All-atomic Modelling of the Compaction of Montmorillonite Clays: Fabric Evolution and Energy Conversion

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ABSTRACT

Compaction is an essential compression process for sedimentary soils. Compared to in-depth studies on granular soil behaviors, numerical modelling of clay compaction is still in its infancy. This study presents

an all-atomic modelling framework to investigate the compaction of anhydrous montmorillonite from initially fully exfoliated platelets. The total number of inter-particle contacts increased and the meso-pores were dominant during the formation of card-house structures. As the local fabrics evolved to book-house structures, the contact evolutions became predominant and partial meso-pores transformed into micro-pores. The coordinated deformations during the formation of compacted aggregates dramatically increased inter-particle contacts and so the micro-pores became dominant. After rebound, the inter-particle contacts decreased and partial micro-pores were restored. The total potential energy decreased during contact evolutions due to the significant reduction in interaction potential energy between clay particles, while hysteresis was observed during coordinated deformations and rebound due to the changes in internal potential energy within deformed clay particles. The internal potential energy was primarily determined by the electrostatic forces except under significant deformations, where the van der Waals forces became dominant. The interaction potential energy remained unchanged with specific contact types, but decreased significantly due to electrostatic interaction when contacts evolved. As computational capacity develops, a greater number of larger hydrated clay particles can be used to improve simulations of compaction and other macroscopic behaviors via all-atomic molecular dynamics simulations.

AUTHOR KEYWORDS

1   **INTRODUCTION**

2   Compaction, mainly by expulsing air from voids (Benhamida et al. 2005), is an essential compression

3   process for soil sedimentary. The evolution history of fabrics and interactions during compaction shows

4   significant influences on the response of soils in the subsequent engineering applications (Tarantino and

5   De Col 2008). Changes in the interactions between particles under external loads result in fabric evolutions

6   via the rotation, translation and deformation of particles, which consequently lead to the compaction of

7   soils. Therefore, quantitative descriptions of fabric evolution and inter-particle interactions can help to

8   interpret the compaction behavior of soils from particle-scale.

9   For granular soils (e.g., sands), particle-scale behavior is governed by contact forces (i.e., skeletal force)

10   (Santamarina 2003). The fabric evolutions induce changes in the internal-anisotropic contact network,

11   which plays an important role in the compaction of granular soils (Imseeh et al. 2018). Observational

12   imaging techniques, such as X-ray computed tomography (Zhao et al. 2015) and scanning electron

13   microscope (Santagata et al. 2014), have been used to demonstrate the relationships between particle

14   morphology (Nie et al. 2021), fabric evolution (Cheng and Wang 2018), and their influences on the

15   compaction of granular soils (Yin et al. 2023). Due to the multi-scale characteristic lengths of granular soils

16   (Xiong et al. 2022), it is challenging to explore impacts of a single variable merely via observational imaging

17   techniques. The discrete element method (DEM) has been employed as a numerical simulation approach

18   to investigate granular soil behaviors (Cundall and Strack 1979; Zhao and Guo 2013), because it allows for

19   the control of target parameters and continuous monitoring of fabric (Jiang 2019). The DEM framework

20   for granular soils, especially the contact constitutive models, was summarized to establish the benchmark

21   of particulate simulations (O'Sullivan 2011), and it was used to simulate fabric evolution under different

22   loading conditions (Vijayan et al. 2020). These simulations revealed that an increase in the coordination

23   number of soil particles densified the contact network to bear the increasing loading stresses (Gu et al.

24   2015), and the contact evolutions along the principle stress direction contributed to the hysteretic effect

25   (Vijayan et al. 2020).

26   There is however a significant difference in the inter-particle mechanics of granular soils and clay (swelling

27   clay in particular). Clay particles have a large specific surface area (van Olphen 1977) and high surface

28   chemical activity, making the influences of non-contact forces (i.e., van der Waals force, electrostatic force,

29   etc.) between clay particles significant. Hence, the particle-scale behavior of clay is controlled by both

30   contact and non-contact forces (Pedrotti and Tarantino 2018). The factors affecting non-contact forces are

31   highly complex and dependent on mineral structure, charge density and pore media characteristics.

32   Moreover, flat clay particles are prone to deformations with low rigidity (Honorio et al. 2018), which is in

33   stark contrast to granular soil particles. Various morphologies can be formed under different

34   environmental conditions, including single crystal layers, quasi-crystals and aggregates (Quirk and Aylmore

35   1971). To address these challenges, numerical simulations have become a valuable way for demonstrating

36   clay compaction, particularly with the development of computational capacity.

37   DEM for clays differs significantly from DEM for granular soils in both model construction and force field

38   selection. Straight line segments were proposed as discrete element models for clay particles (Fig. 1(a))

39   and hinged nodes were set to approximate particle bending (Anandarajah 1994). Compression behavior

40   was simplified as a two-dimensional (2D) problem (Anandarajah and Lu 1991) and described by

41 mechanical contact, double-layer repulsive and van der Waals attractive forces (Anandarajah 1994). This

42 modelling framework allowed exploration of the relationships between particle behaviors and

43 macroscopic characteristics for clays with low computation costs. Anadarajah's framework has

44 subsequently been modified and extended to three-dimensional problems with 3D-blocks (Yao 2002), and

45 used to simulate the compression of kaolinite (Khabazian et al. 2018; Jaradat and Abdelaziz 2019; Pagano

46 et al. 2020) and montmorillonite (Bayesteh and Mirghasemi 2013) respectively, with a particular focus on

47 the impacts of initial orientation arrangements and ion concentrations. In the recent years, more and more

48 researchers attempted to modify the calculation method of inter-particle forces in DEM. A DLVO-based

49 interaction model, calibrated by the Atomic Force Microscope (AFM) force measurement (Guo and Yu

50 2017), was employed to study the suspension structure of kaolinite in terms of particle orientations and

51 contact numbers (Guo and Yu 2019). A simplified linear force model was adopted to approximate the

52 conventional non-linear interactions and reduce the computational costs in the consolidation simulations

53 of kaolinite (Jaradat and Abdelaziz 2023). A multifaceted interaction model with a specified "pull-off force"

54 between attractive surfaces was proposed (de Bono and McDowell 2023a) to simulate the compression

55 and shearing of kaolin clay (de Bono and McDowell 2023b). However, several challenges remained. The

56 2D-lines with hinged nodes could not simulate the conversion of internal potential energy, while the rigid

57 3D block-shaped particles neglected deformations of clay particles. Additionally, the homogeneous nature

58 of discrete elements could not completely account for the non-homogeneous distribution of clay particle

59 charges caused by isomorphous substitutions and the (de-)protonation of edges. As for inter-particle

60 interactions, since current experimental techniques were unable to directly calibrate model parameters,

61 the approximate methods to calculate repulsive electrostatic force (Lu and Anandarajah 1992) and

62   attractive van der Waals force (Anandarajah and Chen 1995) between two inclined finite particles in DEM

63   were not valid for clays with high surface potential (e.g. montmorillonite) (Bayesteh and Mirghasemi 2013)

64   and easily resulted in errors dependent on particle sizes and inter-particle distances (Chen and

65   Anandarajah 1996).

66   Coarse-grained (CG) simulation method (Suter et al. 2009; Bandera et al. 2021) is an efficient method to

67   perform particle-scale simulations of clay and has also been used to investigate clay agglomeration

68   (Ebrahimi et al. 2016a; Zhu et al. 2019) and compaction (Sjoblom 2016). Each clay particle was modelled

69   by an ellipsoid (Fig. 1(b)), or an assembly of two types of beads (Fig. 1(c)), which represented the main

70   structure and edge of clay particles, respectively. Simulation results successfully revealed the influence of

71   contact number on clay stiffness and clarified that the reorientation during unloading and reloading was

72   the main cause of the hysteresis effect (Sjoblom 2016). However, the CG models were generally rigid and

73   incapable of simulating particle deformations within particles, and the CG simulation method also faced

74   challenges in accurately describing inter-particle interactions. Interactions among CG particles were

75   usually implemented by the DLVO model (Sjoblom 2016; Aminpour and Sjoblom 2019) or other CG force

76   fields (e.g. Gay-Berne potential calibrated by all-atomistic molecular dynamics simulations) (Ebrahimi 2014;

77   Schaettle et al. 2018). The DLVO model may overestimate the electrostatic repulsive force near the surface

78   and neglected the attractive force at the positive-charged edges, resulting in a tendency for dispersion

79   rather than flocculation (Ebrahimi et al. 2014). CG models with Gay-Berne potential (Gay and Berne 1981;

80   Everaers and Ejtehadi 2003) could not simultaneously reflect two free energy valleys of face-face

81   interaction between Na-montmorillonite particles and overestimated the edge-edge interaction at short

82   distance (Ebrahimi et al. 2014; Ebrahimi et al. 2016b).

83   All-atomic modelling for clays became feasible (Cygan et al. 2021) with the development of a general force

84   field for clay minerals, termed CLAYFF (Cygan et al. 2004), which enabled the investigation of clay

85   compaction by all-atomic molecular dynamics (MD) simulations (Fig. 1(d)). All-atomic clay models could

86   describe the local characteristics in mineral structures and charge distributions, and reflect the

87   deformations of flexible clay particles. Combined with laboratory tests and microscopic observations, all-

88   atomic MD simulation reproduced the compaction process of clay aggregates and proposed a prediction

89   model for cohesive energy density (Ahmed and Abduljauwad 2017). Researchers also explored the

90   compaction and dehydration process of clay-like nanoparticle aggregates (Ho et al. 2017; Underwood and

91   Bourg 2020) with all-atomic models, showing the influences of drainage rate and water content on fabric,

92   thermodynamics and mechanical properties, which verified the effectiveness of all-atomic MD simulations

93   in modelling clay behaviors. However, current all-atomic MD simulations mainly focused on the geometric

94   descriptions of fabric evolutions and their influences, lacking analysis of the nanoscale mechanical

95   motivations behind these evolutions.

96   This study presents an all-atomic MD modelling framework to investigate clay compaction from the views

97   of fabric evolution and potential energy conversion. The all-atomic clay models, simulation force field and

98   analysis methods are introduced. In the following section, the fabric evolution is quantitively described by

99   orientation order parameter, variations of inter-particle contacts, pore size distribution and pore space

100   tomography. The laws of potential energy conversion and dissipation in clays are analyzed by potential

101   energy density calculations. The internal potential energy within a typical particle and interaction potential

102   energy between two typical particles are investigated in terms of electrostatic and van der Waals terms

103   for different stages in the compact-rebound process.

104   **MODEL AND METHOD**

105   In this study, to model the fabric evolution of clay during the compact-rebound process at atomistic-scale,

106   an assembly of clay particles are considered within a contained volume. The particles are modelled at the

107   atomic level using the open-source software LAMMPS (Thompson et al. 2022), with inter-atomic force

108   field described in the following.

109   **Molecular models**

110   An all-atomic clay particle model was constructed following the approach of Ho et al. (2017). In this study,

111   Na-montmorillonite (Na-MMT) with stoichiometry of $Na_{0.75}Si_8[Al_{3.25}Mg_{0.75}]O_{20}(OH)_4$ was selected as the

112   representative clay mineral. Initially, a crystalline layer with dimensions of 13.73 nm × 11.88 nm × 0.92 nm

113   was created using the unit cell structure of pyrophyllite proposed by Skipper et al. (1995). The crystalline

114   layer was then cleaved into a hexagonal particle (Underwood and Bourg 2020) with an edge length of 6.86

115   nm along the main growth directions of phyllosilicate crystal chain ([0 1 0], [1 1 0] and [1 -1 0]) (White and

116   Zelazny 1988), as depicted in Fig. 2(a). The undercoordinated oxygen atoms exposed at the edges were

117   healed with hydrogen atoms in neutral-pH environment (Martins et al. 2014; Lavikainen et al. 2015).

118   Isomorphous substitutions were performed by randomly replacing aluminum atoms at the octahedral

119   sheets with magnesium atoms, following the Lowenstein's Law (Lowenstein 1954). This resulted in a

120   permanent charge of -196e (e = $1.602×10^{-19}$ C) and a surface charge density of -0.128 C/m^2 in a clay particle,

121   which conformed to the experimental results (-0.193 – -0.125 C/m^2) (Sperry and Peirce 1999). To maintain

122   overall electroneutrality, those negative charges were compensated by sodium ions (Na$^+$). Unlike earlier

123   rigid particle models for montmorillonite (Ebrahimi et al. 2014; Kang et al. 2020), the clay particles in this

124   study are flexible and capable of deformations. Subsequently, 100 clay particles, each of which consisted

125   of about 10 thousand atoms, were randomly placed in a 70.8-nm cubic simulation box, forming a loose

126   packing with a void ratio of 30.0 to prevent particle overlapping in the initial model, as illustrated in Fig.

127   2(b). It is noteworthy that a parametric study with 200 clay particles was also conducted to investigate the

128   influences of model size. The results indicated that variation laws of fabric evolution and energy conversion

129   during the compact-rebound process were model size independent.

130   **Force field parameters and simulation details**

131   The inter-atomic interaction force field in this study referred to the widely-used CLAYFF (Cygan et al. 2004),

132   which has been successfully employed to simulate clay mineral systems (Mazo et al. 2008; Anoukou et al.

133   2015; Prishchenko et al. 2018; Cygan et al. 2021; Zhu et al. 2021). The total potential of the system U^{total},

134   consisted of the non-bonded interaction potential $U^{\text{non-bonded}}$, and the bonded interaction potential U^{bonded}.

135   The former included Coulombic electrostatic interaction U^{Coul}, and van der Waals interaction U^{vdW}. The

136   latter was composed of a bond-stretching term U^{bond}, and an angle-bending term U^{angle} :

137    $$U^{\text{total}} = U^{\text{non-bonded}} + U^{\text{bonded}} = U^{\text{Coul}} + U^{\text{vdW}} + U^{\text{bond}} + U^{\text{angle}} \tag{1}$$

138    $$U^{\text{Coul}} = \frac{e^2}{4\pi\epsilon} \sum_{i \neq j} \frac{q_i q_j}{r_{ij}} \left(r_{ij} \leq r_c \right) \tag{2}$$

139    $$U^{\text{vdW}} = \sum_{i \neq j} 4\varepsilon_{ij} \left[\left(\frac{\sigma_{ij}}{r_{ij}} \right)^{12} - \left(\frac{\sigma_{ij}}{r_{ij}} \right)^6 \right] \left(r_{ij} \leq r_c \right) \tag{3}$$

140 $$U^{\text{bond}} = D_0 \left\{ 1 - \exp\left[-\alpha \left(r_{ij} - r_0 \right) \right] \right\}^2$$ (4)

141 $$U^{\text{angle}} = k_0 \left(\theta_{ijk} - \theta_0 \right)^2$$ (5)

142 The cutoff distance r_c for the short-range part of non-bonded interaction was 1.0 nm. The short-range part

143 of U^{Coul} was given by Coulombic law, where ϵ is the dielectric constant and q is the atomistic partial charge,

144 while the long-range part of U^{Coul} was calculated by Particle-Particle and Particle-Mesh (PPPM) solver

145 (Hockney and Eastwood 1989). Most of the atomistic partial charges were obtained from quantum

146 chemistry calculation as described in CLAYFF, but the partial charges of edge oxygen atoms were re-

147 assigned due to the (de-)protonated process at the edges (Lammers et al. 2017). The short-range U^{vdW} was

148 calculated by Lennard-Jones potential, where $\varepsilon_{ij} \left(= \sqrt{\varepsilon_i \varepsilon_j} \right)$ and $\sigma_{ij} \left(= \left(\sigma_i + \sigma_j \right)/2 \right)$ are energy parameter

149 and distance parameter between atom i and atom j, respectively. It includes a repulsive term (i.e., r^{-12} term,

150 referring to Pauli repulsion) and an attractive term (i.e., r^{-6} term, referring to London dispersion) (Wang et

151 al. 2020). Only the bond-stretching terms of hydroxyl groups were considered, where D_0 is the energy

152 constant, α is the damping parameters and r_0 is the equilibrium length of bonds in hydroxyl groups. The

153 angle-bending terms between the metal atoms and the hydroxyl groups (Metal-O-H) were described by

154 harmonic function, where k_0 is the force constant and θ_0 is the equilibrium angle of Metal-O-H (Cygan et al.

155 2004). These parameters were empirically derived from experimental data of mineral crystal structures

156 (Pouvreau et al. 2017; Pouvreau et al. 2019). Besides, to ensure the stability of clay particle models in long-

157 term MD simulations, additional angle-bending potential with a large force constant (k_0 = 800 kcal·mol⁻

158 ¹·rad⁻²) was assigned to the Metal-O-Metal groups at the edges. (Ho et al. 2017) Detailed force field

159 parameters are listed in the Supplementary Materials.

160   The analyses for the all-atomic MD simulations, which were undertaken on Intel(R) Xeon(R) Gold 6226R

161   CPUs (2.90 GHz), involved three steps: initial structure optimization, compaction and rebound. First, the

162   loose packing (Fig. 2(b)) was energy-minimized to obtain an optimized structure. The system temperature

163   was then set to 10 K to reduce the thermodynamic fluctuations, and the initial velocities of atoms were

164   assigned based on a Gaussian distribution. After eliminating the initial stress in the model with an anneal

165   simulation under canonical ensemble (NVT, with constant particle numbers, volume and temperature),

166   the isothermal-isobaric ensemble (NPT, with constant particle numbers, pressure and temperature) was

167   applied with an isotropic pressure P = 1 atm in three dimensions of the simulation box until the size of the

168   simulation box reached a stable state. The obtained model (43.7 nm × 44.4 nm × 32.2 nm), with a void

169   ratio e_0 of about 4.45 and a dry density ρ_{d0} of about 0.50 g/cm^3, was regarded as the initial state for the

170   compact-rebound simulation. Subsequently, one-dimensional compaction was performed at a rate of 0.02

171   nm/ps along the z-dimension, while the x and y dimensions remained fixed. This compaction rate was

172   selected to reduce the sampling errors and avoid the influences of overfast relative movements of particles

173   on fabric evolution. The atom positions were remapped according to the changing size of the simulation

174   box under the NVT ensemble. The compaction continued until ρ_d reached 1.98 g/cm^3, and the maximum

175   simulated pressure reached about 17.4 MPa, which slightly exceeded static compaction or geological

176   pressure (0.01 GPa), but was far inferior to dynamic compaction pressure (1 GPa) (Ahmed and

177   Abduljauwad 2017). After compaction, the rebound simulation was performed inversely. An equilibrium

178   period of 1 ns and a sampling period of 0.5 ns were conducted after each step of compaction and rebound.

179   The entire compact-rebound simulations lasted for about 40 ns.

180   A time step of 1.0 fs was used, and periodic boundary conditions (Frankel and Smit 2002) were applied to

181   each boundary surface of the domain. System equilibrium criterion was based on the stricter

182   thermodynamics equilibrium condition as opposed to a mechanical equilibrium condition chiefly used in

183   DEM simulations. Throughout the compact-rebound process, thermodynamics properties were monitored

184   to ensure the system had reached a thermodynamic equilibrium state after each equilibrium period.

185   The compact-rebound process was quantitatively analyzed in terms of fabric and energy. According to the

186   definition from Mitchell and Soga (2005), the term "fabric" refers to the arrangement of particles, particle

187   groups, and pore spaces. In our study, the correlation parameter between the particle normal vector and

188   the positive direction of z-axis (i.e., the orientational order parameter) S_z (Allen and Tildesley 2017) was

189   calculated to represent the arrangement of particles; the variations of inter-particle contacts were

190   investigated to reflect the arrangement of particle groups since the contact between two clay particles

191   was the fundamental part of particle groups; the pore size distribution (PSD) and pore space tomography

192   were presented to manifest the arrangement of pore spaces.

193   The singular value decomposition (SVD) algorithm (Arun et al. 1987) was first adopted to match the point

194   sets of atoms and obtain the normal vector $\mathbf{n}$ of particle surface (Supplementary Materials). With the

195   obtained normal vectors of particles, S_z could be calculated as follows:

196    $$S_z = \frac{1}{N_p} \sum_{i=1}^{N_p} \left| \frac{3\left(\mathbf{n}_i \cdot \mathbf{e}_z\right)^2 - 1}{2} \right|$$ (6)

197   where $N_p = 100$ is the total number of clay particles and $\mathbf{e}_z$ is the unit vector of z-direction. For randomly-

198   aligned clay sample, the normal vectors of particles are independent of the z-direction and $S_z = 0$; for

199   transverse-isotropic clay sample, the normal vectors of particles are parallel to the *z*-direction and $S_z = 1$.

200   Six basic types of inter-particle contacts were considered according to the contact sites and particle

201   morphologies: Vertex-Vertex (VV), Vertex-Edge (VE), Vertex-Face (VF), Edge-Edge (EE), Edge-Face (EF) and

202   Face-Face (FF) contacts, as shown in Fig. 3. A recognition algorithm was developed based on the geometric

203   criteria for different types of contacts (Supplementary Materials). The numbers of inter-particle contacts

204   were then compiled via the algorithm and the average contact numbers per particle (*CN*) were calculated

205   to avoid the impact of system size. Specifically, two particles with $\left| \mathbf{n}_i \cdot \mathbf{n}_j \right| \geq 0.97$ could be regarded as parallel;

206   otherwise, two inclined clay particles were in contact if the minimum distance between octahedral metal

207   atoms d_s was smaller than 1.5 nm. According to the stability analysis in Supplementary Materials, the rank

208   of stability for different contact types was as follows: FF > (EF > VE > EE) > (VF > VV). In this paper, the

209   evolution of inter-particle contacts refers to the transformation from low-stability contacts to high-stability

210   contacts, while degeneration refers to the reverse process.

211   To analyze the pore size distribution (PSD) of clay, pore diameter *D* at any given point $P\left(x_p, y_p, z_p\right)$ was

212   defined as the maximum diameter of a sphere that encompassed point *P* without overlapping neighboring

213   atoms (Bhattacharya and Gubbins 2006) (refer to Fig. S3(a) for a schematic graph). The detailed calculation

214   method is given in the Supplementary Materials. To visualize the morphologic changes of pore space, the

215   simulation box was divided into a cubic grid with a side length of about 0.15 nm and the space state of

216   each grid cell was determined based on the distances between the center of grid cell and atoms in the

217   system. To ensure a smooth transition between pore space and non-pore space, a Gaussian function was

218   adopted as a switching function (Supplementary Materials) (Lu and Chen 2012). Finally, the software Visual

219   Molecular Dynamics (VMD) (Humphrey et al. 1996) was utilized to generate a contour map of the space

220   state of each grid cell, which represented the tomography of pore space.

221   The fabric evolution induced potential energy conversion. In order to assess the stability of clay fabrics

222   under different dry densities, the variations in total potential energy stored within a unit volume of clay

223   (i.e., total potential energy density) compared to the initial state with $\rho_{d0} = 0.50$ g/cm^3, $\Delta\Omega_{sys}$, was taken as

224   a characterization parameter. During the sampling period, the total potential energy U^{total} and volume of

225   clay V were collected, and $\Delta\Omega_{sys}$ could be calculated as:

$$\Delta\Omega_{sys} = \Delta\Omega_{in} + \Delta\Omega_{ia} = \left\langle \frac{U^{total}}{V} \right\rangle - \left\langle \frac{U_0^{total}}{V_0} \right\rangle \tag{7}$$

227   where U_0^{total} is the total potential energy of the initial system, V_0 is the initial volume of clay and the notation

228   < > represents the ensemble-averaged value of variables. $\Delta\Omega_{sys}$ could be further decomposed into the

229   variations in internal potential energy density within clay particles $\Delta\Omega_{in}$ and interaction potential energy

230   density between clay particles $\Delta\Omega_{ia}$.

231   As for potential energy conversion of a single particle, variations in internal potential energy Δu_{in}^{total} and

232   interaction potential energy Δu_{ia}^{total} were analyzed. Δu_{in}^{total} was comprised of a Coulombic electrostatic term

233   Δu_{in}^{Coul}, a van der Waals term Δu_{in}^{vdW} and a bonded term Δu_{in}^{bonded}. It was dependent on the deformations of

234   each particle; thus, an index K was defined to quantify the deformations of particles along their normal

235   vector with respect to their initial undeformed state:

$$K = \sum_{i=1}^{N} \delta_{zi}^2 \tag{8}$$

237   where δ_{zi} is the deflection of the ith octahedral metal atom in the deformed particle (see Supplementary

238   Materials for a schematic diagram) and N is the total number of octahedral metal atoms in the particle. A

239   larger value of K indicated a greater particle deformation along the normal vector and a larger value of

240   Δu_{ia}^{total}. Similarly, Δu_{ia}^{total} included a Coulombic electrostatic term Δu_{ia}^{Coul} and a van der Waals term Δu_{ia}^{vdW}.

241   **FABRIC EVOLUTION**

242   Typical states in the simulated compact-rebound process were shown in Fig. 4. Particles were initially

243   scattered with large voids between them and were randomly-aligned with respect to the z-direction (Fig.

244   4(a), P = 0.1 MPa, ρ_d = 0.50 g/cm^3). Particles gradually approached each other under external pressure,

245   leading to a reduction in inter-particle distances (Fig. 4(b), P = 0.6 MPa, ρ_d = 0.89 g/cm^3). The ensuing

246   compaction led to the increase of the number of parallel-stacked particles and the normal vectors of

247   particles tended to align with the compaction direction (Fig. 4(c), P = 1.4 MPa, ρ_d = 1.33 g/cm^3). Further

248   compaction caused the coordinated deformations between particles, leading to a transverse-isotropic clay

249   system (Fig. 4(d), P = 17.4 MPa, ρ_d = 1.98 g/cm^3), which was similar to the soil layers after geological

250   sedimentation. After unloading, there was a slight recovery with layered structure being maintained, and

251   partial small pores recovered (Fig. 4(e), P = 63.1 kPa, ρ_d = 1.63 g/cm^3). A video of the entire simulated

252   compact-rebound process and the loading/unloading curve in the e-log(P) space are provided in the

253   Supplementary Materials (see Fig. S2).

254   As shown in Fig. 5, S_z increased stepwise from 0.02 to 0.48 as the compaction progressed. The maximum

255   S_z was smaller than the observed value (0.70) in the orientation test of the hydrated Na-montmorillonite

256   (Carrier et al. 2016). At Stage A (ρ_d < 0.89 g/cm^3), S_z dramatically increased to 0.23, indicating that the

257   interaction between scattered particles played an important role in guiding the rotation of particle normal

258   vectors. At Stage B ($0.89 \leq \rho_d < 1.33$ g/cm^3), the Pauli repulsion between particles inhibited the further

259   orientational rotation of clay particles (Underwood and Bourg 2020), resulting in a slow increment of S_z .

260   At Stage C ($1.33 \leq \rho_d < 1.98$ g/cm^3), S_z continuously increased to 0.48 due to the rotations and transversely

261   slides between particles during the coordinated deformations within the parallel-stacking particles. At

262   Stage D ($1.98 \geq \rho_d > 1.63$ g/cm^3), S_z decreased gradually with the recovery of elastic particle deformations,

263   and the value after rebound was slightly smaller than that before rebound under identical dry density,

264   which suggested that partial orientational rotations during the coordinated deformation stage within

265   book-house structures were irreversible.

266   **Local fabrics**

267   Consideration of a sub-domain of 12 particles gives insight into local fabric evolution. A typical path of the

268   compact-rebound process is shown in Fig. 6, with evolution from (a) card-house structure → (b) book-

269   house structure → (c) compacted aggregate → (d) post-unloading structure. During the early stage of

270   compaction, a voluminous card-house structure was formed primarily through vertex/edge contacts

271   (Pedrotti and Tarantino 2018) (Fig. 6(a)). This open fabric was rich in interconnected meso-pores due to

272   the relatively large inter-particle distances. As compaction progressed, the inter-particle contacts evolved

273   via the rotation around contact sites and the reduction in inter-particle distances. The card-house structure

274   gradually collapsed, transforming to a book-house structure (Mitchell and Soga 2005) (Fig. 6(b)). Within

275   the parallel-stacked particles (i.e., "books"), micro-pores appeared at the edges owing to the imperfect

276   stacking of monolayered particles (Michot and Villiéras 2006), which has been observed via cryo-electron

277   tomography (Whittaker et al. 2020). Flexible clay particles, characterized by a propensity for tangling

278   (Honorio et al. 2018), contributed to transverse slides and coordinated deformations within "books" in the

279   book-house structure, which is similar to the cake fragment of montmorillonite after high pressure freeze-

280   drying of the suspensions observed by cryo-SEM (Mouzon et al. 2016). This led to further volume reduction

281   with the shrinkage of micro-pores, ultimately forming a compacted aggregate (Fig. 6(c)). After releasing

282   the external pressure, particles rebounded and partial deformations were restored (Fig. 6(d)).

283   **Inter-particle contacts**

284   Evolution of the average contact numbers per particle are depicted as the variation curves with ρ_d in Fig.

285   7. It is the first time to quantitatively explore the changes in arrangements of particle groups via the

286   thorough analyses of inter-particle contact variations during the compact-rebound process. The average

287   number of total contacts per particle CN_{total} showed a stepwise increase during the compact process, as

288   shown in Fig. 7(a). At the formation stage of inter-particle contacts (Stage A), as dispersed clay particles

289   approached each other, many inter-particle contacts were newly formed, leading to an increase in CN_{total}

290   from 2.25 to 2.77. Stage B was dominated by inter-particle contact evolution, along with a small increase

291   in CN_{total} to 3.04. At the stage of coordinated deformations between particles (Stage C), the further

292   reduction in the inter-particle distances led to a continuous increase in CN_{total} to around 4.00. However, the

293   enhanced van der Waals repulsive force (i.e., Pauli repulsion) hindered the further reduction in inter-

294   particle distances, causing a slowdown in contact number growth at the late stage. At Stage D, elastic

295   deformations within particles were recovered and the increasing inter-particle distances directly resulted

296   in the degeneration or loss of inter-particle contacts, with CN_{total} decreasing to about 3.41.

297   The formations and evolutions of inter-particle contacts coexisted in the compact-rebound process, with

298   different dominant forms at each stage, as shown in Fig. 7(b). At Stage A, there was a predominance of

299   newly-formed VV, VF, EE contacts, while at Stage B the evolution from VE and EF contacts to FF contacts

300   was prominent, contributing to a decrease of CN_{VE} and CN_{EF} to about 0.50 and a sustainable increase in

301   CN_{FF} from 0.92 to 1.25. As particles slid transversely and inter-particle distances decreased, Stage C was

302   characterized by the formations of VE, EF and FF contacts, with CN_{VE}, CN_{EF} and CN_{FF} increasing to 0.67, 0.83

303   and 1.69, respectively. After unloading at Stage D, partial EF and FF contacts were detached or gradually

304   degenerated into VV and VF contacts. As a result, CN_{EF} and CN_{FF} decreased to 0.46 and 1.41, respectively.

305   In summary, the stepwise variation stages of inter-particle contacts were alternately characterized by the

306   formation of low-stability contacts and the evolution from low-stability contacts (i.e., VE and EF contacts)

307   to high-stability contacts (i.e., FF contacts) during compaction.

308   **Pore structure**

309   According to the International Union of Pure and Applied Chemistry (IUPAC) (Rouquerol et al. 1994;

310   Kaufhold et al. 2013), pores with diameters of 2–50 nm were categorized as meso-pores (Fig. 6(a)) and

311   pores smaller than 2 nm were categorized as micro-pores (Fig. 6(b)). Cumulative pore size curves and PSD

312   histograms for different dry densities were plotted in Fig. 8. Meso-pores dominated during the formation

313   and collapse of card-house structures (ρ_d ≤1.33 g/cm^3), while micro-pores dominated during the

314   coordinated deformation of book-house structures and the rebound process (ρ_d > 1.33 g/cm^3). Due to the

315   small particle sizes, macro-pores of montmorillonite (50-100 nm) observed via mercury intrusion

316   porosimetry (Souli et al. 2013; Wei et al. 2013) did not emerge in this simulation.

317   The mean pore diameter $\bar{D}$ was defined as the pore diameter smaller than which the pores accounted

318   for 50% of total pores. In the initial model (Fig. 8(a), ρ_d = 0.50 g/cm³), meso-pores accounted for about

319   95.2% of the total pore volume. The PSD showed a broad single peak with $\bar{D}$ of around 7.0 nm and a

320   maximum pore diameter of about 12.2 nm. As compaction progressed (Fig. 8(b), ρ_d = 0.89 g/cm³), $\bar{D}$

321   decreased to about 4.4 nm and a minor micro-pore peak gradually generated. The collapse of card-house

322   structures resulted in a unimodal pore size distribution with $\bar{D}$ of about 2.5 nm (Fig. 8(c), ρ_d = 1.33 g/cm³).

323   Throughout these stages, pores were mostly interconnected and open (Insets in Fig. 8(a-c)). In the

324   compacted aggregate, micro-pores accounted for 79.2% of the total pore volume. The inter-particle

325   coordinated deformations reduced the sizes of micro-pores, with $\bar{D}$ of the narrow unimodal curve

326   reducing to about 1.2 nm (Fig. 8(d), ρ_d = 1.98 g/cm³). Pore spaces became fragmented with discrete and

327   closed pores as deformed particles cleaved through them (Inset in Fig. 8(d)). Unloading caused partial

328   micro-pores to transform into meso-pores, with a reduction of micro-pore proportion to 59.0% (Fig. 8(e),

329   ρ_d = 1.63 g/cm³), and pores reconnected as inter-particle distances increased (Inset in Fig. 8(e)). Compared

330   to the brick-like fabric formed by rigid particle models (Honorio et al. 2018; Khabazian et al. 2018), a fabric

331   with low porosity, small open-pores and a higher presence of lenticular pores was reproduced due to the

332   flexible all-atomic clay particle models used in this study.

333   **POTENTIAL ENERGY CONVERSION**

334   **Potential energy density of the whole system**

335   Clearly, a more mechanically stable fabric possessed a lower value of $\Delta\Omega_{sys}$. As shown in Fig. 9, $\Delta\Omega_{sys}$

336   decreased to -1.50×10⁻²⁰ J/nm³ as the inter-particle contacts gradually evolved into the most stable FF

337   contacts at Stage A and B. However, further compaction at Stage C reduced the inter-particle distances

338   and increased the steric repulsion, resulting in an increase in $\Delta\Omega_{sys}$ to 4.04×10^{-20} J/nm³. At Stage D with $\rho_d >$

339   1.63 g/cm³, $\Delta\Omega_{sys}$ decreased to -2.21×10^{-20} J/nm³ due to the recovery of elastic deformations and the

340   enlargement of inter-particle distances. The energy dissipation throughout the entire compact-rebound

341   process was 2.21×10^{-20} J/nm³ greater than the input volume work, and it could be inferred that part of the

342   formation and evolution of inter-particle contacts (Stage A and B) contributed most to the dissipation.

343   Furthermore, $\Delta\Omega_{sys}$ after rebound was lower than that before unloading at the same dry density, indicating

344   that the remaining irreversible energy dissipation emerged during the coordinated deformations and

345   rebound process.

346    $\Delta\Omega_{in}$ and $\Delta\Omega_{ia}$ were of the same order of numerical magnitude, indicating that the influences of the

347   deformations of montmorillonite particles on the potential energy conversion could not be ignored, as

348   shown in Fig. 9. During the compact process, $\Delta\Omega_{in}$ increased to 2.26×10^{-18} J/nm³ due to the intensified

349   particle deformations, while $\Delta\Omega_{ia}$ decreased to -2.22×10^{-18} J/nm³ as a result of the evolution of inter-

350   particle contacts. At Stage A and B, where particle deformations were comparatively insignificant,

351   evolution of inter-particle contacts played a major role, and $\Delta\Omega_{ia}$ was chiefly responsible for the reduction

352   in $\Delta\Omega_{sys}$. Conversely, particle deformations dominated during Stage C, and the increase in $\Delta\Omega_{in}$ was the

353   main reason for the rapid increase in $\Delta\Omega_{sys}$. During Stage D, $\Delta\Omega_{in}$ decreased by 7.74×10^{-19} J/nm³ due to the

354   recovery of elastic deformations, while $\Delta\Omega_{ia}$ increased by 7.12×10^{-19} J/nm³ due to the enlargement of inter-

355   particle distances and the degenerations of contacts. Therefore, the reduction in $\Delta\Omega_{in}$ was the main cause

356   of the decline in $\Delta\Omega_{sys}$ during the rebound stage. To our knowledge, it is the first time in the literatures to

357   monitor the potential energy conversion during the compaction-rebound process and link it to the fabric

358   evolution observed.

359   Although all particles exhibited similar variation trends in Δu_{in}^{total} and Δu_{ia}^{total}, typical particles No.61 and

360   No.98 were selected for further investigation at particle level in the following two sections, due to their

361   representative deformation characteristics and relatively complete evolution path.

362   **Internal potential energy within a clay particle**

363   Compared to the initial undeformed state, the deformation index K and Δu_{in}^{total} of the typical particle No.98

364   were plotted in Fig. 10. The deformations of particle No.98 during the compact-rebound process could be

365   categorized as five phases in terms of K in Fig. 10(a). At Phase I ($\rho_d \leq 0.69$ g/cm^3), the particle was not

366   subjected to significant external forces and experienced minimal deformation ($K < 8.50$ nm^2). At Phase II

367   ($0.69 < \rho_d \leq 1.14$ g/cm^3), torsional deformation developed and K increased to 190.42 nm^2. At Phase III (1.14

368    $< \rho_d \leq 1.59$ g/cm^3), the particle transformed to a saddle-shape and K sharply increased to 518.67 nm^2. At

369   Phase IV ($\rho_d > 1.59$ g/cm^3), the relaxation of tilting-up vertexes led to a reduction in curvature and the

370   particle's shape transformed to an arch, with K decreasing to 416.17 nm^2. At Phase V (i.e., the rebound

371   process), partial elastic deformations were released and K gradually decreased to 338.58 nm^2. The curved

372   platelets in this simulation resemble the montmorillonite clay particles observed via SEM in the previous

373   literatures (Ece et al. 1999; Leong et al. 2022).

374   As mentioned in the *MODEL AND METHOD*, Δu_{in}^{total} consisted of Δu_{in}^{Coul}, Δu_{in}^{vdW} and Δu_{in}^{bonded}. It was found that

375   the repulsive interactions between oxygen atoms contributed most to Δu_{in}^{Coul} during particle deformations,

376   and Δu_{in}^{vdW} raised with the increase of particle volume strain (Supplementary Materials). As shown in Fig.

377   10(b), the contribution of Δu_{in}^{vdW} to Δu_{in}^{total} was less significant than that of Δu_{in}^{Coul} , and Δu_{in}^{bonded} was omitted

378   because it was neglectable compared to Δu_{in}^{Coul} and Δu_{in}^{vdW} .

379   In the compact process, Δu_{in}^{total} increased with dry density and had great relevance to stages of particle

380   deformations. At Phase I, with small deformations, both Δu_{in}^{Coul} and Δu_{in}^{vdW} remained close to zero. At Phase

381   II and III, inter-atomic distances decreased and the redistribution of Na^+ occurred as the particle deformed,

382   as shown in Fig. 10(c), which resulting in the increase of Δu_{in}^{Coul} and Δu_{in}^{vdW} to 3.23×10^{-17} J and 1.10×10^{-17} J,

383   respectively. At Phase IV, Δu_{in}^{vdW} increased to 2.19×10^{-17} J but Δu_{in}^{Coul} kept relatively unchanged, indicating

384   that the increase in Δu_{in}^{total} was attributed to the contribution of Δu_{in}^{vdW} at this phase.

385   In the rebound process (i.e., Phase V), Δu_{in}^{total} decreased from 5.24×10^{-17} J to 4.04×10^{-17} J, among which

386   Δu_{in}^{vdW} decreased by 1.70×10^{-17} J while Δu_{in}^{Coul} increased by 0.52×10^{-17} J, suggesting the reduction of Δu_{in}^{total} in

387   the rebound process chiefly resulted from the decrease of van der Waals repulsion as partial elastic

388   deformations recovered and volume strain decreased. However, Δu_{in}^{vdW} became greater and Δu_{in}^{Coul} became

389   smaller than those before rebound with identical dry density, which indicated irreversible changes in van

390   der Waals and Coulombic electrostatic interactions emerged during the rebound process, due to plastic

391   deformations (such as, atomic dislocations and ruptures) and the non-recoverability of Na^+ redistribution.

392   **Interaction potential energy between clay particles**

393   Δu_{ia}^{total} , Δu_{ia}^{Coul} and Δu_{ia}^{vdW} between the typical particles No.61 and No.98 are plotted in Fig. 11. In the

394   compaction process, Δu_{ia}^{total} decreased with dry density in stages. Obviously, Δu_{ia}^{total} reached a plateau once

395   a specific inter-particle contact (i.e., VE, EF and FF contacts) was formed. Δu_{ia}^{total} decreased by 0.28×10^{-18} J

396   as particles evolved from VE contact into EF contact, and decreased by 9.12×10^{-18} J as they evolved from

397   EF contact into FF contact. It could be inferred that Δu_{ia}^{total} decreased with the enlargement of contact areas.

398   Calculations of interaction force between particles (Supplementary Materials) indicated that the attractive

399   electrostatic force of cations on negatively-charged clay crystalline layer slightly exceeded the summation

400   of repulsive electrostatic forces between clay crystalline layers and between cations, while the van der

401   Waals force between atoms at the contact sites was repulsive and that between atoms far from contact

402   sites was attractive. As the particle rotated around the contact sites, positive work was done by

403   electrostatic attraction and the van der Waals attraction between atoms far from the contact sites, while

404   the van der Waals repulsion at the contact sites almost did no work due to the constant contact distances,

405   leading to the decreases of Δu_{ia}^{Coul} and Δu_{ia}^{vdW} as the inter-particle contacts evolved. This is similar to the

406   detachment mechanism in the concept model proposed by Pedrotti (Pedrotti and Tarantino 2018),

407   resulting in an irreversible volume change.

408   A hysteresis loop of Δu_{ia}^{total} was observed throughout the compaction and rebound process. Under identical

409   dry density, there was a slight change in Δu_{ia}^{vdW} before and after rebound, indicating that it was almost

410   reversible. However, there was a significant difference in Δu_{ia}^{Coul} before and after rebound, highlighting that

411   the irreversibility of electrostatic interaction was attributed to the hysteresis observed in the variation

412   curve of Δu_{ia}^{total} .

413   **CONCLUSIONS**

414   This paper proposes an all-atomic molecular dynamic modelling framework to investigate the compact-

415   rebound process of anhydrous montmorillonite platelets from the view of fabric evolution and potential

416   energy conversion. The fabric evolution, which was characterized by card-house structure, book-house

417   structure, compacted aggregate and post-unloading structure, was quantified via orientation order

418   parameter, variations in inter-particle contacts, pore size distribution and pore space tomography.

419   Potential energy density calculations were performed to analyze the laws of energy conversion and

420   dissipation in clays. The variations of the internal potential energy and interaction potential energy of

421   typical particles were investigated in terms of electrostatic and van der Waals terms for different stages in

422   the compact-rebound process.

423   Compared to the brick-like fabric formed by rigid particle models, a fabric with smaller open pores and

424   more lenticular pores was reproduced with flexible clay particle models in this study. The average number

425   of total contacts per particle increased stepwise in the compact process and decreased in the rebound

426   process. Generation of low-stability contacts (e.g., VV, VF and EE) dominated during the formation of card-

427   house structures. During the collapse of card-house structures into book-house structures, the fabric was

428   dominated by the evolution from low-stability contacts to high-stability contacts (e.g., EF and FF). Partial

429   contacts formed at the later coordinated deformation stage were lost or degenerated during rebound.

430   The variations in total potential energy density included the internal potential energy density and

431   interaction potential energy density, which were of the same order of magnitudes in the compact-rebound

432   process. The internal term increased during compaction and decreased during rebound, while the

433   interaction term showed the opposite trend. The total potential energy density decreased when inter-

434   particle contacts evolved to more stable types, which mainly resulted from a significant reduction in

435   interaction potential energy density between particles. Afterwards, it raised dramatically during the stage

436   of coordinated deformations and gradually decreased during rebound, primarily owing to the

437   corresponding variations in the internal potential energy within particles.

438   The deformation of a clay particle and its induced variations in the internal potential energy within the

439   particle during the compact-rebound process could be simulated with the flexible clay particle model. The

440   internal potential energy of a typical particle increased during compaction but decreased during rebound.

441   The electrostatic term consistently contributed more to the internal potential energy within the particle

442   than the van der Waals term, but the latter played an indispensable role as particle deformations became

443   relatively large in the later compact and rebound stages. The interaction potential energy between

444   particles remained unchanged once a specific contact type was formed, while it decreased stepwise

445   primarily due to the electrostatic interaction when the inter-particle contact evolved into a more stable

446   type and the contact area enlarged. The hysteresis of the total interaction potential energy observed in

447   the compact-rebound process was mainly attribute to the irreversibility of the electrostatic interaction.

448   Computational expense for such models was significant. The simulation presented in this study took nearly

449   6 months of consecutive calculations on high-performance computer clusters. The size of particles was

450   small (about 1/10 of the transverse size of actual montmorillonite particles) and monotypic, which

451   resulted in large values of energy density, fictitious high stiffness of clay (Ebrahimi et al. 2016b) and

452   intensive thermodynamic fluctuations with respect to those obtained in the laboratory tests. There was a

453   lack in considering the influences water molecules, which could hydrate surface cations to form the

454   electrical double layers, in turns changing the inter-particle interactions. In future work, better simulations

455   of compaction and other macroscopic behaviors of clays via all-atomic MD can be achieved with

456   consideration of a greater number of larger hydrated clay particles as computational capacity develops.

457

458   **NOTATION**

459   CN average number of contacts per particle (subscripts stand for specific contact types)

460   D pore diameter

461    $\bar{D}$ mean pore diameter (i.e., pores smaller than this size account for 50% of the total pores)

462   e void ratio (e_0 is the void ratio of initial model before compaction)

463   K deformation index of the particle

464    $\mathbf{n}$ normal vector of particle surfaces

465   N total number of octahedral metal atoms in each particle

466   N_p total number of clay particles in the simulation box

467   S_z orientation order parameter of particles with respect to the z-axis

468   u_{ia}^{Coul} Coulombic electrostatic term of interaction potential energy between two clay particles

469   u_{ia}^{vdW} van der Waals term of interaction potential energy between two clay particles

470   u_{ia}^{total} total interaction potential energy between two clay particles

471   u_{in}^{bonded} bonded term of internal potential energy within a single clay particle

472   u_{in}^{Coul} Coulombic electrostatic term of internal potential energy within a single clay particle

473   u_{in}^{vdW} van der Waals term of internal potential energy within a single clay particle

474   u_{in}^{total} total internal potential energy within a single clay particle

475	U^{angle}	the angle-bending potential energy of the whole system
476	U^{bond}	the bond-stretching potential energy of the whole system
477	U^{bonded}	the bonded potential energy of the whole system
478	U^{Coul}	the Coulomb electrostatic potential energy of the whole system
479	U^{vdW}	the van der Waals potential energy of the whole system
480	$U^{non-bonded}$	the non-bonded potential energy of the whole system
481	U^{total}	the total potential energy of the whole system (U_0^{total} is the initial total potential energy)
482	V	volume of clay (V_0 is the volume of the initial model before compaction)
483	δ_z	z-deflection of the deformed particle
484	ρ_d	dry density (ρ_{d0} is the dry density of initial model before compaction)
485	Ω_{ia}	interaction potential energy density of the whole system
486	Ω_{in}	internal potential energy density of the whole system
487	Ω_{sys}	total potential energy density of the whole system

488

489 DATA AVALABILITY STATEMENT

490 All data, models, and code generated or used during the study appear in the submitted article.

491

496

497   SUPLEMENTAL MATERIALS

498   Figs. S1–S12 and Tables S1-S4 are available online in the ASCE Library (www.ascelibrary.org). The video of

499   the whole compress-rebound process can also be found online in the ASCE Library (www.ascelibrary.org).

500

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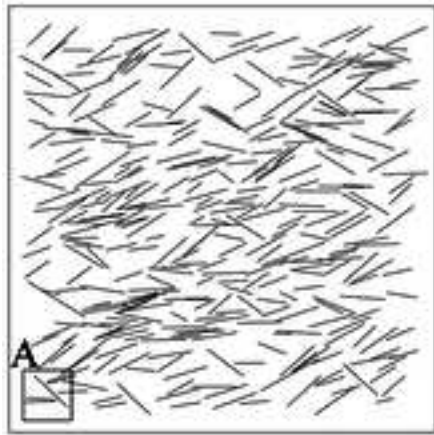
(a) 2D-line discrete element model

Adapted from H. Bayesteh and A. A.
Mirghasemi (2013). Copyright 2012
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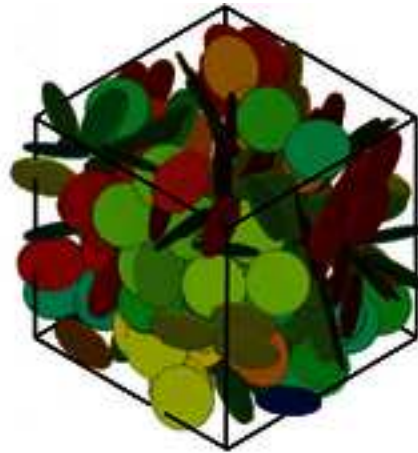
(b) Ellipsoidal particle model

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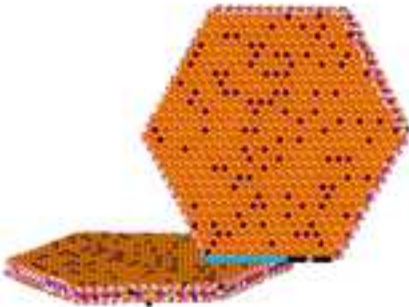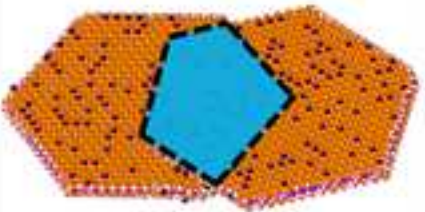LLC.
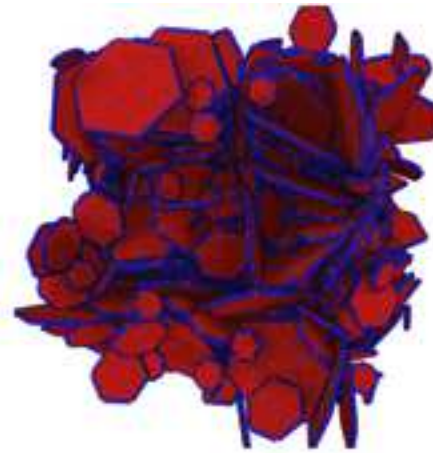
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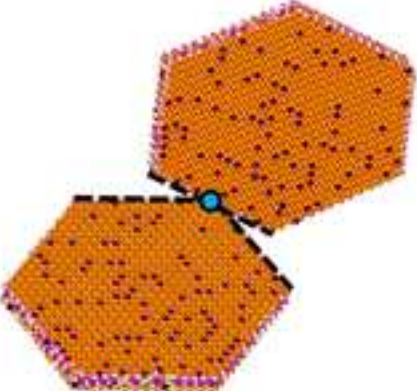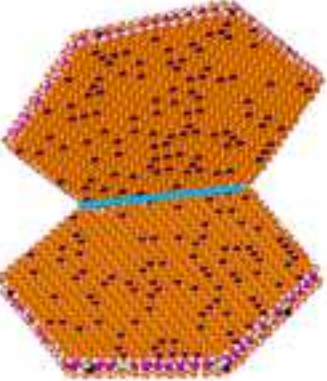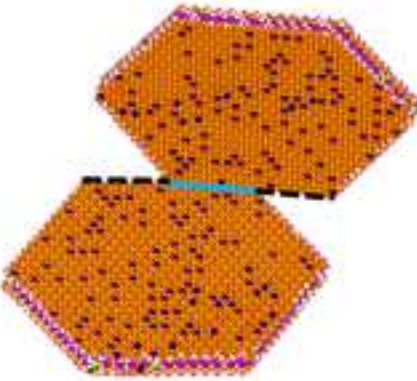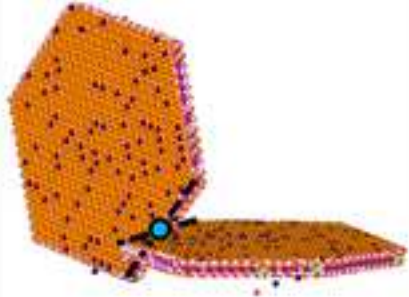
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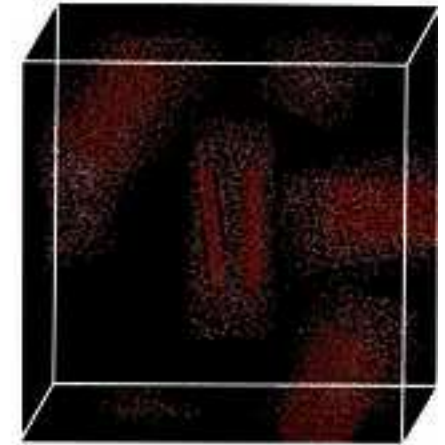
(d) All-atomic model

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(a)   (b)

Figure 3
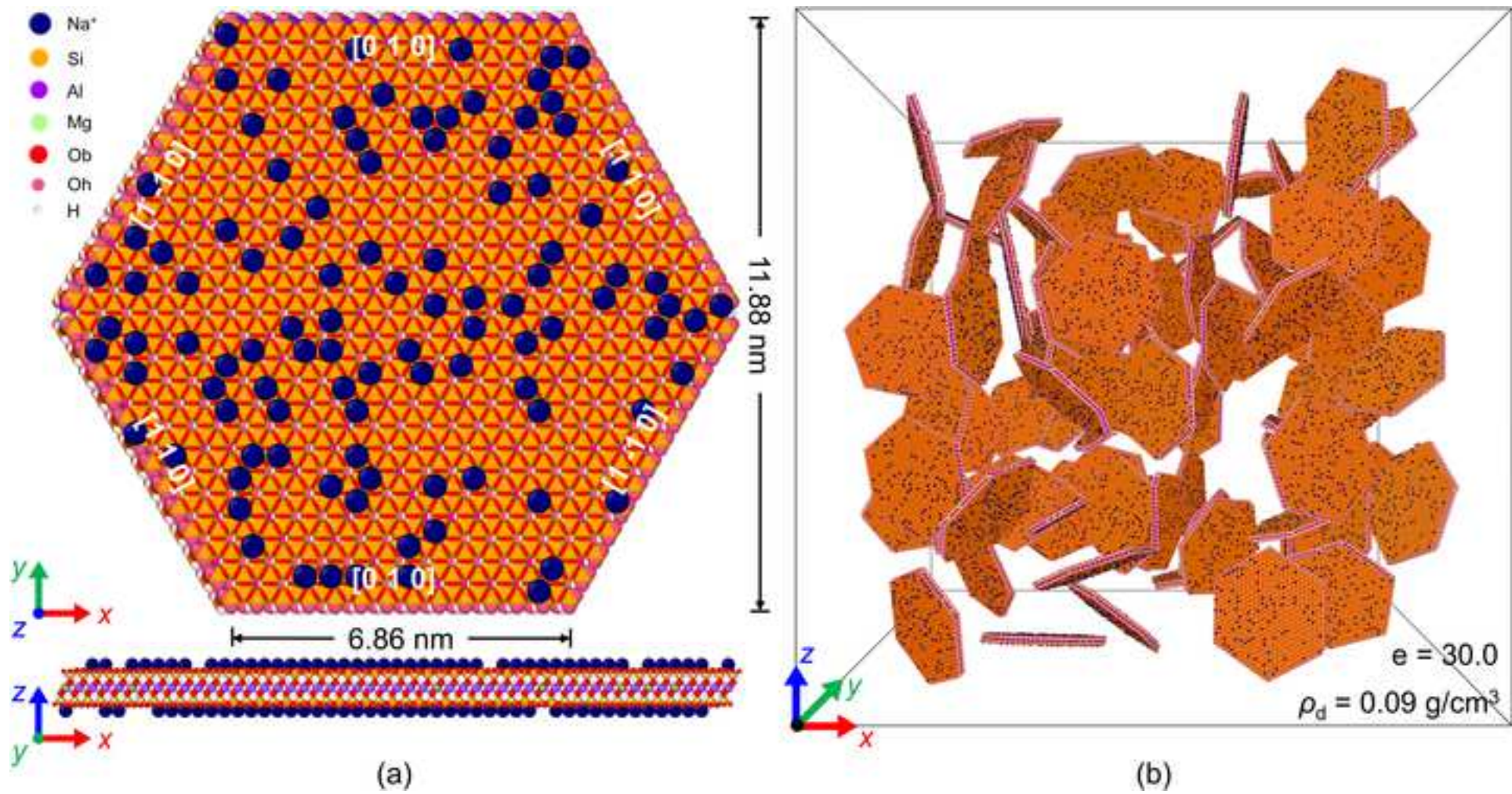
Vertex - Vertex	Vertex - Edge	Vertex - Face

Edge - Edge

Type I (full contact)	Type II (partial contact)	Type III (point contact)

Edge - Face

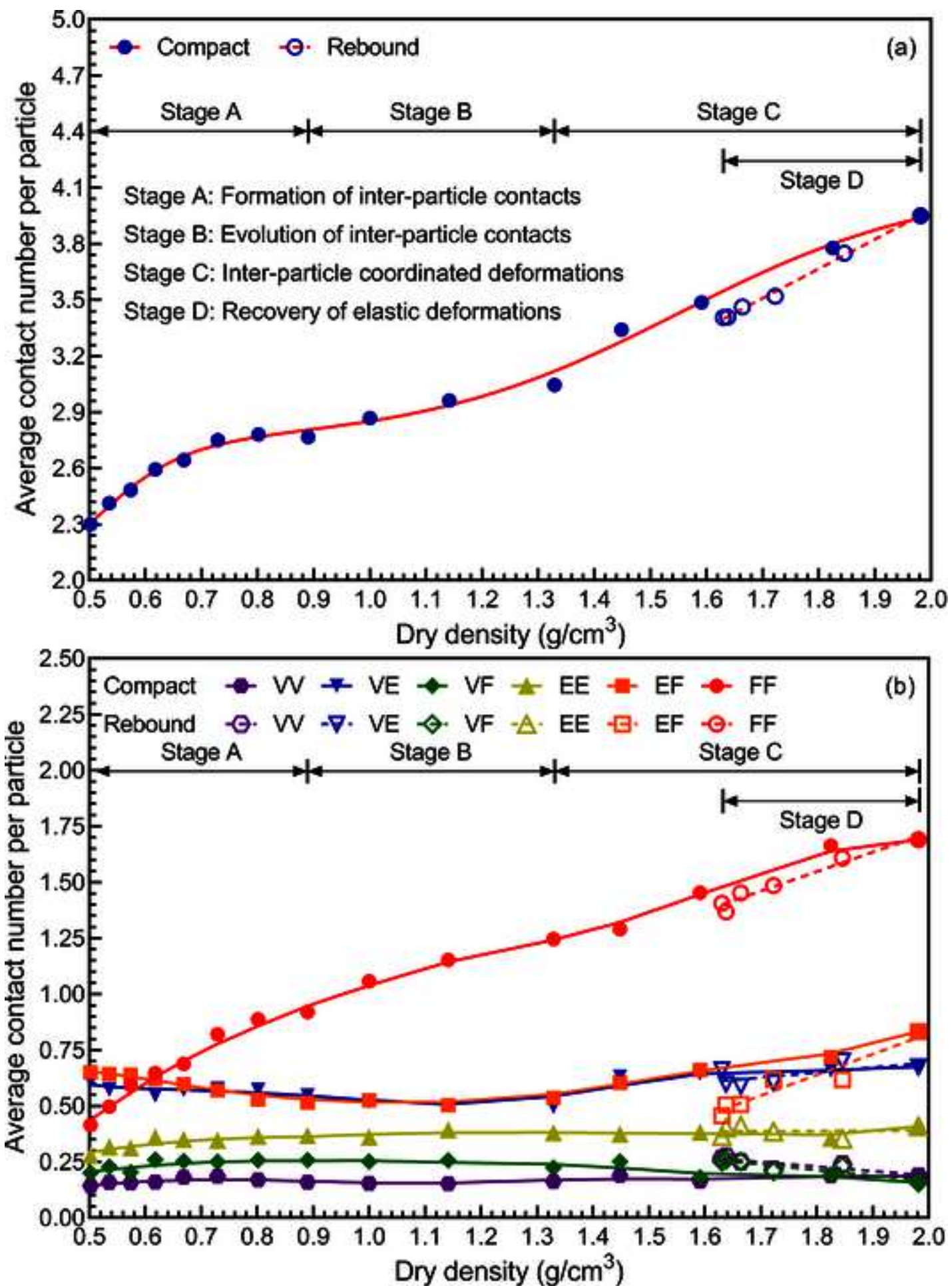| Type I (full contact)	Type II (partial contact)	Face - Face

Figure 4

(a) $e = 4.45$ $\rho_d = 0.50 \ g/cm^3$

(b) $e = 2.08$ $\rho_d = 0.89 \ g/cm^3$

(c) $e = 1.06$ $\rho_d = 1.33 \ g/cm^3$

(d) $e = 0.38$ $\rho_d = 1.98 \ g/cm^3$

(e) $e = 0.68$ $\rho_d = 1.63 \ g/cm^3$

Figure 5

Figure 6

Meso-pores
2 nm < D < 50 nm

Micro-pores
D ≤ 2 nm

fabric evolution

coordinated deformation

card-house structure
(a)

"book" in book-house structure
(b)

coordinated deformation

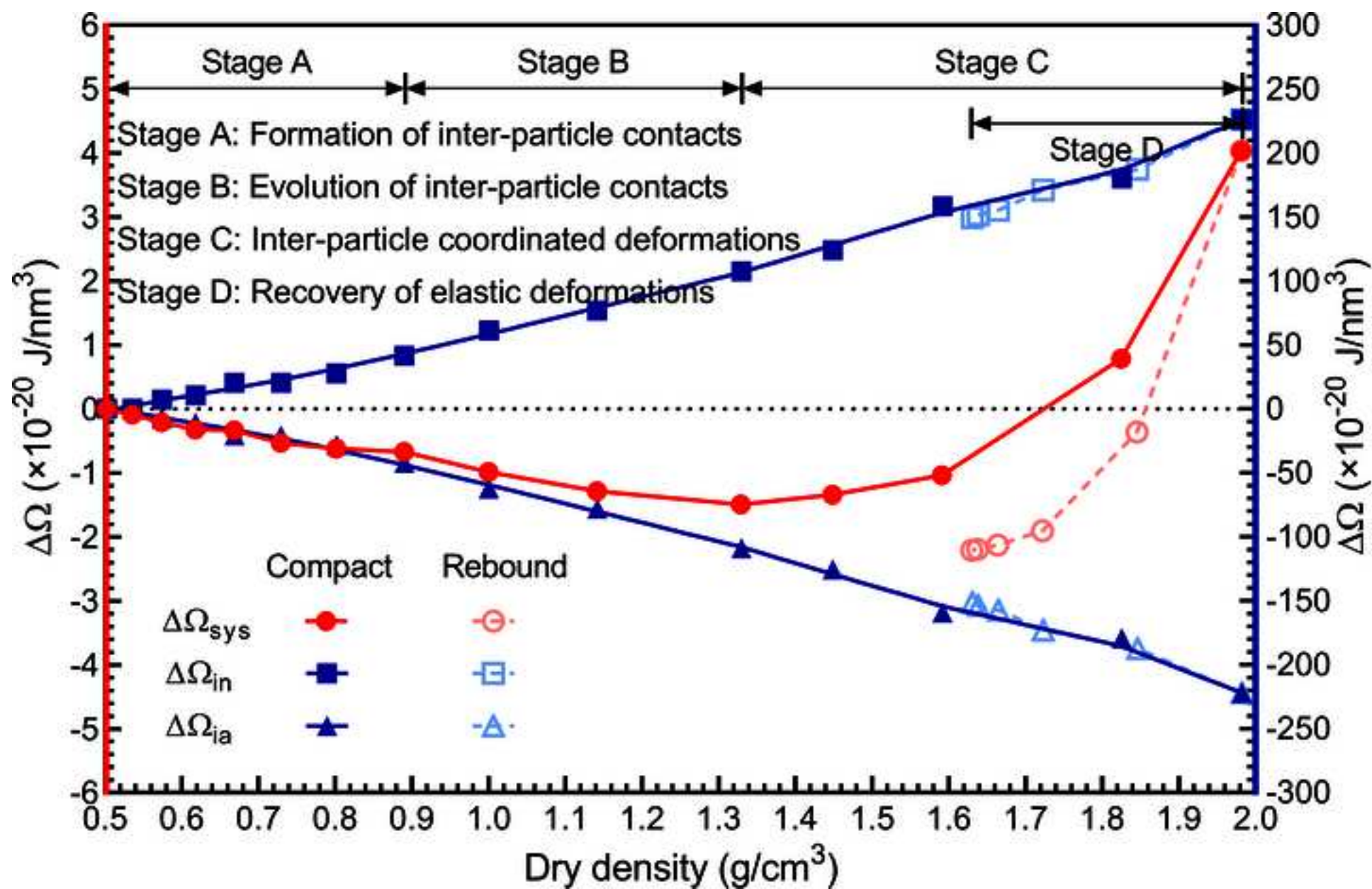rebound

compacted aggregate
(c)

post-unloading structure
(d)

Figure 7

Click here to access/download;Figure;Figure 7. CN.tif ±

Figure 7. (a) Average contact number per particle versus dry density for Compact and Rebound. Stage A: Formation of inter-particle contacts; Stage B: Evolution of inter-particle contacts; Stage C: Inter-particle coordinated deformations; Stage D: Recovery of elastic deformations. (b) Average contact number per particle for VV, VE, VF, EE, EF, FF contact types.

Figure 8   Click here to access/download;Figure;Figure 8. PSD.tif ⬇

Figure 9

Figure 11

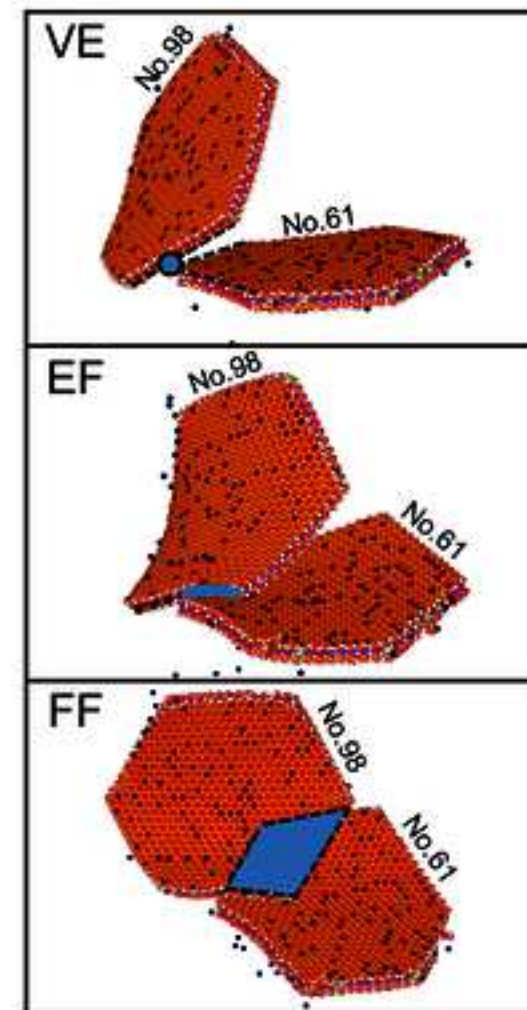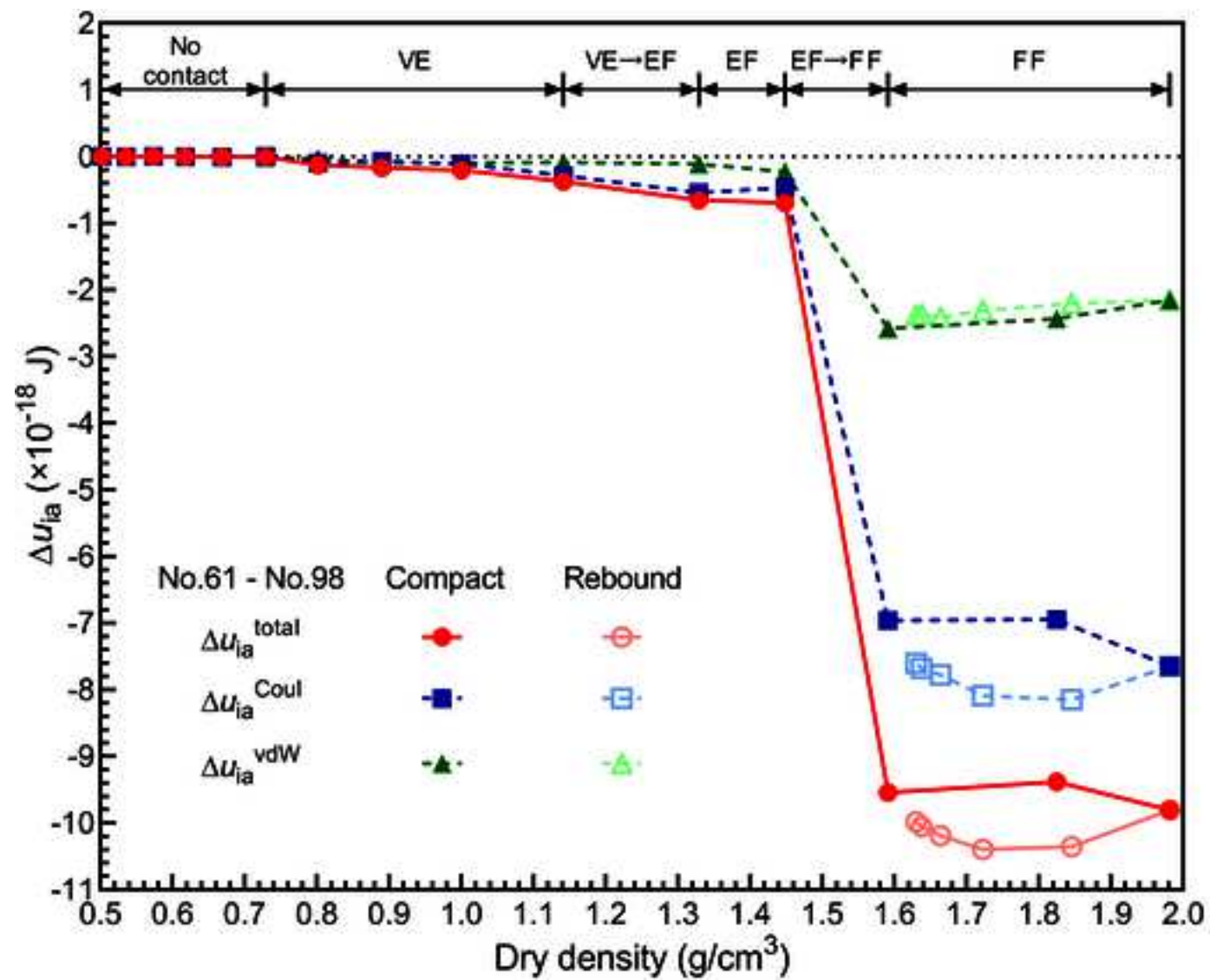Figure 11. Interaction Potential Energy
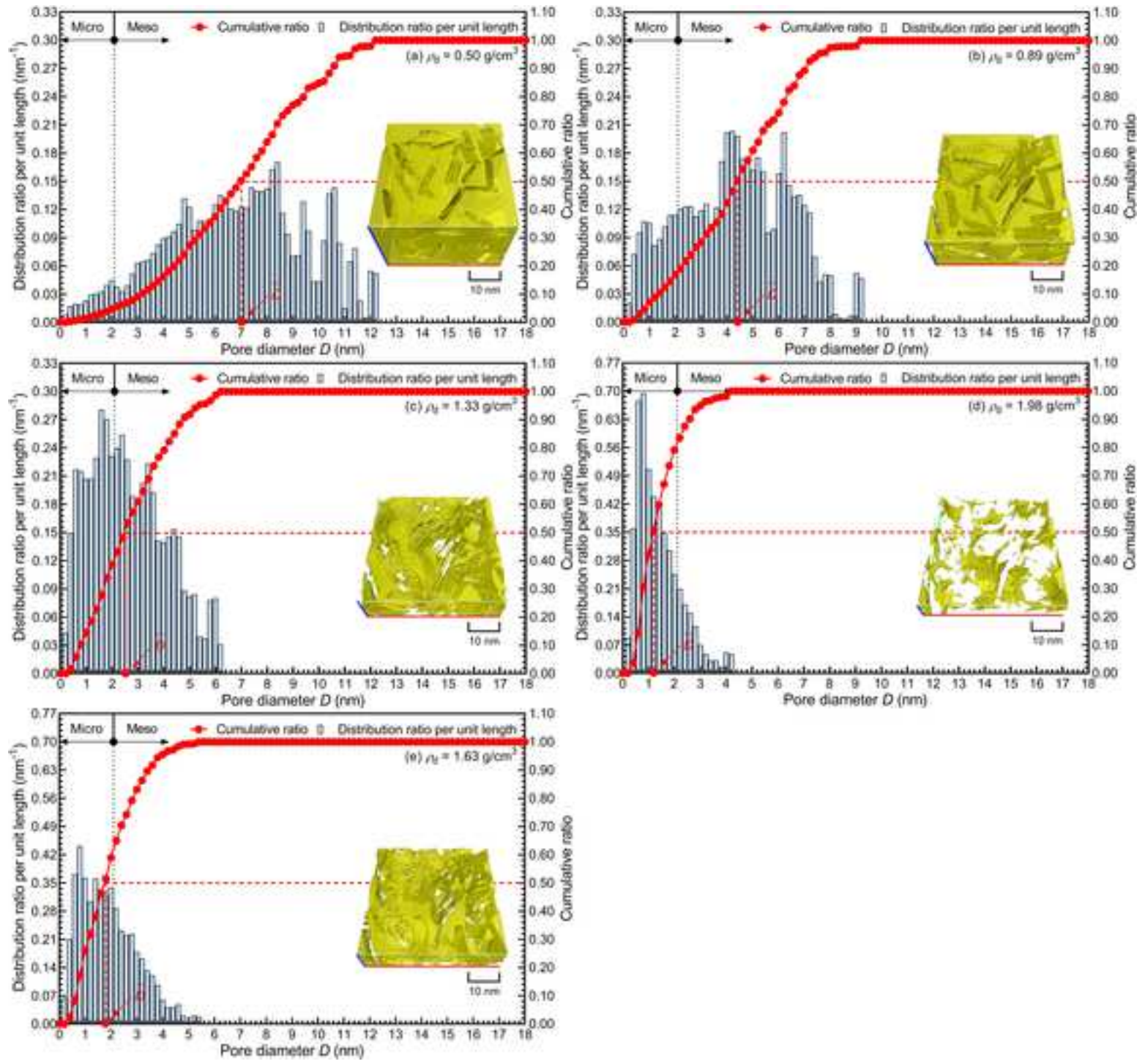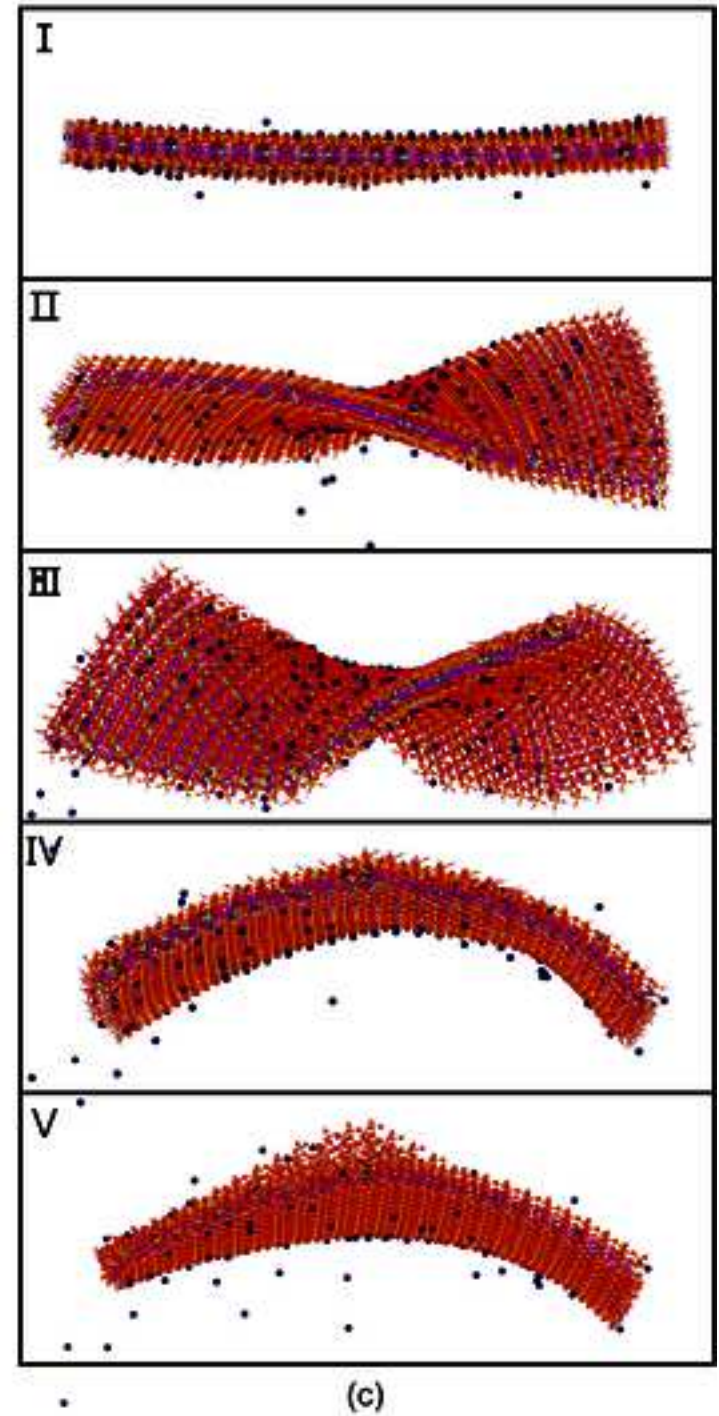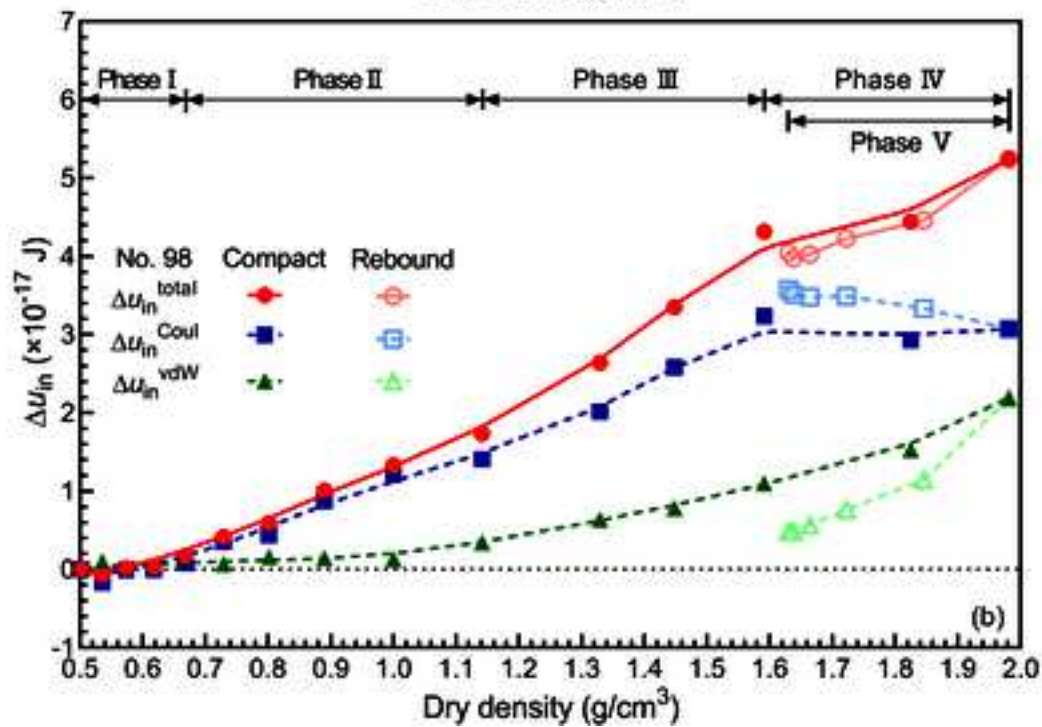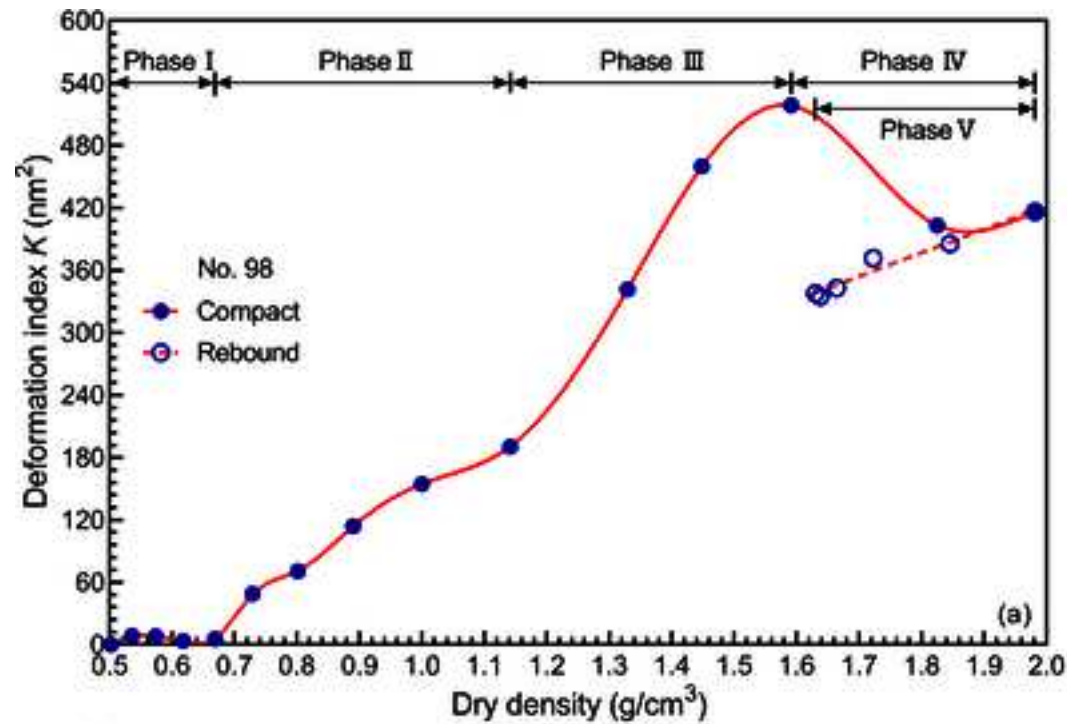
Figure Caption List

Fig. 1. Schematic simulation models of clays adaptive from previous researches. (a) 2D-line discrete element model (Anandarajah 1994); (b) Ellipsoidal particle model (Ebrahimi et al. 2014); (c) United-bead particle model (Aminpour and Sjoblom 2019); (d) All-atomic model (Ahmed and Abduljauwad 2017).

Fig. 2. Molecular models for clay particles and their initial assemblage. (a) All-atomic model for a single clay particle; (b) Initial loose packing of 100 particles.

Fig. 3. Diagram of different types of inter-particle contacts. Blue represents the contact point, line or area.

Fig. 4. Snapshots of typical states of the whole system during the compact-rebound process.

Fig. 5. Variation curve of orientation order parameter S_z with dry density.

Fig. 6. Snapshots of the evolution of local clay fabric of 12 clay particles during the compact-rebound process.

Fig. 7. Variations of average number of (a) total contacts and (b) various types of contacts per particle during the compact-rebound process.

Fig. 8. Pore size distribution histogram and cumulative curve for simulated clay with (a) ρ_d = 0.50 g/cm3, (b) ρ_d = 0.89 g/cm3, (c) ρ_d = 1.33 g/cm3, (d) ρ_d = 1.98 g/cm3 and (e) ρ_d = 1.63 g/cm3 (post-unloading). Insets are the pore space of the simulated clay with corresponding dry densities.

Fig. 9. Variations of potential energy density during the compact-rebound process. Red circles represent $\Delta\Omega_{sys}$ plotting on the left y-axis, blue squares represent $\Delta\Omega_{in}$ plotting on the right y-axis, and blue triangles represent plotting on the right y-axis.

Fig. 10. Variations of particle No.98 in (a) deformation index K, (b) internal potential energy and (c) representative deformation snapshots.

Fig. 11. Variations in interaction potential energy between particles No.61 and No.98.

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