## **Supplementary Information for**

# Transitions between Nanomechanical and Continuum Mechanical Contacts: New Insights from Liquid Structure

<sup>1,2</sup>Shujian Chen<sup>\*§</sup>, <sup>3</sup>Weiqiang Chen<sup>§</sup>, <sup>4</sup>Yubing Ouyang, <sup>2</sup>Stephan Matthai<sup>\*</sup>, <sup>2</sup>Lihai Zhang

<sup>1</sup>School of Civil Engineering, The University of Queensland, Brisbane, Queensland 4072, Australia

<sup>2</sup>Department of Infrastructure Engineering, The University of Melbourne, Parkville 3010, Australia

<sup>3</sup>State Key Laboratory of Geomechanics and Deep Underground Engineering, China University of Mining and Technology, Xuzhou 221116, China

<sup>4</sup>Department of Civil Engineering, Monash University, Clayton 3168, Australia

\*E-mail: shujian.chen@uq.edu.au (S.J. Chen); stephan.matthai@unimelb.edu.au. (S. Matthai).

This material includes supplementary notes, figures and their captions.



**Figure S1.** (a) Typical setup for the molecular model which simulated the contact between two surfaces. It should be noted that the asperities extend periodically in the z-direction. The region within the red box is our region of interest and an extra free water layer is added around it. (b) An enlarged view of the contact point. (c, d) Color code of different types of atoms and molecules including water (c) and silica (d).

**Table S1.** Model arguments, fitting result for equivalent starting point  $\delta_0$  of Hertzian contact and respective critical contact force  $F_c$  for each case

Case	1	2	3	4	5	6	7	8	9
$R_1$ (nm)	3.0	3.0	3.0	3.0	4.5	4.5	4.5	4.5	4.5
$R_2$ (nm)	3.0	3.0	3.0	3.0	3.0	3.0	4.5	4.5	4.5
$\mathbf{d}_1$	001	010	210	210	010	210	001	010	210
$\mathbf{d}_2$	001	010	010	210	010	210	001	010	010
$\delta_{0}\left(\mathrm{nm} ight)$	0.549	0.326	0.522	0.690	0.411	0.613	0.721	0.435	0.613
$R^2$	0.923	0.988	0.895	0.724	0.982	0.719	0.921	0.989	0.920
$F_c$ (nN)	99.3	123.2	77.8	89.9	147.9	107.9	148.9	184.9	116.7
Case	10	11	12	13	14	15	16	17	18
$R_1$ (nm)	4.5	6.0	6.0	6.0	6.0	6.0	6.0	6.0	6.0
$R_2$ (nm)	4.5	3.0	3.0	4.5	4.5	6.0	6.0	6.0	6.0
$\mathbf{d}_1$	210	010	210	010	210	001	010	210	210
$\mathbf{d}_2$	210	010	210	010	210	001	010	010	210
$\delta_{0}\left(\mathrm{nm} ight)$	0.658	0.473	0.633	0.489	0.736	0.525	0.382	0.499	0.342
$R^2$	0.769	0.967	0.866	0.982	0.881	0.998	0.998	0.996	0.998
$F_c$ (nN)	134.8	164.3	119.9	211.3	154.1	198.6	246.5	155.6	179.8

**Supplementary Note 1. The Energy Functional Form for ClayFF.** The energy formula for ClayFF has contributions from the Coulombic interactions, the short-range interactions (also referred to as the van der Waals (vdW) term) and the bonded interactions including the bond stretch and angle bend energy terms given as eq S1<sup>1</sup>:

$$E_{total} = E_{Coul} + E_{vdw} + E_{bond \ stretch} + E_{angle \ bend}.$$
(S1)

The Coulombic energy is shown in eq S2:

$$E_{Coul} = \frac{e^2}{4\pi\epsilon_0} \sum_{i \neq j} \frac{q_i q_j}{r_{ij}}$$
(S2)

where the Coulombic energy is inversely proportional to the distance of separation  $r_{ij}$ ,  $q_i$  and  $q_j$ are the partial charges while e is the charge of the electron and  $\epsilon_0$  is defined as the dielectric permittivity of vacuum which is equal to  $8.85419 \times 10^{-12}$  F/m

The vdW energy term can be expressed by the Lennard-Jones (12-6) function (eq S3):

$$E_{vdw} = \sum_{i \neq j} D_{0,ij} \left[ \left( \frac{R_{0,ij}}{r_{ij}} \right)^{12} - 2 \left( \frac{R_{0,ij}}{r_{ij}} \right)^6 \right]$$
(S3)

where  $D_{0,ij}$  and  $R_{0,ij}$  are the empirical energy and distance parameters, respectively.

Supplementary Note 2. Extended Examination on the Efficiency of Pressure-Relief Mechanism of Free Water Layer in NVT Ensemble. Figure S2 compares the potential energy of interfacial water molecules and that in the free water layer during the contact process and it is found that the free water layer serves as a buffer zone and the high potential energy of squeezed-out water molecules would dissipate in this zone. Because the water pressure is the gradient of water potential energy and  $E_{ppm}$  in the free water layer fluctuates at a minimum range (standard deviation is 1.34% of average value) during the contact process, the pressure of the free water layer should be constant during the contact process. The discussion in this note indicates that by setting a free water layer, NVT can be used in the MD simulation of the contact process because

pressure build up caused by contact would be efficiently dissipated by the surrounding free water layer. Both NVT and NPT can be adopted in this wet quartz contact simulation.



**Figure S2.** The potential energy of interfacial water and free water layer during the contact process for the case of  $R_1 = R_2 = \infty$  nm,  $\mathbf{d} = (010)$ .

Supplementary Note 3. Change of Energy Components During the Compression Process. The energy components are examined to validate the simulation setup. The energies of the angle bend  $(E_{angle})$  and bond stretch  $(E_b)$  reflect the change in the interaction between water molecules and the surface –OH groups because the interaction between Si and O are simulated via nonbond forces. Figure S3b show that increasing the radii of the asperities results in a proportional increase of the bond energy. The results show that the interfacial water nanofilm between asperities demonstrates some "viscosity effect" <sup>2-3</sup>, namely, the water needs a certain period to relax and reach equilibrium. This effect reduces as two asperities moved closer because the amount of interfacial water molecules reduces. When the two asperities started touching ( $\delta = 0$ nm), this effect disappears (as shown in Figure S3). Based on the examination here, it is found that the relaxation stage used during the intervals is essential to allow viscous movement of the water molecules. Figure S3 shows that the relaxation time of 40 ps is enough to minimize the energy build up due to the viscosity of the water. To extract locations and energy of water molecules after these viscous movements, we extended another 40 ps for data collection after the 40 ps relaxation for the case with radii being 3.0, 4.5, 6.0,  $\infty$  nm and the lattice direction being (010). For the other scenarios shown in Figure 5 and Table S1 where deformation of quartz is of interest, we only use a relaxation time 40 ps for collected data on the compression force– deformation relationship only.



**Figure S3.** Energy components during compression. (a–c) The energy of angle bend ( $E_{angle}$ ), bond stretch ( $E_b$ ) and van der Waals forces ( $E_v$ ) of cases with different radii. The data points in all the graphs indicate the results after a relaxation of 40 ps using NVT while the lines represent the smoothed (by Fourier filtering) data for the whole simulation process.

#### Supplementary Note 4. Procedures for Obtaining the Spatial Distribution Function (SDF)

of Interfacial Water. Here, we explain how the SDF is obtained to allow the examination of the nanoscale arranging and ordering effect of interfacial water molecules. As illustrated in Figure S4a, to produce an SDF of interfacial water, we first take a sample of a single water molecule as the central molecule, regard it as a solute in a solvent of water. The SDF, therefore, describes the likelihood of other water molecules occurring in the three-dimensional space around this molecule. Then, we overlap several central water molecules (note that overlapping means that not only the mass centers but also the two hydrogens of different central water molecules should be translated and rotated to the same spatial locations), and then their surrounding water molecules (Figure S4a). After that, the probability distribution of surrounding water molecules can be obtained based on the overlapped surrounding water molecules from different central water molecules. Figure S4b shows the slices in three different planes from this spatial probability distribution. Figure S4c shows the final model of 3D water solvation shell (or SDF) which is

produced by rendering the isosurface. The spatial positions of interfacial water molecules are sampled every 0.2 ps in a relaxation stage of 40 ps. Namely, 200 snapshots are used to produce the model of water solvation shell (or SDF). As shown in Figure S4c, the produced water solvation shell (or SDF) clearly shows the probability density of surrounding water molecules and preferred location of water molecules. Compared with other methods like Radial Distribution Function (RDF) <sup>4</sup>, the SDF proposed here can provide more comprehensive visualization clearly showing the mutual location relations between surrounding molecules and hydrogen and oxygen of the central water molecules.



Figure S4. Evaluation and Visualization of the Water Solvation Shell (or SDF). (a) Obtaining the probability distribution of the water occurrence in the solvation shell by choosing different

water molecules as the center, and then translating the coordinates of all water molecules in the gap to the origin of the coordinate system. After translation, the coordinates of all the water molecules are rotated to align the hydrogen atoms. (b) Slices from the 3-D histogram obtained by overlapping the coordinates of water molecules by sampling 2000 water molecules as the centers. (c) 3-D visualization of the distribution of water molecules by rendering a number of isosurfaces.

#### Supplementary Note 5. Extended Data for the Projection of Water Occupancy (PWO). This

note explains the clustering of water coordinate data in the x–y-plane and the link between the clustering and layer-like structure formed by interfacial water molecules. As shown in Figure S5, in support of Figure 2c–h in the main body of this paper, the spatial positions of interfacial water molecules are sampled in the same intervals as SDF. To reveal the layer-like arrangement of liquid structure in the interfacial water nanofilm <sup>5</sup>, we projected the coordinates of water molecules (by the center of mass) to the x–y-plane. Projected coordinates from the 200 frames were overlapped to capture the thermal movement of the water molecules. Figure S5 shows the existence of areas with highly clustered data points. Note that the occurrence of water molecules is presented as light blue dots and it is evident that in some locations, dots would cluster. This can be the result of thermal vibration of water molecules around the energetically preferred locations in the space. To eliminate the effect of thermal vibration and clearly identify these locations, further processes are needed, which are demonstrated in Supplementary Note 6.



**Figure S5**. The Occurrence of Water Molecules between Two Surfaces. The data points (light blue dots) are the mass center locations of water molecules that are recorded over a 40 ps period. Typical results are demonstrated here for flat surface (a–d) and a curved surface with radius equal to 3 nm (e, f) during the contact process.

Supplementary Note 6. 2-D Histogram of Water Coordinate Projection and Gaussian Denoising. As shown in Figure S6, the information of water coordinate data in Figure S5 is converted into histograms using a bin width of 0.025 nm. One can notice the high level of noise in these histograms ( $\sigma = 0$  nm). To identify the location where the water molecule preferred to reside locally, we applied Gaussian denoising to smooth the histogram. Figure S6a–c shows the 2-D histograms for two flat surfaces with different  $\delta$  and  $\sigma$ , while Figure S6e-f indicates an alternative case of asperities with a curved surface. As demonstrated in Figure S6, the denoising effects with different filter sizes are examined and compared, we reckon that with a filter size of 0.5 nm, the 2-D histograms clearly show local maxima points while still preserving the location of the data clusters (Figure S5) in the histogram. The local maxima were then identified and presented in Figure 2c–h in the main text.



Figure S6. 2D Histograms of the Water Coordinate Projection and the Effect of Gaussian Denoising. Histogram of projected water coordinate data to the x-y-plane for the two surfaces without (a-c) or with (e) surface curvature. The noise in the histogram is removed using Gaussian filters with filter size  $\sigma = 0$ , 0.125, 0.25 and 0.5 nm.

Supplementary Note 7. Extended Examination on the Preferred Locations of Water Molecules between Curved Surfaces. The results presented in Figure 2c-h illustrate the significant change in the preferred locations of water molecules during contact. As shown in Figure 2c-f, the collapse of layers of water is clearly visible during the contact of flat surfaces. In addition, Figure 2g-h clearly shows the change of spatial arrangement of preferred locations of water molecules between curved surfaces, here we present a more detailed explanation and analysis of this arrangement. Figure S7 illustrates the distribution of points, which represents the preferred location of water molecules, and the network connecting these points. The curved geometry introduces a wider range of preferred packing modes in addition to the layered packing for flat surface (Figure 2c-f). These packing modes lead to an extremely complex nanomechanical system. As demonstrated in Figure S7, there are local regions in the water nanofilm with different levels of ordering effect, which is affected by the vertical gap size but

also varies horizontally. For example, the high-energy regions shift horizontally from Figure S7g–h. As the vertical gap between the two surfaces reduces from g to h, these high-energy regions shift to regions with bigger gaps. In other words, the curved surfaces can be, to some extent, considered as a combination of flat surfaces with different gap size. As the two surfaces compress, it can be seen the high-energy and high-confinement areas become more prominent from the profile of the moving mean  $L_{ij}$  indicating the transition into a nanomechanical stage. Another particular case is for Figure S7e, where a very good confinement effect occurs to wide regions in the gap, allowing optimum packing and minimum energy for water molecules.

The discussion in this note illustrates the complexity of the problem to describe the nanomechanical contact for surfaces with complex geometry. Development of an accurate formulation for such a nanomechanical model is beyond the scope of this paper but important for follow-up studies. To lay the ground for such future work, herein we illustrate the correlation between moving  $L_{ij}$  and the mean potential energy of interfacial water molecules. This correlation can be established by the following equation (eq S4).

$$E_{l} = \frac{1}{m} \sum_{j=1}^{j=m} \frac{1}{n_{j}} \sum_{i=1}^{i=n_{j}} (L_{ij} - 0.275 \ nm)$$
(S4)

where *m* is the number of bins in the x-direction and  $n_j$  is the number of  $L_{ij}$  in the *j* bin. In our calculation, we divide the spread of the gap between the two surfaces in Figure S7 into bins with size = 0.2 nm because in each bin the space in the y-direction allowed for atoms to pack as can be seen as constant. Therefore, the average  $L_{ij}$  within this bin will represent the order of packing for that specific gap size of the bin. When the mean of  $L_{ij}$  in a bin is closer to 0.275 nm, the packing is, therefore, more ideal with low energy. To obtain an overall indicator of packing order  $E_l$  in the entire gap we calculate the mean of all the bins.  $E_l$  is compared with the mean potential energy of water molecules  $E_{ppm}$  in the main article (Figure 3c). The correlation here demonstrates the importance of surface geometry and the packing space of water.



**Figure S7.** Analysis of the arrangement of preferred location (projected to x–y-plane) of water molecules. (a–h) Delaunay triangulation based on the preferred locations of water as the discrete points. To avoid the influence of very closely located points, which can be a result of local vibration of water molecules near the preferred locations, we combine several closely located points with mutual distances less than 0.138 nm (50% of the approximate size of a water molecule) into one (by using the mean coordinates of these points). The area of each triangle,  $A_t$ , is color coded. Larger  $A_t$  indicates lower order of water in that region and vice versa. The distance between each pair of points,  $L_{ij}$ , is plotted as scatter points while the moving mean of  $L_{ij}$  is indicated using solid black lines. The distribution of  $L_{ij}$  is compared with the ideal size of water, 0.275 nm (indicated by the dash blue line), to reveal high-energy regions and high-confinement regions. In the high-confinement region, the  $L_{ij}$  is close to the ideal size of water of 0.275 nm while in the high-energy regions,  $L_{ij}$  is significantly higher than 0.275 nm.

**Hydrophilic and Hydrophobic Surfaces.** The nanomechanical properties of interfacial water between quartz surfaces are investigated by comparing the behavior of interfacial water between quartz surfaces with that between ideal surfaces (namely, ideal hydrophilic and hydrophobic surfaces). In the context of hydrophilic surfaces, interfacial water molecules can interact with the surfaces and be attracted by the surfaces. Therefore, interfacial water molecules between hydrophilic surfaces occupy a "thicker" zone (Figure S8a–d) than that between hydrophobic

Supplementary Note 8. The Compression of Interfacial Water Nanofilm between Ideal

surfaces, which is "thinner" (Figure S8e–h). In addition, Figure S8a–h shows that the interfacial water molecules between hydrophobic surfaces are orderly aligned as 2–3 layers while are more disordered between hydrophilic surfaces.



**Figure S8.** Simulated compression of interfacial water nanofilm between ideal hydrophilic and hydrophobic surfaces. (a–d) The compression process of the ideal hydrophilic surfaces. (e–h) The compression process of the ideal hydrophobic surfaces. (i) The schematic diagram of the compression process of the 3-D model by compressing the simulation box in the z-direction.

Supplementary Note 9. Changes of 3-D SDF for Interfacial Water between Quartz Surfaces.

This note represents the changes of SDF for interfacial water between quartz surfaces in a threedimensional sense (3-D SDF), which is supplementary visualization and support for the 2-D SDF used in Figure 2b. As is shown in Figure S9, the changes mainly happened at the slice plane as is shown in Figure 2b. Therefore, we visualize the change of SDF in a two-dimensional sense as in the main text for simplicity. As is introduced in the main text, the clockwise shifts of position I at peaks of potential energy fluctuation and counterclockwise shifts at valleys of potential energy fluctuation can be observed from Figure S9a–d.



**Figure S9.** (a–d) The top view of 3-D SDF. (e–h) The front view of 3-D SDF for interfacial water during the contact process of two quartz surfaces.

**Supplementary Note 10. The Hydrophobicity Shift of Quartz Surface.** By comparing Figure S9 with Figure S10 and Figure S11, we analyzed the shift of hydrophobicity at the quartz interface due to the compression. To analyze the hydrophobicity shift of quartz surface, the 3-D SDF for ideal hydrophilic and hydrophobic surfaces were produced as Figure S10 and Figure S11, respectively. It is obvious that the quartz surface tends to be hydrophilic when the gap is 0.7, 0.9 and 1.0 nm, and when the gap decreases to 0.55 nm, the quartz surface changed to be hydrophobic. The discussion here supports Figure 4 in the main text. For clarity and simplicity, the 2-D SDF sliced from the top view plane is shown in the main text to illustrate this shift.



**Figure S10.** (a–d) The top view of 3-D SDF. (e–h) The front view of 3-D SDF for interfacial water during the contact process of two hydrophilic surfaces.



**Figure S11.** (a–d) The top view of the 3-D SDF. (e–h) The front view of the 3-D SDF for interfacial water during the contact process of two hydrophobic surfaces.

### Supplementary Note 11. Details of In-Plane Uniaxial Compression Tests on α-Quartz Blocks along Different Crystal Orientations. The uniaxial compression process was recorded

to investigate the continuous deformation characteristics of intact quartz crystals. As is shown in Figure S12, in-plane uniaxial compression tests were conducted to obtain the basic mechanical properties of  $\alpha$ -quartz, that is, Young's modulus and the Poisson ratio of  $\alpha$ -quartz along the crystal orientations of **d** = (001), (010) and (210). The top and bottom boundaries of rectangular blocks are taken as the loading end with a loading rate of  $6.0 \times 10^{-5}$  nm/fs. The atom's velocity in the top and bottom boundaries along the loading direction (y-direction)  $v_y$  is fixed at  $6.0 \times 10^{-5}$  nm/fs and 0 nm/fs, respectively, but the velocity in the x-direction  $v_x$  is not fixed. To get the lateral strain of rectangular blocks precisely, the width of rectangular blocks during the compression process was decided by the average positions of capped hydrogens in the left and right boundaries. Based on the deformation characteristics as shown in Figure S12, it is found that the atomic structures of intact quartz crystal after deformation are without evident strain concentrations.



**Figure S12.** (a, b) and (c, d) show the rectangular blocks of  $\alpha$ -quartz loaded along the lattice direction of (010) and (210), respectively.

Supplementary Note 12. Approximate Elastic Characteristics of  $\alpha$ -Quartz. Young's modulus E and the Poisson ratio  $\mu$  of  $\alpha$ -quartz were calculated to validate the use of elastic characteristics of a continuous model to model  $\alpha$ -quartz deformation. Figure S13 and Figure S14 show the simulation scatter and regression lines of uniaxial compression tests as introduced by Supplementary Note 11. To investigate the elastic characteristics of quartz crystal in this loading condition, a straight line through the origin is adopted to fit the simulation data, that is, axial

stress  $\sigma$  versus axial strain  $\varepsilon$  and lateral strain  $\varepsilon_h$  versus axial strain  $\varepsilon$ . It is found that the regression result is susceptible to the range of axial strain  $\varepsilon$  selected (Figure S13 and Figure S14) and the goodness of fit,  $R^2$  is barely satisfactory in a small axial strain range (0–0.01 and 0–0.02). However, when the axial strain range is fixed to be 0-0.05, the results show that the linear elastic model provides a better fit to the simulation scatter. The goodness of fit,  $R^2$ , to the simulation data are all above 90% for the 0–0.05 axial strain range. The calculated Young's modulus E and Poisson ratio  $\mu$  are all greater than those in the resonance spectrum and other experiments <sup>6-7</sup>, being E = 79.41 GPa,  $\mu = 102.8$  GPa, respectively. The higher elastic modulus from the MD simulation is partially induced by the in-plane confinement<sup>8</sup>. In addition, we also noticed the hardening effects <sup>9</sup> to the quadratic form of the forcefield functional used in ClayFF<sup>1</sup>. Our results here indicate that the ClayFF only guarantees the elasticity in a short range with moderately high E and  $\mu$ . Because the purpose of this study is to verify the validity of continuum mechanical presentation for the contact based on atomic models, focus is not given to the optimization of ClayFF for a more accurate representation of E and  $\mu$ , which can be done more properly in a separate study. Here, we adopt the value of E and  $\mu$  between strain = 2% and strain = 5% in a corresponding continuum mechanical model to verify the transition from nanomechanical to continuum contact behavior.



**Figure S13.** The linear regressions of Young's modulus for in-plane uniaxial compression tests along the three lattice directions ( $\mathbf{d} = 001, 010, 210$ ) with the strain of 1% (a, d, g), 2% (b, e, h) and 5% (c, f, i).



**Figure S14.** The linear regressions of Poisson's ratio for in-plane uniaxial compression tests along the three lattice directions ( $\mathbf{d} = 001, 010, 210$ ) with the strain of 1% (a, d, g), 2% (b, e, h) and 5%(c, f, i).

Supplementary Note 13. The Mesh Convergence in the FEM Modeling. As illustrated in Figure S15, two semicylinders are simulated to contact and compress with mesh size  $\sim 0.02R$  and  $\sim 0.01R$  (Figure S15a, b), both figures indicate that the stress is highly concentrated within the compression area where two semicylinders contact. Moreover, Figure S15c shows that the mesh size is converged for a force–displacement relationship. The size of  $\sim 0.01R$  is adopted in the estimation of boundary effects.



**Figure S15.** Mesh convergences for the finite element simulations. (a) and (b) Deformation and von Mises stress distribution in the simulation of semicylinder contact with the mesh size  $\approx$  0.02R (a) and 0.01R (b). (c) Convergence of the force–displacement relationship.

Supplementary Note 14. Consideration of the Boundary Effect in the Hertzian Contact Model. As addressed earlier, a tip-like boundary condition (Figure S1) is used in the simulation here to optimize computational cost in MD. This boundary condition should be considered in the corresponding Hertzian contact model. To understand further the boundary effect, Figure S16 compares the stress distribution of two cases using semicylindrical (a) and tip-like boundary (b). The stress distribution shown here follows a similar pattern near the tip, especially when the contact area is small. However, the stress distribution in the tip-like boundary condition intensifies significantly under a large contacting area. Therefore, the normalized compression force is different as shown in Figure S16c which indicates that the Hertzian contact model for cylinders or semicylinders should be modified to take into account the boundary condition used here. We have found that the Hertzian contact equation for this case (eq 1 in the main text) can be modified to adapt the boundary condition in MD simulation using  $\alpha'/R = \alpha/R(4F^{0.45} + 0.73)$ , where F is the compression force while  $\alpha'$  is the displacement at the boundaries of the tip. For simplicity, in the main text, we still referred to this modified model as a Hertzian contact model because it preserves the same assumptions as the Hertzian contact theory <sup>10-11</sup>.



**Figure S16.** Effect of boundary condition on deformation and stress distribution. (a) and (b) Deformation and stress distribution by using the semicylindrical (a) and the tip-like (b) boundary condition. (c) the force–deformation  $(\alpha'/R)$  relationship for semicylindrical and tip-like boundary conditions in comparison with Hertzian and the modified Hertzian contact model (Hertzian tip).

Supplementary Note 15. Critical contact force  $F_c$  to cause initial yielding is given for the plane strain case as below <sup>12-13</sup> (eq S5):

$$F_{c} = \frac{L\pi^{2}D_{1}D_{2}(V_{1} + V_{2})(CS_{y})^{2}}{2(D_{1} + D_{2})}$$
(S5)

Where  $CS_y = \min(C_1S_{y1}, C_2S_{y2})$ ,  $C_i = 1.164 + 2.975 \ \mu_i - 2.906 \ \mu_i^2$ , and  $S_{yi}$  is the yield strength of  $\alpha$ -quartz, which is suggested to be  $S_{yi} = 0.10 \ E_i$  in previous studies <sup>14-15</sup>. According to the Mises yield criteria,  $S_{yi} = 0.12 \ E_i$  was derived based on the plane strain conditions adopted in this paper. According to eq S5,  $F_c$  for each case is listed in Table S1.

#### REFERENCES

(1) Cygan, R. T.; Liang, J.; Kalinichev, A. G. Molecular Models of Hydroxide, Oxyhydroxide, and Clay Phases and the Development of a General Force Field. *J. Phys. Chem. B* **2004**, *108* (4), 1255-1266.

(2) Granick, S. Motions and Relaxations of Confined Liquids. *Science* **1991**, *253* (5026), 1374-1379.

(3) Horn, R.; Smith, D.; Haller, W. Surface Forces and Viscosity of Water Measured between Silica Sheets. *Chem. Phys. Lett.* **1989**, *162* (4-5), 404-408.

(4) Levine, B. G.; Stone, J. E.; Kohlmeyer, A. Fast Analysis of Molecular Dynamics Trajectories with Graphics Processing Units–Radial Distribution Function Histogramming. *J. Comput. Phys.* **2011**, *230* (9), 3556-3569.

(5) Speedy, R. J.; Madura, J. D.; Jorgensen, W. L. Network Topology in Simulated Water. J. Phys. Chem. 1987, 91 (4), 909-913.

(6) Heyliger, P.; Ledbetter, H.; Kim, S. Elastic Constants of Natural Quartz. J. Acoust. Soc. Am. **2003**, 114 (2), 644-650.

(7) Hartley, N.; Wilshaw, T. Deformation and Fracture of Synthetic A-Quartz. *Journal of Materials Science* **1973**, *8* (2), 265-278.

(8) Chen, C.; Zhang, N.; Shen, W. J.; Li, W. Z.; Song, Y. C. Interaction between Hydroxyl Group and Water Saturated Supercritical Co<sub>2</sub> Revealed by a Molecular Dynamics Simulation Study. *J. Mol. Liq.* **2017**, *231*, 185-191.

(9) Llewellyn, D. Work Hardening Effects in Austenitic Stainless Steels. *Mater. Sci. Technol.* **1997**, *13* (5), 389-400.

(10) Johnson, K. One Hundred Years of Hertz Contact. Proc. Inst. Mech. Eng. 1982, 196 (1), 363-378.

(11) Puttock, M., J.; Thwaite, E, G. Elastic Compression of Spheres and Cylinders at Point and Line Contact. *National Standards Laboratory Technical Paper No. 25. Commonwealth Scientific and Industrial Research Organization, Australia* **1969**.

(12) Green, I. Poisson Ratio Effects and Critical Valus in Spherical and Cylindrical Hertzian Contacts. *Intl. J. Appl. Mech. Eng.* **2005**, *10* (3), 451–462.

(13) Sharma, A.; Jackson, R. L. A Finite Element Study of an Elasto-Plastic Disk or Cylindrical Contact against a Rigid Flat in Plane Stress with Bilinear Hardening. *Tribol. Lett.* **2017**, *65* (3), 112.

(14) Seo, Y.-S.; Ichikawa, Y.; Kawamura, K. Stress-Strain Response of Rock-Forming Minerals by Molecular Dynamics Simulation. *Journal of the Society of Materials Science, Japan* **1999**, *48* (3Appendix), 13-20.

(15) Subramanian, S.; Yip, S. Structural Instability of Uniaxially Compressed A-Quartz. *Comp Mater Sci* **2002**, *23* (1-4), 116-123.