then &
  "change_box all y delta 0 ${delta} units box"
if "${dir} == 3" then &
  "change_box all z delta 0 ${delta} units box"
if "${dir} == 4" then &
  "change_box all yz delta ${delta} units box"
if "${dir} == 5" then &
  "change_box all xz delta ${delta} units box"
if "${dir} == 6" then &
  "change_box all xy delta ${delta} units box"
```



```
minimize ${etol} ${ftol} ${maxiter} ${maxeval}
```

```
# Delete dir to make sure it is not reused
```

```
variable dir delete
```