

Highlights

A Phase-Field Discrete Element Method to study chemo-mechanical coupling in granular materials*

Alexandre Sac--Morane, Manolis Veveakis, Hadrien Rattez

- Development of an extension of the discrete element method using a phase-field formulation (solved with finite elements) to incorporate grain shape and its evolution.
- Highlight the influence on the grain shape during an oedometer test under acid injection.
- Model the phenomenon of pressure solution at the microstructural level.

A Phase-Field Discrete Element Method to study chemo-mechanical coupling in granular materials

Alexandre Sac--Morane^{a,b,*}, Manolis Veveakis^a, Hadrien Rattez^b

^a*Multiphysics Geomechanics Lab, Duke University, Hudson Hall Annex, Room No. 053A, Durham, 27708, NC, USA*

^b*Institute of Mechanics, Materials and Civil Engineering, UCLouvain, Place du Levant 1, Louvain-la-Neuve, 1348, Belgium*

Abstract

This paper presents an extension of the discrete element method using a phase-field formulation to incorporate grain shape and its evolution. The introduction of a phase variable enables an effective representation of grain geometry and facilitates the application of physical laws, such as chemo-mechanical couplings, for modeling shape changes. These physical laws are solved numerically using the finite element method coupled in a staggered scheme to the discrete element model. The efficacy of the proposed Phase-Field Discrete Element Model (PFDEM) is demonstrated through its ability to accurately capture the real grain shape in a material subjected to dissolution only and compute the stress evolution. It is then applied to model the phenomenon of pressure solution involving dissolution and precipitation in granular materials at the microscale and enables to reproduce the creep re-

*It is important to notice that color need to be used for any Figures in print.

*Corresponding author

Email addresses: `alexandre.sac-morane@uclouvain.be` (Alexandre Sac--Morane), `manolis.veveakis@duke.edu` (Manolis Veveakis), `hadrien.rattez@uclouvain.be` (Hadrien Rattez)

sponse observed experimentally. This framework contributes to the enhanced understanding and simulation of complex behaviors in granular materials and sedimentary rocks for many geological processes like diagenesis or earthquake nucleation.

Keywords: discrete element model, phase field, granular material, pressure solution, chemo-mechanical couplings

1. Introduction

The intricate interplay between chemical and mechanical processes in soil and rocks has emerged as a key factor to consider for many engineering applications like underground storage or geothermal energy [1] or to understand geological processes like diagenesis or earthquake nucleation [2]. In particular, underground storage strategies envisioned to store large amounts of CO_2 , captured from large point sources like power generation facilities, or H_2 , produced during overproduction periods of renewable energies like solar or wind, involve the injection of a fluid into a reservoir rock. This fluid reacts with the surrounding rock inducing dissolution and/or precipitation that affects rocks' permeability [3], but also their mechanical behavior [4]. To ensure the long-term success and safety of such storage solutions, the role of chemo-mechanical couplings needs to be understood and modeled as they can influence settlement at the surface [5], the stability of the caprock [6, 7] or induced seismicity [8].

Chemo-mechanical couplings are also fundamental to many geological processes. For example, understanding of diagenetic transformations and in particular the phenomenon of pressure solution [9] relies on a comprehen-

19 sive grasp of these couplings. During the diagenetic processes sediments are
 20 transformed into sedimentary rocks. These processes influence reservoir qual-
 21 ity in hydrocarbon exploration and the formation of economically valuable
 22 mineral deposits. Moreover, Pressure solution [10], has also a pivotal role in
 23 earthquake nucleation and recurrence. It involves three chemo-mechanical
 24 processes at the micro-scale [11]: dissolution due to stress concentration at
 25 grain contacts, diffusive transport of dissolved mass from the contact to the
 26 pore space, and precipitation of the solute on the less stressed surface of the
 27 grains. These processes lead to a time-dependent compaction of the rock
 28 by changing its microstructure, pore structure and composition[11]. It also
 29 induces a modification of the strength of a wide range of geological materials.
 30 As such, it has been suggested as a mechanism for long-term evolution of fault
 31 strength during interseismic periods [12], but also to control faults' frictional
 32 behavior at low speed and thus influences earthquake nucleation[10].

33 Chemo-mechanical couplings have already been investigated with finite-
 34 element models at the meso/macro scale [13] but this kind of formulation
 35 strongly depends the chosen constitutive equations, which are not well con-
 36 strained as they depend on the specific microstructure and chemical reac-
 37 tions considered. To better understand the driving processes simulations
 38 on the microstructure should be considered. To do so, the discrete model
 39 (DEM) [14] is used in the present study as it enables the micromechani-
 40 cal analysis of granular materials and sedimentary rocks by reproducing the
 41 interactions among individual grains and grains' rearrangement [15]. Tra-
 42 ditional DEM formulations assume grains as disks in two dimensions (2D)
 43 or spheres in three dimensions (3D). The dissolution/precipitation is then

44 taken into account as a homogeneous decrease/increase of the grain diame-
 45 ter [16, 17] or with an addition/subtraction of a dissolved/precipitated layer
 46 thickness at the contacts [18]. The extrapolation of the results obtained from
 47 those approaches to geological applications is limited as they only consider
 48 disks or spheres as grains, but real particles often exhibit highly irregular
 49 shapes. These complex geometries of the grains significantly influence the
 50 macroscopic mechanical behavior of granular materials [19, 20]. Therefore,
 51 accurate models should aim to capture this complexity. Various approaches
 52 have been developed within the framework of DEM to account for irregular
 53 grain shapes. These include incorporating rolling resistance calibrated with
 54 grain geometry [21, 22], utilizing grain clusters [23], using ellipsoids [24], em-
 55 ploying superquadric particles [25], and even considering polyhedral shapes
 56 [26, 27, 28]. More recently, a level-set discrete element model was developed
 57 to capture the complex shapes of grains and reproduce experimental results
 58 [29, 30]. However, none of those approaches consider a grain shape evolution
 59 due to chemo-mechanical couplings.

60 Considering the granular material as a phase, the phase-field theory (PF)
 61 [31, 32, 33, 34] provides a suitable framework for modeling the local addi-
 62 tion or reduction of material quantity using physics-based laws. Dissolu-
 63 tion/precipitation at the contact interfaces is controlled by introducing me-
 64 chanical and chemical energy into the Allen-Cahn formulation of the phase
 65 variables, while the mass conservation and the solute diffusion are addressed
 66 through a coupled diffusion formulation of the solute concentration [35]. No-
 67 tice that phase-field description is not sufficient alone to describe the mi-
 68 crostructure evolution. In granular matter, grain reorganization is described

as discrete displacements of the grains. Phase-field formulation is solved with a Finite Element Method, so it is a continuous description, which is less suited than DEM approaches to model at the grain scale large deformation and contact topology changes as involved in grain reorganization.

Previous studies have already explored the coupling between finite element modeling and discrete element methods to study deformable grains [36] or grain growth and sintering [37]. In this study, we present an extension of the discrete element method to simulate irregular particle shapes in granular materials and their heterogeneous evolution using the phase-field variable as a geometric descriptor. We apply this method to two cases: (i) an oedometric test with partial dissolution and (ii) the pressure solution phenomenon involving two or multiple grains. The first case highlights the influence of grain shape and dissolution on the mechanical behavior, while the second case investigates the effects of heterogeneous dissolution and precipitation on the rate of material compaction.

2. Methodology

The objective of this paper is to present a novel coupling between a Phase-Field (PF) model and a Discrete Element Model (DEM). In this section, the algorithms and constitutive equations for discrete element models and the phase field are presented separately. The framework of the model is then presented to give an overview of the algorithm and how each model involved in the coupling scheme is used.

2.1. DEM

Time integration

93 The linear kinematics of grains are described by their center $\mathbf{C}$. Various
 94 integration methods can be used [38], but for computational efficiency, an
 95 explicit method is chosen, the Symplectic Euler method. It is selected for
 96 its suitability and accuracy in the simulation [38], with time discretization
 97 playing a significant role in the simulation outcome. The rotation of the
 98 grains is also considered, especially in shearing problems. The rotation inte-
 99 gration follows the same scheme. A rigid body rotation is used to compute
 100 the coordinates of the grain vertices.

101 Mass m and inertia I of the grains, used to solve the momentum balances
 102 [15], must be updated during a simulation as chemical reactions will modify
 103 the shape of the grain (dissolution/precipitation occurring). A Monte Carlo
 104 method [39] is used after each phase-field simulation to solve this problem,
 105 see Section 2.3.

106

107 *Contact detection*

108 As the particles are polygonal, an algorithm is needed to detect contact
 109 between grains i and j . First, the nearest vertex of grain j to grain i needs
 110 to be determined. This is achieved by applying a virtual displacement, as
 111 described in Equation 1, to the vertices of grain j .

$$\mathbf{p}^v = \mathbf{p} + \max(R_i, R_j) \times \frac{\mathbf{C}_j - \mathbf{C}_i}{\|\mathbf{C}_j - \mathbf{C}_i\|} \quad (1)$$

112 Here, $\mathbf{p}^v$ represents the virtual coordinates of the vertex, $\mathbf{p}$ denotes the
 113 coordinates of the vertex, $R_{i,j}$ is the maximum radius of the grain i, j (grains
 114 have different radii depending on the direction), and $\mathbf{C}_{i,j}$ denotes the coor-

115 dinates of the center. This virtual translation assumes that the grains are
 116 not in contact (even if they were before this operation). After the virtual
 117 displacement, the distances between vertices of grains i and j are computed.
 118 The vertices used for contact detection are determined by considering the
 119 nearest ones after the virtual displacement.

120 Next, the algorithm of the common plane is applied to compute the con-
 121 tact normal vector $\vec{n}$ [25, 26, 27] and the normal distance between grains
 122 is defined as $\delta_n = d^j - d^i$ where $d^{i,j}$ is the distance between grain and the
 123 common plane. It is important to note that particles are in contact only if
 124 the overlap δ_n is negative.

125 This contact algorithm, especially the virtual displacement approach, is
 126 conventional for spherical particles but its robustness is questionable for very
 127 complex shapes. Other proximity detection algorithms are available in the
 128 literature [40, 41, 42] and work better in the latter case. However, the per-
 129 formances of this algorithm are acceptable for grains with shapes close to
 130 spheres (as is the case here). A more suitable algorithm for contact detec-
 131 tion will be the focus of future works.

132 *Interaction between grains*

133 The interaction between two grains is illustrated in Figure 1 and described
 134 by the following constitutive laws. The normal response consists of a nonlin-
 135 ear spring and a dashpot in parallel, along with a no-tension joint. Similarly,
 136 the tangential response includes a nonlinear spring and a dashpot in parallel,
 137 a slider, and a no-tension joint. This description used here is the classical
 138 idealized interaction between two bodies. A more realistic one will be the

139 subject of further work. Indeed, it appears in Section 4 that the contacts can
 140 become flat. Considering this observation, a distributed contact stiffness or
 141 a surface (2D)/ volume (3D) stiffness would be more suitable.

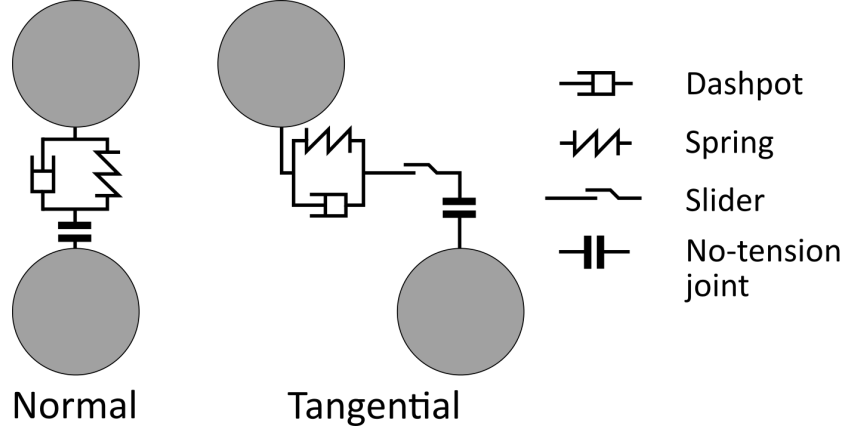


Figure 1: Model for the interaction between two particles.

142 The normal spring in the contact model is defined by Equation 2. It is
 143 worth noting that some equivalent parameters, such as the Young's modulus
 144 Y_{eq} and the radius R_{eq} , are used and depend on the grain properties.

$$k = \frac{4}{3}Y_{eq}\sqrt{R_{eq}}$$

where $\frac{1}{Y_{eq}} = \frac{1-\nu_i^2}{Y_i} + \frac{1-\nu_j^2}{Y_j}$ and $\frac{1}{R_{eq}} = \frac{1}{R_i} + \frac{1}{R_j}$ (2)

145 Here, k represents the stiffness of the normal spring, $Y_{i,j}$ is the Young's
 146 modulus of the grain i or j , $\nu_{i,j}$ is the Poisson's ratio of the grain i or j , and
 147 $R_{i,j}$ is the mean grain radius of the grain i or j .

148 To introduce energy dissipation and reach an equilibrium, a linear damp-
 149 ing term is added. The damping coefficient η is defined in Equation 3 based

150 on a restitution coefficient ($e = 1$ for perfect elastic contact, $e = 0$ for com-
151 plete energy dissipation).

$$\eta = 2 \times \gamma \sqrt{m_{eq} \times k}$$

where $\gamma = -\frac{\ln(e)}{\sqrt{\pi^2 + \ln(e)^2}}$ and $m_{eq} = \frac{m_i \times m_j}{m_i + m_j}$ (3)

152 Here, η denotes the damping coefficient, $m_{i,j}$ is the mass of the grain i or
153 j , k is the stiffness of the normal spring, and e is the restitution coefficient.

154 Next, the Hertz theory is applied in Equation 4 to obtain the normal
155 force [43, 44, 45]. A 3D formulation has been applied as the PFDEM aims
156 to be applied for 3D geometries. It is important to note that this mechanical
157 response is nonlinear with respect to the overlap. The stiffness increases as
158 the overlap becomes larger. The damping term is also included.

$$F_n = F s_n + F d_n = -k \times \delta_n^{3/2} - \eta \times (\vec{v}_i - \vec{v}_j) \cdot \vec{n} \quad (4)$$

159 Here, $\vec{v}_{i,j}$ is the speed of the grain i or j , and $\vec{n}$ is the normal vector of
160 the contact.

161 Concerning tangential behavior, a tangential overlap vector $\vec{\delta}_t$ is com-
162 puted incrementally by $\Delta \vec{\delta}_t$, defined Equation 5.

$$\Delta \vec{\delta}_t = [\vec{v}_{ij} - \vec{n}(\vec{n} \cdot \vec{v}_{ij})] \times dt$$

where $\vec{v}_{ij} = (\vec{v}_i - \vec{v}_j) + R_i \vec{n} \times \vec{\omega}_i + R_j \vec{n} \times \vec{\omega}_j$ (5)

163 Here, $\vec{v}_{i,j}$ is the grain linear velocity vector, $\vec{\omega}_{i,j}$ is the grain angular
164 velocity vector, dt is the time step.

165 It is important to notice that the orientation of the tangential vector can
 166 evolve with time. An update of this vector is done at each step, see Equation
 167 6.

$$\vec{\delta}_t = \vec{\delta}_t' - \vec{n}(\vec{n} \cdot \vec{\delta}_t') \quad (6)$$

168 Here, $\vec{\delta}_t$ is the tangential displacement updated and $\vec{\delta}_t'$ is the tangential
 169 displacement not updated.

170 The tangential spring in the contact model is defined by Equation 7.
 171 Similar to the normal spring, it uses equivalent parameters (shear modulus
 172 G_{eq} and radius R_{eq}) that depend on the grain properties. Additionally, this
 173 parameter is obtained using the Mindlin and Deresiewicz theory [44, 45, 46].
 174 The tangential stiffness depends on the normal overlap δ_n , with a stiffer spring
 175 for larger overlaps. Moreover, it is nonlinear, depending on the value of the
 176 norm of the tangential overlap $\overline{\delta}_t$, with a softer spring for larger tangential
 177 overlaps.

$$\begin{aligned} k_t &= k_{t0} \sqrt{1 - \frac{2 \times k_{t0} \times \overline{\delta}_t}{3 \times \mu \times F s_n}} \\ \text{where } k_{t0} &= 8 \times G_{eq} \sqrt{R_{eq} \times \delta_n} \\ \text{and } \frac{1}{G_{eq}} &= \frac{1-\nu_i}{G_i} + \frac{1-\nu_j}{G_j} \text{ and } G_i = \frac{Y_i}{2(1+\nu_i)} \end{aligned} \quad (7)$$

178 Here, k_t represents the stiffness of the tangential spring, μ is the friction
 179 coefficient, and $G_{i,j}$ is the shear modulus of the grain i or j .

180 Similar to the normal contact, a linear damping term is added for energy
 181 dissipation, and the damping coefficient is defined in Equation 8.

$$\eta_t = 2 \times \gamma \sqrt{m_{eq} \times k_t} \quad (8)$$

182 To obtain the tangential force vector $\overrightarrow{Fs_t}$, the Coulomb criterion is ap-
 183 plied, allowing sliding between grains.

$$\begin{aligned} \overrightarrow{Fs_t} &= -k_t \times \overrightarrow{\delta_t} \\ \overline{Fs_t} &\leq \mu F s_n \end{aligned} \quad (9)$$

184 The total tangential force is the sum of the spring term and the damping
 185 term, as explained in Equation 10.

$$\overrightarrow{F_t} = \overrightarrow{Fs_t} + \overrightarrow{Fd_t} = -k_t \times \overrightarrow{\delta_t} - \eta_t \times (\overrightarrow{v_i} - \overrightarrow{v_j}) \cdot \overrightarrow{t} \quad (10)$$

186 Note that the equations for the Discrete Element Method outlined in this
 187 Section and used in the present paper are frequently applied in the literature
 188 for geomaterials [15]. Multiple adaptations and expansions of this model
 189 exist, tailored to the specific applications targeted or couplings considered
 190 [47] and could be easily added to the present framework.

191 *Influence of grain discretization*

192 The discretization of the grain (= the number of vertices) is the main issue
 193 for the quality of the result for polyedral particles. Hence, if the discretization
 194 is coarse, the shape is not well described and some errors can occur for contact
 195 computation. The goal of this paragraph is to study the influence of this
 196 parameter on the quality of the results.

197 First, two simple cases illustrated in Figure 2 are considered: two grains
 198 in contact and one grain in contact with a wall. Those cases represent the
 elementary phenomena in a granular sample.

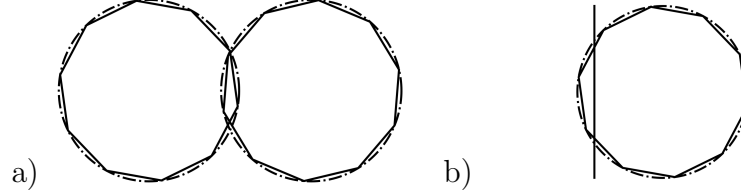


Figure 2: Two cases are considered: a) a contact grain-grain and b) a contact grain-wall. The dashed lines represent perfectly circular grains.

199
 200 Different grains with 10 to 60 (with a step of 5) vertices are generated
 201 to see the influence of grain discretization. Then, the grains are randomly
 202 rotated and the overlap is estimated with the common-plane algorithm [27].
 203 The statistical result obtained with 100 repetitions is shown in Figure 3. It
 204 appears the variation of estimated overlap (see the size of the boxes) decreases
 205 with grain discretization.

206 This study is repeated for different real overlaps. Indeed, for coarse dis-
 207 cretization and low contact force, a theoretical contact cannot be detected
 208 because of the discretization. Figure 4 shows that grain discretization greatly
 209 influences the results' quality for small overlap, as is the case in the granular
 210 sample.

211 Secondly, a more complex case is considered: a granular assembly com-
 212 posed of 300 grains is loaded under oedometric conditions. To see the in-
 213 fluence of grain discretization, a sample composed of perfect disks is loaded.
 214 Then the grains are divided into 20, 30, 40 or 60 vertices. For each case,
 215 10 simulations are run. Figure 5 shows the evolution with time of the to-

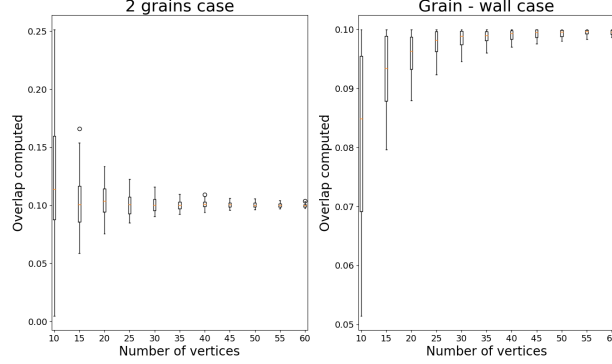


Figure 3: Influence of the grain discretization on the overlap estimation (the targetted value is 0.10).

tal kinetic energy, the coefficient $K_0 = \sigma_{II}/\sigma_I$, the upper wall position and the force applied on the upper wall for one run and different numbers of vertices. The number of vertices for one grain has a large influence on the global behavior of the sample. Indeed, with few vertices, a lot of fluctuation is introduced in the computing of the contacts. This fluctuation is reflected in the estimation of the coefficient K_0 . Moreover, the total kinetic energy stays at a large value with few vertices by grains. The equilibrium can not be reached.

A statistical analysis of the total kinetic energy and the estimation of the coefficient K_0 of all the runs is presented in Figure 6. For all runs, the difference between the third quartile and the first one is estimated. Then, a mean value is computed for the different numbers of vertices. It appears the fluctuation is decreasing with the quality of the discretization.

It appears the discretization of the grains is a main parameter to capture accurately the interactions at the particle level and to minimize the noise in

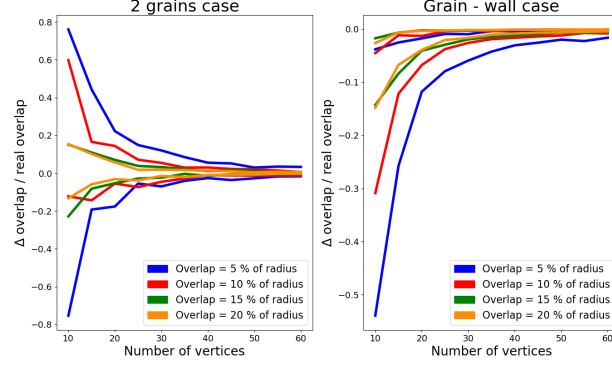


Figure 4: Influence of the grain discretization on the overlap estimation for different solicitations. Only the first and the third quartiles of each data set are plotted.

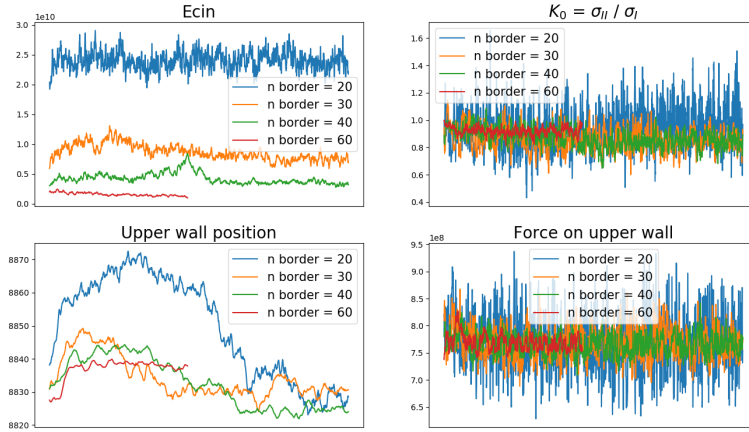


Figure 5: Evolution with time of the total kinetic energy, the coefficient $K_0 = \sigma_{II}/\sigma_I$, the upper wall position and the force applied on the upper wall for different numbers of vertices.

231 more complex granular media. At least 60 nodes are considered in this work
 232 to discretize the grains (80 nodes are considered for more complex shapes).

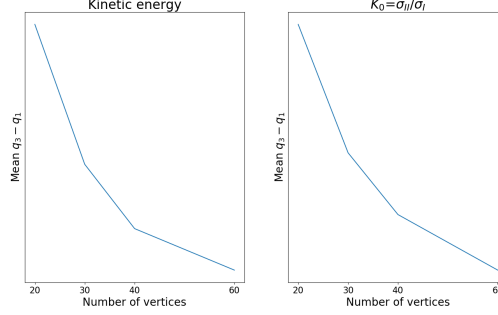


Figure 6: Statistical analysis about the fluctuation of the coefficient $K_0 = \sigma_{II}/\sigma_I$ for different numbers of vertices.

2.2. Phase-Field

A granular medium is composed of grains and pores. As illustrated in Figure 7, this porous matter is represented here in the phase-field space by a combination of phase variables (one phase per grain). This phase variable equals 1 if it is inside the grain associated and equals 0 elsewhere.

The phase-field method, specifically the Allen-Cahn equation described in Equation 11, is employed to describe the dissolution/precipitation of the material [33]. In this approach, nonconserved order parameters η_j are utilized to represent the phase transformation.

$$\frac{\partial \eta_j}{\partial t} = -L_j \left(\frac{\partial (f_{loc} + E_d)}{\partial \eta_j} - \kappa_j \nabla^2 \eta_j \right) \quad (11)$$

In this equation, the terms L_j and κ_j correspond to the order parameter mobility and the gradient energy coefficient, respectively. The order parameter mobility affects the overall dissolution/precipitation kinetic, while the gradient energy coefficient influences the interface width and reaction. The term E_d encompasses additional sources of energy introduced into the sys-

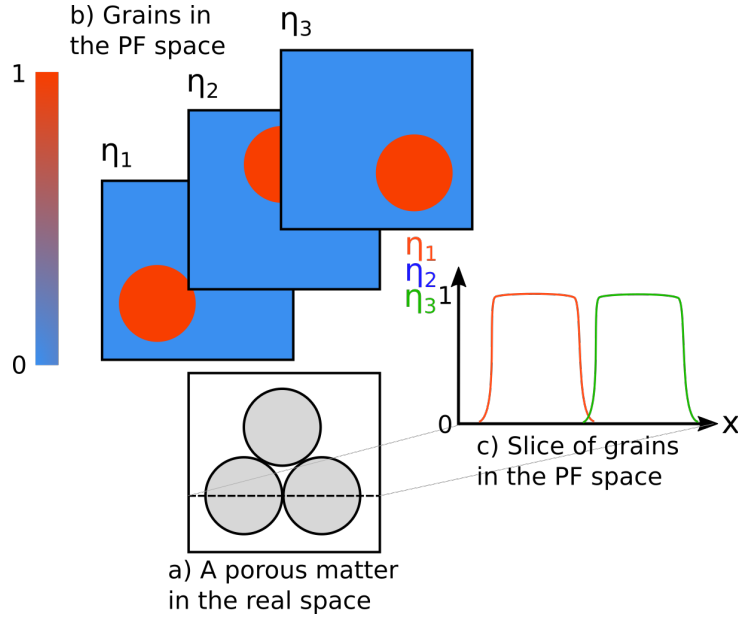


Figure 7: The phase-field method is applied to granular matter: a) a granular matter is composed of grains and by pore, b) one phase variable η_i is generated by grain, c) the phase variable is equal to 1 if it is inside the grain associated and equal to 0 elsewhere.

tem, such as mechanical, chemical, or thermal loading. The local free energy density f_{loc} is specified in Equation 12 and is visualized in Figure 8.

$$f_{loc} = 16 \times h \times \left(\sum_j \eta_j^2 (1 - \eta_j)^2 \right) \quad (12)$$

The local free energy density f_{loc} employs a double-well function with a barrier height h [48]. Notice that the minima of this potential energy are located at $\eta_j = 0$ and $\eta_j = 1$. At the equilibrium and without external destabilization, the phase variables stay at $\eta_j = 0$ or $\eta_j = 1$ (no dissolution/precipitation). Figure 8 illustrates the double-well function as a representation of the local free energy density f_{loc} and its derivative regarding the

255 phase variable η .

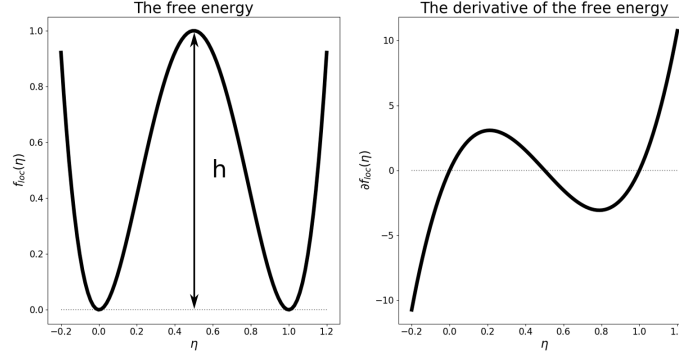


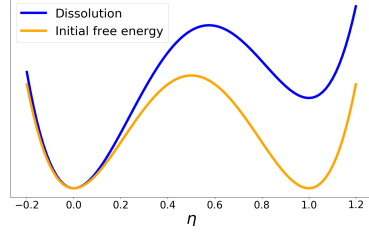
Figure 8: A double-well function as a local free energy density f_{loc} and its derivative $\frac{\partial f_{loc}}{\partial \eta}(\eta)$ (the energy barrier $h = 1$ in this example).

256 To obtain localized dissolution/precipitation, the idea is to add external
 257 source term E_d to tilt the initial free energy to favor dissolution/precipitation,
 258 as depicted in Figure 9. When the dissolution phenomenon dominates, the
 259 double-well function is tilted towards $\eta_j = 0$, causing the material to aim
 260 for dissolution. Conversely, when the precipitation phenomenon dominates,
 261 the double-well function is tilted towards $\eta_j = 1$, indicating the material's
 262 tendency to precipitate.

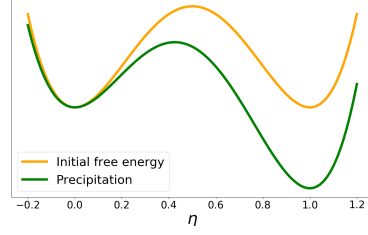
263 To model solute concentration in the pore fluid, a new variable, denoted
 264 as c , is introduced in the phase-field formulation. The Equation 11 is solved
 265 along with a new equation for the variable c , as given in Equation 13.

$$\frac{\partial c}{\partial t} = - \sum_j \left(\frac{\partial \eta_j}{\partial t} \right) + \kappa_c \nabla^2 c \quad (13)$$

266 The Equation 13 represents the conservation of solute mass and is solved
 267 using the variations of phase-field variables η_j as a source term. Here, κ_c



Dissolution destabilization



Precipitation destabilization

Figure 9: The double-well function is destabilized to favor dissolution/precipitation.

268 represents the gradient energy coefficient that controls the diffusion rate.
 269 Notably, the gradient energy coefficient κ_c is assumed to be 0 within the
 270 grains, so that the solute concentration c is nonzero outside the grains and
 271 diffuses from the contact area to the pore surface. The algorithm's imple-
 272 mentation for building the heterogeneous gradient energy coefficient κ_c is
 273 described in Appendix B. The terms $\frac{\partial c}{\partial t} = -\sum_j \frac{\partial \eta_j}{\partial t}$ in Equation 13 ensure
 274 the conservation of mass. When a grain (η_j) dissolves, solute concentration
 275 (c) is generated, and vice versa, when η_j is generated through precipitation,
 276 some solute disappears.

277 Equations 11 and 13 are solved with the open-source finite element soft-
 278 ware MOOSE [49]. This software uses automatic differentiation to solve the
 279 system of equations with an implicit formulation.

280 A special focus is placed on the mesh dependency of the problem. Indeed
 281 the mesh resolution influences the resolution of the Phase-Field equations
 282 and the discretization of the grain, see Section 2.1.

283 The mesh should be fine enough to obtain accurate results for solving the

284 Phase-Field equations. The critical element is the interface thickness. In the
 285 literature, the criteria $w \sim 4\Delta x - 8\Delta x$ and $\Delta x \leq R/5$ are advised [48, 50]
 286 where w is the interface width, Δx is the mesh size and R is the grain radius.
 287 Combining the two criteria, a third one is obtained: $w \leq 4R/5$. In this
 288 study, a finer interface has been selected ($w \sim R/10 \iff \Delta x \sim R/40$) to be
 289 sure the mesh has no influence on the numerical results and to have a fine
 290 interface compared to the grain size [50].

291 Concerning the discretization of the grain, the mesh limits the maximal
 292 number of vertices defining a grain, see the algorithm presented in Section
 293 2.3. Considering the mesh size described earlier in this section ($\Delta x \sim R/40$)
 294 it appears that the maximum discretization is ≈ 160 nodes (2 vertices per
 295 slice). This value is much larger than the discretization considered in Section
 296 2.1.

297 To summarize, assuming a fine interface compared to the size of the grain
 298 induces mesh independence of the numerical results.

299 *2.3. PF-DEM Couplings*

300 The data exchange and global scheme of the PF-DEM couplings are il-
 301 lustrated in Figure 10 and in Algorithm 1, respectively.

302 In the phase-field simulation, the sample geometry is discretized into a
 303 mesh, and the grains are represented by phase variable maps. A grain de-
 304 tection algorithm is applied to determine the new grain shape and solute
 305 configuration based on the phase-field outputs. On the other hand, in the
 306 discrete element modelization, grains are represented as a collection of ver-
 307 tices. This model aims to compute the new positions of the grains and the

308 mechanical energy at each contact.

309 It is important to note that although PF and DEM operate on different
 310 time scales, they are still connected. The mechanical equilibrium (DEM) is
 311 assumed to be instantaneous, while grain dissolution or precipitation occurs
 312 over longer time periods, typically several hours to days.

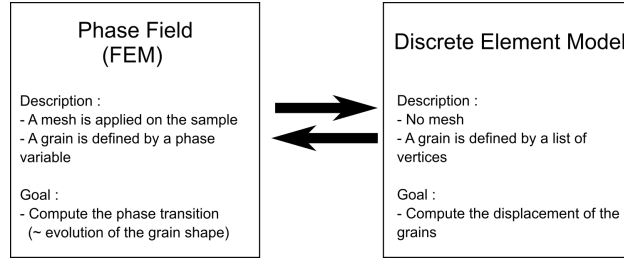


Figure 10: Data exchange between the phase-field simulation and discrete element modelization.

313 After a phase-field step, the system does not immediately reach a mechan-
 314 ical equilibrium due to ongoing dissolution or precipitation. Therefore, the
 315 new grain boundaries need to be detected. Figure 11 illustrates the division
 316 of the phase-field into multiple layers (along the different axes), assuming the
 317 grain boundary occurs at $\eta_j = 0.5$ and using a linear approximation.

318 A polygonal particle is obtained based on the vertex positions. To reduce
 319 computational cost, only a certain number of vertices are used. As explained
 320 in Figure 12a, all nodes are considered initially. Then, the perimeter of the
 321 polygon is computed by summing the distance of the nearest nodes all over
 322 the grain. An average node distance is computed considering the perimeter
 323 and the grain discretization $d_{min}^{dis} = Perimeter / n_{discretization}$. Finally, an
 324 iterative step is used, considering a random starting vertex. As illustrated
 325 in Figure 12b, the vertices list is read until the distance between the two

Algorithm 1 Global scheme of the simulation. Two time scales are used: one for DEM and one for PF.

```

while work not finished do
    Build grains from PF data
    Update geometric parameters
    while not DEM equilibrium do                                ▷ Start the DEM simulation
        Compute solicitations
        Solve the momentum balances
        Check the equilibrium criteria for DEM
    end while
    Recreate the maps of phase variable
    Solve PF equations                                          ▷ Start the PF simulation
end while

```

326 vertices (starting and the list's ones) is at least the average node distance
327 d_{min}^{dis} . The iteration keeps going, considering the list's vertex as the new
328 starting one. This method helps to minimize the number of nodes and to
329 have a homogeneous discretization of the grain.

330 The discretization of the grain boundary is crucial, and the quality of
331 overlap estimation depends strongly on this parameter, as explained in Sec-
332 tion 2.1. The geometric properties of the grain, such as its center, surface
333 area, and inertia, can be computed from the list of vertices using a Monte
334 Carlo method [39] described in Appendix A.

335 Once the grain boundaries are known, a series of DEM iterations takes
336 place until the system reaches an equilibrium. To dissipate mechanical en-
337 ergy, some damping is introduced, as explained earlier in this section. The

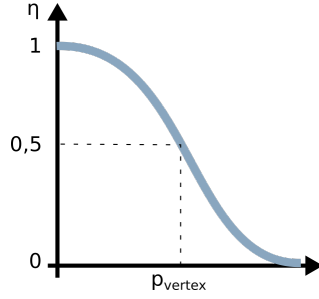


Figure 11: The phase field is divided into layers, and the grain boundary is assumed to occur at $\eta_j = 0.5$.

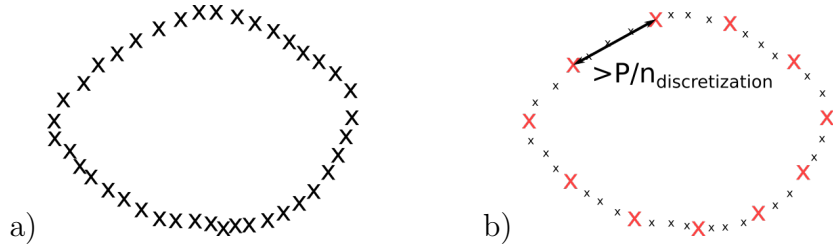


Figure 12: Even if a) the grain is described as a list of vertices, b) only a part of this list is considered to save computational cost.

338 convergence criteria for detecting the equilibrium may vary depending on the
 339 problem. In this work, the equilibrium is detected when the three following
 340 criteria are verified :

- 341 • the sum of the kinetic energy over the grains is smaller than a threshold
 342 value.
- 343 • the confining pressure applied is in the window defined by 0.95 and
 344 1.05 the confining pressure targetted.
- 345 • the position of the top wall (controlled to apply the load) is stable. The
 346 variance of the latest positions is below a threshold value.

Moreover, a maximum number of iterations is set to avoid an infinite loop. Once the equilibrium is reached before the maximum number of iterations, new phase maps are constructed to update the geometry of the phase field problem based on the results of the DEM step.

Two methods have been designed to construct the new phase maps: the Deconstruction-Rebuild and the Interpolation algorithms. A comparison of their features is shown in Table 1.

	DR	Int
Assume the interface width w constant	x	
Generate a new mesh at each PF simulation	x	
Conservation of the information between PF simulations		x
Can track the solute concentration c		x

Table 1: Comparison of the Deconstruction-Rebuild (DR) and the Interpolation (Int) algorithms to build phase map after a DEM step.

In the simulations presented in Section 3, a Deconstruction-Rebuild algorithm is employed, assuming that the interface width (w) remains constant throughout the study. Indeed, the solute concentration does not need to be tracked as it is assumed immediately evacuated [17, 51]. Moreover, this algorithm is faster than the Interpolation one. Given the knowledge of the center and vertices of a grain (the Deconstruction part), a distribution of the radius R following the angle can be computed (the choice of the angle origin is arbitrary; for this study, the +x-axis is considered). Subsequently, the new phase variable η_j is calculated using a cosine profile described by Equation 14 for all nodes of the mesh (the Rebuild part).

$$\eta_j = \begin{cases} 1 & \text{for } r \leq R - \frac{w}{2} \\ 0.5 \times \left(1 + \cos\left(\pi \frac{r - R + \frac{w}{2}}{w}\right)\right) & \text{for } R - \frac{w}{2} \leq r \leq R + \frac{w}{2} \\ 0 & \text{for } r \geq R + \frac{w}{2} \end{cases} \quad (14)$$

364 Here, r represents the distance between the node and the center, R is the
 365 radius in the considered direction, and w denotes the interface width.

366 In the simulations presented in Section 4, an Interpolation algorithm is
 367 used. This algorithm considers that the interface width (w) is not constant
 368 during the study, particularly increasing in the contact zone during pressure
 369 solution modelling. Moreover, it becomes important to track the solute con-
 370 centration c . The new phase map is obtained through interpolation from the
 371 previous one. As illustrated in Figure 13, at each iteration, the next mesh
 372 is computed based on the previous mesh by applying an inverse rigid body
 373 motion. Since the two meshes often do not perfectly match, bilinear (2D) or
 374 trilinear (3D) interpolation is performed to estimate the values of the next
 375 mesh.

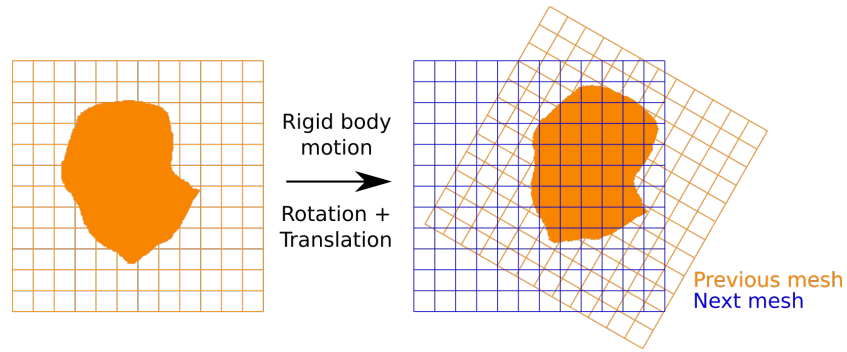


Figure 13: Scheme of the interpolation algorithm. The next mesh is computed from the previous mesh by applying an inverse rigid body motion.

376 The deformation induced by the phase-field simulation and the grain dis-
 377 placements computed in the discrete element modelization are closely in-
 378 terconnected. It is important to ensure that the deformation remains in
 379 the small-strain range. For fluid-structure interaction problems, an Aitken
 380 method [52] is commonly used to adapt the deformation of the structure.
 381 Here, the deformation of the grain during a phase-field simulation can be
 382 controlled with an adaptive time step (relative to shape evolutions). A clas-
 383 sical threshold values system is used in this problem. The duration of the
 384 simulation, and consequently the grain deformation during the phase-field
 385 simulation, depends on the mean absolute value per contact node of the to-
 386 tal energy E_d . This parameter is related to the deformation known in the
 387 phase-field simulation. The importance of this method is illustrated in Figure
 388 14 where a lateral displacement for 2 grains pressure-solution configuration,
 389 see Section 4, is plotted without the inspired Aitken method. Oscillations
 390 appear, revealing large deformation in the Phase-Field simulations. At step
 391 i , a large amount of grain dissolves, a lot of solute is available, and the grains
 392 get closer. At step $i + 1$, the solute available generates a large amount of
 393 grain precipitation, and the grains get further. The inspired Aitken method
 394 allows the use of an efficient Phase-Field simulation duration, verifying a
 395 small deformation criterion.

396 By initializing the sum of the absolute total energy as $E_{d,abs}$ and the
 397 number of contact nodes as *node_contact*, the mesh is scanned. If a contact
 398 zone is detected (where η_i and η_j are both larger than 0.5), the absolute value
 399 of the total energy E_d is added to the indicator $E_{d,abs}$, and the number of
 400 contact nodes is incremented. The mean absolute value per contact node of

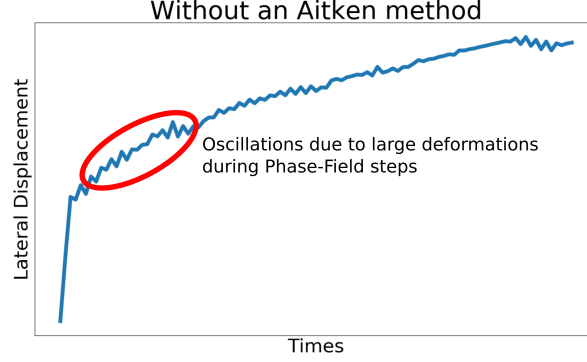


Figure 14: Lateral displacement for a 2 grains pressure-solution configuration, see Section 4, without an inspired Aitken method. Oscillations appear because of the large deformation in the Phase-Field simulations.

401 the total energy $\overline{E_{d,abs}}$ is then computed = $E_{d,abs}/node_contact$. This mean
 402 parameter plays the role of the discriminant value and depending on the
 403 threshold values, a time step dt is determined for the phase-field simulation.

404 3. Application to Irregular Shapes and homogeneous dissolution

405 This Section aims to demonstrate the capability of the PFDEM to handle
 406 irregular grain shapes. The simulations conducted here are intended to be
 407 compared with previous experiments and numerical simulations conducted by
 408 Shin et Santamarina and Cha et Santamarina [51, 53]. Figure 15 illustrates
 409 the setup where a sample is subjected to a constant stress in oedometric
 410 conditions while acid is injected. The granular material consists of two types
 411 of grains: dissolvable and undissolvable.

412 The distribution between dissolvable and undissolvable grains is deter-
 413 mined by the ratio of the total surface area of dissolvable grains to the total

414 surface area of all grains. As the acid is injected, it tends to reduce the size of
 415 the dissolvable grains. It has been observed that the coefficient $K_0 = \sigma_{II}/\sigma_I$
 416 evolves with the dissolution process due to internal reorganization around
 417 the dissolvable grains.

418 The simulation campaign parameters are provided in Table 2. The mean
 419 behavior is obtained by extrapolating results from three simulations for each
 420 configuration.

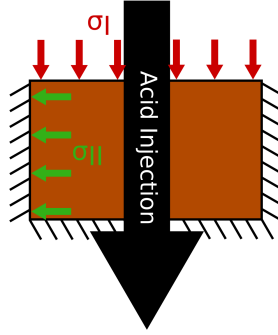


Figure 15: Scheme of the sample under oedometric condition and acid injection.

Parameters	Unit	Value
Number of grains	-	300
Particle Size Distribution for undissolvable grains (radius and percentage of the number of undissolvable grains)	μm (%)	420 (17), 385 (33), 315 (33), 280 (17)
Percentage of grain dissolvable (= $S_{dissolvable}/(S_{dissolvable} + S_{undissolvable})$)	%	5 or 15

Particle Size Distribution for disks as dissolvable grains (radius and percentage of the number of dissolvable grains)	$\mu m (\%)$	360 (17), 330 (33), 270 (33), 240 (17)
Particle Size Distribution for squares as dissolvable grains (dimension and percentage of the number of dissolvable grains)	$\mu m (\%)$	500 (17), 460 (33), 380 (33), 440 (17)
Young modulus	GPa	70
Poisson's ratio	-	0.3
Density	kg/m^3	2500
Friction grain-grain	-	0.5
Friction grain-wall	-	0
Restitution coefficient	-	0.2
Mobility for phase-field	-	1
Gradient coefficient for phase-field	-	3
Time step for discrete element model	sec	$\Delta t_{crit}/8$
Linear force applied on the upper wall	N/m	$4 \times Y R_{mean}/2000$
Percentage of the mean dimension dissolved between each iteration	%	0.5

Table 2: Parameters used for the simulation campaign, concerning the acid injection inside a sample under oedometric condition.

421 In the simulations presented in this study, the undissolvable grains are
422 always assumed to be disks, while the dissolvable grains can be either disks
423 or squares (although other shapes are possible, the simulation is restricted

to these two). The main objective of this Section is to discuss the influence of grain shape on overall behavior. The latter choice is only possible in the present framework and represents more accurately the experimental setup as the undissolvable grains are glass beads and the dissolvable ones are salt grains with a cubic shape. The size of the grains is defined as shown in Figure 16, where a disk is characterized by its radius and a square is determined by the length of one side.

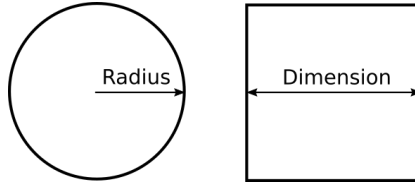


Figure 16: Definition of the characteristic length for a disk and a square.

The choice of grain shape affects certain material properties, such as surface density. For disk particles, the surface density is given by $\sigma_m = \frac{4}{3}\rho R_{\text{mean}}$, while for square particles, it is $\sigma_m = \rho L_{\text{mean}}$, where R_{mean} is the mean radius, L_{mean} is the mean dimension, and ρ is the density.

The time step for the DEM is computed based on the critical time step defined for a sphere using the Hertzian contact model, as shown in Equation 15 [54]:

$$\Delta t_{\text{crit}} = \frac{\pi L_{\text{min}}}{0.1631\nu + 0.88} \sqrt{\frac{\rho}{G}} \quad (15)$$

Here, L_{min} represents the minimal characteristic length for the grains (half of the dimension for squares), ν is the Poisson's ratio, ρ is the density, $G = \frac{Y}{2(1+\nu)}$ is the shear modulus, and Y is the Young's modulus.

441 A linear vertical force is applied to the upper wall to confine the sample.
 442 The intensity of this force is proportional to the normal stiffness, considering
 443 an isotropic 3D material model with $k_n = 4 \times Y \times R_{\text{mean}}$ [55], where Y is the
 444 Young’s modulus and R_{mean} is the mean radius of the undissolvable grains.

445 To establish the initial configuration, perfect disks are assumed (using
 446 the circumscribed circle for non-disk grains). These disks are randomly gen-
 447 erated without overlap within the sample and then loaded. Once an initial
 448 equilibrium is reached, the disks are discretized, and their shapes are main-
 449 tained. Another loading step is performed to reach a second equilibrium. A
 450 sample geometry with a height-to-diameter ratio of 0.6 is chosen to mini-
 451 mize boundary effects at the top and bottom [51], and the sample generation
 452 process ensures this geometry is achieved.

453 *3.1. Influence of the number of grains*

454 The definition of the particle size distribution depends on the specific case
 455 and is presented in Table 2. The total number of grains has been chosen to
 456 minimize computational cost while respecting the assumption of a represen-
 457 tative element volume, as determined by a preliminary study presented in
 458 the following.

459 Circular particles are used for this analysis on the representative number
 460 of particles to optimize the computational cost. Parameters from Table 2 are
 461 used to define the material constitutive laws, the DEM time step, and the
 462 particle’s geometry. As described by Table 3, the number of grains varies
 463 between 50 and 700. The table also describes the number of simulations
 464 run for each number of grains (between 5 and 10 times). 15% of the grains
 465 selected randomly are assumed as dissolvable. After equilibrium is found,

the radius of the dissolvable particles decreases by 1% of the initial mean radius ($350 \mu m$).

Number of grains	50	100	200	300	400	500	700
Number of runs	10	10	10	9	10	5	5

Table 3: Different samples are tried with different numbers of grains. Each configuration is run several times to make a statistical study of the results.

The results are presented in Figures 17, 18 and 19. A statistical study is done on the evolution of the $K_0 = \sigma_{II}/\sigma_I$ with dissolved material quantity to determine the representative equivalent volume.

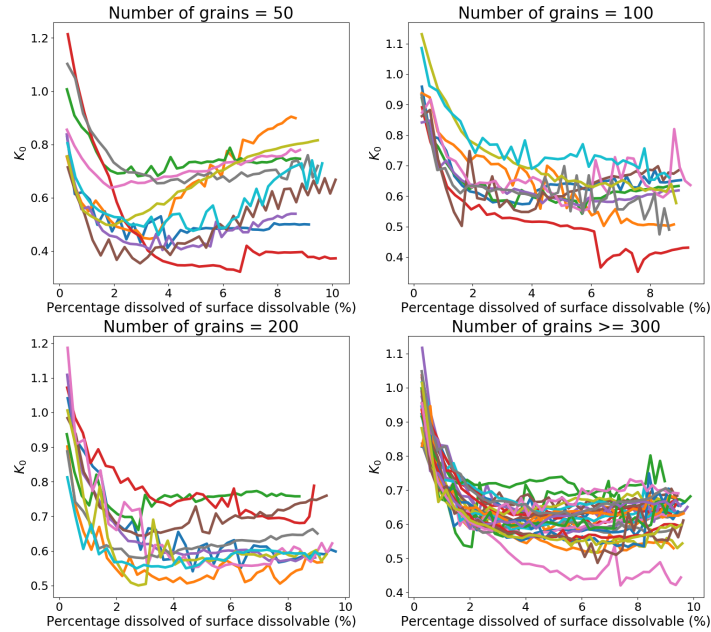


Figure 17: All curves of the preliminary campaign sorted following the number of grains in the sample.

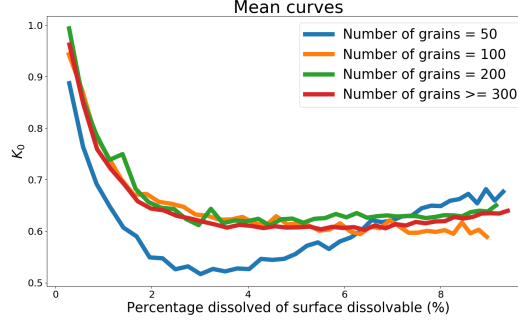


Figure 18: Mean curves for different numbers of grains in the sample.

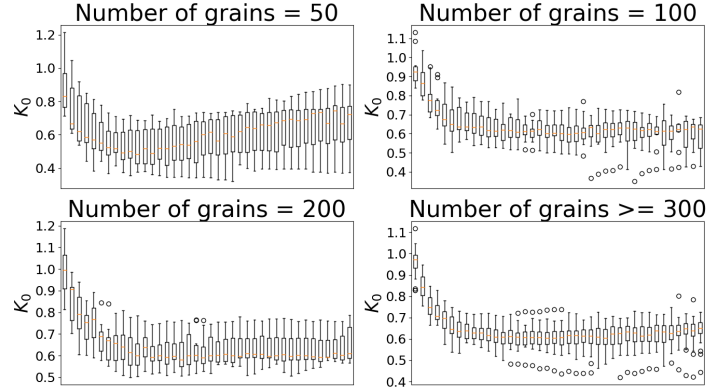


Figure 19: Statistical study to justify that a sample with 200 grains is a representative equivalent volume.

471 We can observe that the results obtained with only 50 grains are scattered
 472 and therefore not representative. Even if Figure 18 shows that mean curves
 473 are the same for samples with 100, 200 or more than 300 grains, Figure 19
 474 highlights the large variance for the case of 100 grains. The representative
 475 equivalent volume can be assumed with 200 grains as the variance is much
 476 lower. As the sample is a granular material, the results are always noisy. Even

477 if the sample is with a representative element volume, several simulations at
 478 the same configuration should be done to work with a mean evolution.

479 *3.2. Influence of the step in the grain dissolution*

480 In this case, the external source term E_d in the phase-field formulation
 481 (as described in Equation 11) models the acid. It is assumed that the solute
 482 quantity generated by dissolution is instantaneously evacuated (the diffusive-
 483 conservative Equation 13 is not solved and the solute concentration c is not
 484 tracked). The formulation of this source term is explained in Equation 16. It
 485 should be noted that this term is applied only to dissolvable grains, as there is
 486 no interaction between grains during the phase-field simulation. Therefore, it
 487 is possible to extract dissolvable grains individually for separate simulations,
 488 and undissolvable grains are not included in the phase-field simulation as
 489 they remain unchanged.

$$E_d = e_{\text{diss}} \times \left(\sum_j \eta_j^2 (3 - 2\eta_j) \right) \quad (16)$$

490 Here, e_{diss} represents the energy of dissolution, and η_j is a phase variable
 491 that models a dissolvable grain.

492 The amount of material dissolved during each phase-field simulation step
 493 depends on the grain size, the energy of dissolution e_{diss} , and the duration of
 494 the simulation. It is important to ensure proper discretization between DEM
 495 steps. In this regard, the DEM step models mechanical equilibrium, while
 496 the PF step models dissolution as the source of perturbation. Mechanical
 497 equilibrium is assumed to be instantaneous, and the amount of material
 498 dissolved between DEM simulations must be sufficiently small. A simulation

campaign presented in the following was conducted to verify this criterion.

To have faster results, perfect circle particles are used. Parameters from Table 2 are used to define the material, the discrete element model time step, and the particle's geometry. According to the preliminary work about the representative equivalent volume presented in this Section, the number of grains is 300. 15% of the grains selected randomly are assumed as dissolvable. As described by Table 4, the radius of the dissolvable particles is decreasing by 0.2 %, 0.5 %, 1.0 % or 2.0 % of the initial mean radius ($350\ \mu m$). The table also describes the number of simulations run for each radius reduction (between 8 and 15 times). The radius reduction between iterations defines the representative dissolution.

Radius reduction (% of the initial mean radius)	0.2	0.3	0.5	0.6	0.7	1.0	2.0	5.0
Number of runs	21	9	13	9	9	13	9	5

Table 4: Different radius reductions between iterations are tried. Each configuration is run several times to make a statistical study of the results.

The results are presented in Figures 20, 21 and 22. A statistical study is done on the evolution of the $K_0 = \sigma_{II}/\sigma_I$ with dissolved material quantity to determine the representative dissolution.

The results obtained with a radius reduction equivalent to 2.0 % of the initial mean radius are dispersed and thus not representative. Moreover, the steps are too large to describe the initial fast decrease of K_0 . Figures 21 and 22 show the curves obtained with a radius reduction lower than 0.5 % of the initial mean radius are reproducible and the initial fast decrease is

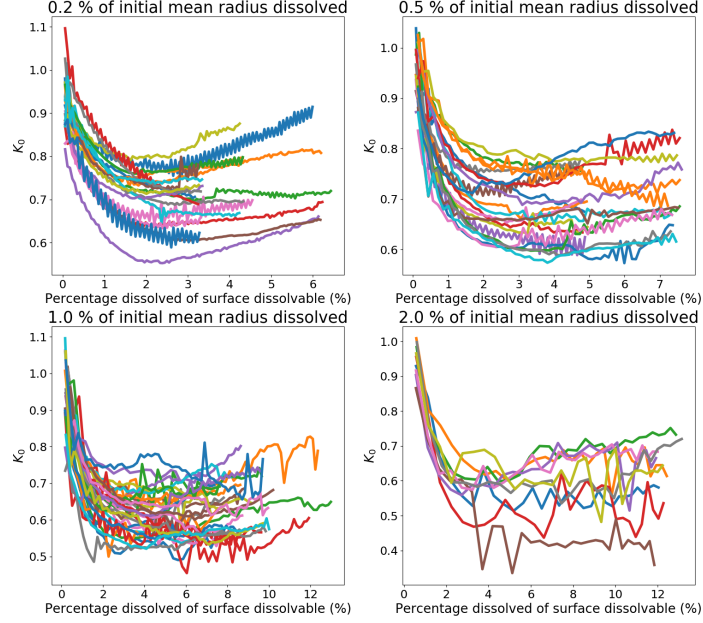


Figure 20: All curves of the preliminary campaign sorted following radius reduction.

well captured by those configurations. The representative dissolution can be assumed with a radius reduction equivalent to 0.5% of the initial mean radius. Notice that in this study no inspired Aitken method is needed. Indeed, the deformation of the grains aims to stay constant during the phase-field simulation. As the sample is a granular material, the results are noisy. Even if the sample is with a representative dissolution, several simulations at the same configuration should be done to work with a mean evolution.

3.3. Influence of the particle shape

The results of the simulation campaign are shown in Figures 23 and 24. Additionally, four example movies are linked to this article, demonstrating

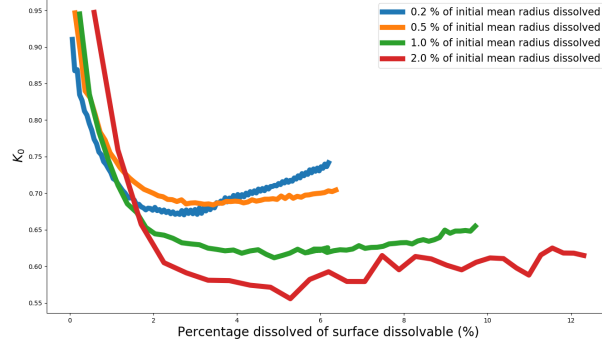


Figure 21: Mean curves for different radius reduction.

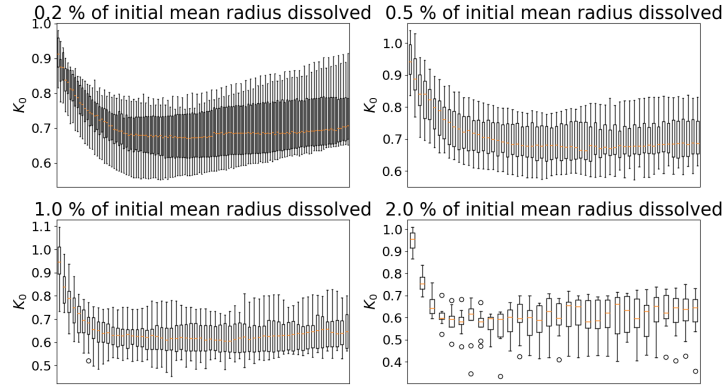


Figure 22: Statistical study to justify that 0.5 % of the initial mean radius dissolved at each iteration is a representative dissolution.

528 the evolution of grain configuration and chain forces for both disk-shaped
 529 and square-shaped dissolvable grains with a dissolvable surface ratio of 0.15
 530 relative to the total surface.

531 Figure 23 illustrates the mean evolution of K_0 (top), compacity (left), and
 532 vertical strain (right) with the dissolution of dissolvable material, consider-

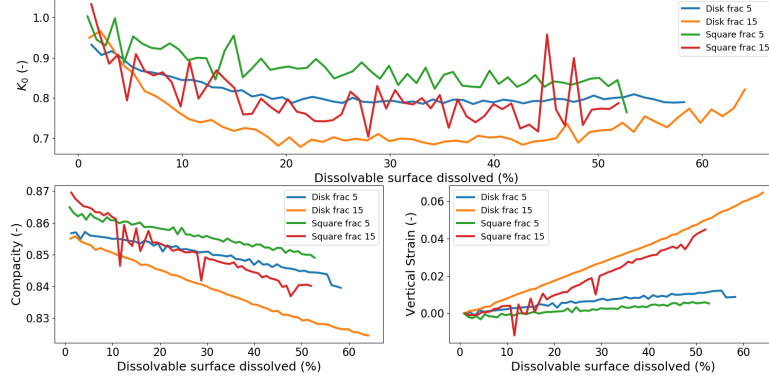


Figure 23: Mean evolution of K_0 (top), of the compacity (left) and the vertical strain (right) with the dissolution of the dissolvable material, considering dissolvable grains as disks or as squares. Two ratios of the dissolvable surface over the total surface are studied.

ing dissolvable grains as disks or squares. Two different ratios of dissolvable
 surface to total surface are studied. As highlighted by Shin et Santamarina
 and Cha et Santamarina [51, 53] and observed in Figure 23, the dissolution of
 grains leads to an evolution (increase or decrease) of the coefficient $K_0 = \frac{\sigma_{II}}{\sigma_I}$.
 In this paper, a stronger reduction in K_0 is observed when the percentage of
 dissolvable grains is larger. This reduction occurs due to the reorganization
 of grains around the dissolved grains, as shown in Figure 24. The num-
 ber of undissolvable-dissolvable contacts decreases with material dissolution,
 while the number of undissolvable-undissolvable contacts remains relatively
 constant.

It should be noted that the simulations by Shin et Santamarina and Cha et
 Santamarina constrained particle rotation to account for grain angularity and
 interlocking, while in our simulations, particle rotation remains free. Figure

Total contact distribution - % dissolvable surface dissolved

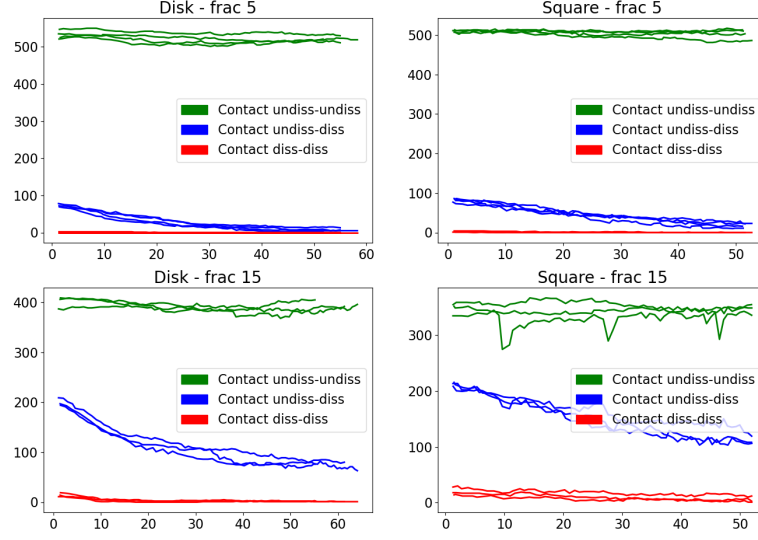


Figure 24: Evolutions of contact distribution with the dissolution of the dissolvable material, considering dissolvable grains as disks or as squares. Two ratios of the dissolvable surface over the total surface are studied.

3 from Cha et Santamarina [53] highlights that hindering the grains rotation
tends to decrease the coefficient K_0 . Hindering particle rotation is often
used as a proxy for grains angularity [56], so their simulations results tend
to show that more angular grains would have a larger decrease of K_0 , which
is the opposite that what we observe on 23. This highlights the importance
of considering explicitly the grain shape to obtain an accurate response of
the system. A parametric study would be relevant to better understand the
influence of confinement, particle size distribution, rolling model, and initial
compressibility to better understand in which conditions a rolling model can
be used as a proxy for grains angularity when chemo-mechanical couplings

556 are involved.

557 Furthermore, simulations with square dissolvable grains exhibit more fluc-
558 tuation, even though the grain discretization was increased to 80 vertices for
559 these simulations. This phenomenon is common in simulation with poly-
560 gons [20, 57, 58] and it is explained due to the applied moments on square
561 particles, arising from the arm moment and the force at the contacts [59].
562 With disk particles, the forces at the contacts are more often directed to-
563 wards the center of the grains, resulting in smaller applied moments. Figure
564 24 also reveals that the number of undissolvable-undissolvable contacts is
565 smaller for square grains compared to disk grains. While this type of contact
566 does not introduce fluctuation (as it involves two disk grains), the contact
567 undissolvable-dissolvable becomes more significant as it includes a square-
568 shaped grain. The difference in contact numbers arises from the fact that
569 there are more dissolvable grains in the case of squares than disks. Conse-
570 quently, the percentage of dissolvable grains is defined as the ratio of dis-
571 solvable surface area ($S_{\text{dissolvable}}$) to the sum of dissolvable and undissolvable
572 surface areas ($S_{\text{dissolvable}} + S_{\text{undissolvable}}$), and the square-shaped grain has a
573 smaller surface area than the disk-shaped grain. For a percentage of 15%,
574 a simulation with 300 grains contains 58 grains (in the case of disks) or 83
575 grains (in the case of squares).

576 Figure 24 demonstrates that the number of undissolvable-dissolvable con-
577 tacts evolves differently depending on the shape of the dissolvable grains,
578 especially when the dissolvable surface ratio is 0.15. The number of contacts
579 for disks decreases in two steps: a sharp slope (around 5 contacts lost per
580 percentage) from 0% to 15% dissolvable surface dissolved, followed by a slight

581 slope (around 1 contact lost per percentage) from 15% to 60% dissolvable
 582 surface dissolved. On the other hand, the number of contacts for squares
 583 decreases in a single step, with a slight slope (around 1-2 contacts lost per
 584 percentage). It is worth mentioning that the initial configuration algorithm
 585 assumes perfect disks. Once the first equilibrium is reached, the shapes of the
 586 particles are generated, and the circumscribed disks are replaced by disks or
 587 squares. In the case of squares, this operation suddenly reduces the number
 588 of undissolvable-dissolvable contacts. When dissolution begins in the case of
 589 disks, many contacts with small overlaps are deleted in the initial iterations.
 590 In contrast, these contacts with small overlaps are already deleted during the
 591 initial configuration generation for squares.

592 Figure 23 illustrates that the evolution of compacity $\left(= \frac{S_{\text{grains}}}{S_{\text{box}}}\right)$ and ver-
 593 tical strain seems to depend solely on the percentage of dissolvable grains.
 594 The slope is sharper when the percentage of dissolvable grains is larger. For a
 595 given percentage, regardless of the shape (disk or square), the same amount
 596 of material is dissolved at each iteration.

597 In conclusion, this Section emphasizes the ability of PFDEM to handle
 598 irregular grains. The shape of the grains has a significant influence on granu-
 599 lar behavior, and considering the true shape instead of perfect disks/spheres
 600 is important.

601 4. Application to Irregular Shapes, heterogeneous dissolution and 602 precipitation

603 In the context of PFDEM simulations, the heterogeneous dissolution/precipitation
604 process can be modeled. This Section aims to demonstrate the capability of
605 PFDEM to simulate such processes. In particular, we consider the exam-
606 ple of the pressure solution phenomenon that involves both processes at the
607 microscale and compare the simulation results with experimental findings
608 reported by De Meer et al. [60].

609 The pressure solution phenomenon arises from the premise that in densely
610 packed granular materials intergranular forces are substantial and this mech-
611 anism is dependent on temperature and mineral composition. This process
612 involves the dissolution of material in high-stress areas and precipitation in
613 low-stress areas, facilitated by diffusive mass transfer through the pore fluid
614 [61, 62, 63].

615 Figure 25 provides a schematic representation of the pressure solution
616 process at the boundaries between two grains. In this scheme, the com-
617 pacted granular medium is subjected to stress, leading to the formation of
618 high-stress and low-stress regions. Within the high-stress regions at the con-
619 tact, dissolution takes place due to a modification of the chemical potential,
620 resulting in the removal of material. This dissolved material is then trans-
621 ported through the pore fluid to the low-stress regions far from the contact,
622 where precipitation occurs, leading to the formation of new material.

623 Here, the Equation 13 is solved to track the solute generation, and its
624 diffusion, and to ensure the total mass is conserved.

625 In the phase-field formulation, Equation 11, the external source term E_d

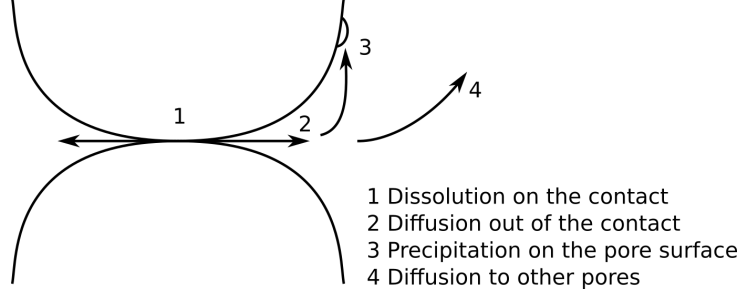


Figure 25: Scheme of pressure solution phenomena.

626 (presented in Equation 11) models both the mechanical energy at the contact
 627 level (E_{mec}) and the chemical energy due to the solute (E_{che}). As explained
 628 in Figure 9, the mechanical energy induces dissolution, whereas the chemical
 629 energy induces precipitation. The formulation of this source term is given by
 630 Equation 17.

$$E_d = E_{mec} - E_{che} = \sum_j \alpha \min(\eta_i) e_{mec} \times \eta_j^2(3 - 2\eta_j) - \chi c \times \eta_j^2(3 - 2\eta_j) \quad (17)$$

631 In Equation 17, E_d is defined as the difference between the mechanical energy
 632 (E_{mec}) and the chemical energy (E_{che}). The mechanical energy term is com-
 633 puted based on the contact level from the DEM simulation and is applied
 634 to the contact area. The solute concentration c is multiplied by a coeffi-
 635 cient χ representing the chemical energy term. A function of interpolation
 636 $\eta_j^2(3 - 2\eta_j)$ is used to apply the source term E_d only at the interface of the
 637 phase variables ($\eta_j \neq 1$ or 0).

638 The source term E_{mec} is non-zero only if both phase variables η_i involved
 639 in the contact are non-zero, approximately representing the contact area.
 640 This assumption generates a heterogeneous dissolution focused on the con-

641 tact area. To ensure stability, it is recommended to work with a source term
 642 E_{mec} lower than 0.2. The coefficient α is used to normalize the mechanical
 643 energy term. The term e_{mec} , defined in Equation 18, represents the poten-
 644 tial mechanical energy at the contact level and is computed as a potential
 645 normalized according to the relation 2.

$$e_{mec} = \frac{\int_{\Omega} F s_n d\delta_n}{\sum_{\Omega} \min(\eta_i)} = \frac{\int_{\Omega} k \delta_n^{3/2} d\delta_n}{\sum_{\Omega} \min(\eta_i)} = \frac{\frac{2}{5} k \delta_n^{5/2}}{\sum_{\Omega} \min(\eta_i)} \quad (18)$$

646 The quantity $\sum_{\Omega} \min(\eta_i)$ represents the sum over the contact area of the
 647 minimal value between phase variables involved η_i at each node. This variable
 648 is equivalent to the contact surface and ensures that the amount of energy in-
 649 troduced in the system from the interaction between grains remains constant.
 650 For example, if only two grains are in contact, the introduced energy term
 651 $\sum_{\Omega} E_{mec}$ is proportional to $\frac{2}{5} k \delta_n^{5/2}$. Thus, the term remains constant with time
 652 as $\sum_{\Omega} E_{mec} = \sum_{\Omega} \alpha \min(\eta_i) e_{mec} = \sum_{\Omega} \alpha \min(\eta_i) \frac{\frac{2}{5} k \delta_n^{5/2}}{\sum_{\Omega} \min(\eta_i)} =$
 653 $\alpha \frac{2}{5} k \delta_n^{5/2}$.

654 As explained in Section 2.3, the deformation induced by the phase-field
 655 simulation and the grain displacements computed in the discrete element
 656 modelization are closely interconnected. Following an inspired Aitken method
 657 [52], a variable time step is used during phase-field simulations to minimize
 658 grain deformation. The values of the thresholds are obtained by trial and
 659 error.

660 4.1. 2 grains configuration

661 A first simulation campaign was conducted using only two grains with the
 662 same radius, and the parameters used for this pressure solution simulation

are listed in Table 5. Since the configuration is relatively simple, the discrete element method (DEM) step can be simplified by using an analytical solution. In this case, the steady overlap between the two grains can be computed from the applied external load, and only an overlap is applied to move the grains.

Parameters	Unit	Value
Mobility for phase-field η_j	-	1
Gradient coefficient for phase-field κ_j	-	0.01
Gradient coefficient for solute κ_c	-	50
Initial time step for phase-field simulation	s	0.2
Duration of the phase-field simulation	s	$6 \times$ the time step
Overlap applied	μm	10% of the initial radius
Chemical energy coefficient χ	-	0.5
Mechanical energy coefficient α	-	Initial $\sum_{\Omega} \min(\eta_i)$

Table 5: Parameters used for the two grains pressure solution simulation.

The configuration of the two-grain system at different times is depicted in Figure 26, and a movie is linked to the article for a more comprehensive visualization. The initial configuration consists of two grains in contact with no solute in the sample. As the simulation progresses, dissolution occurs at the contact, resulting in a decrease in the phase-field variables and the generation of solute. The dissolution process continues, and the solute diffuses throughout the sample. The amount of solute generated decreases as the chemical energy at the contact balances the mechanical energy. Eventually, a steady-state is reached, where grains continue to dissolve at the contact, while an equal amount of solute precipitates far from the contact.

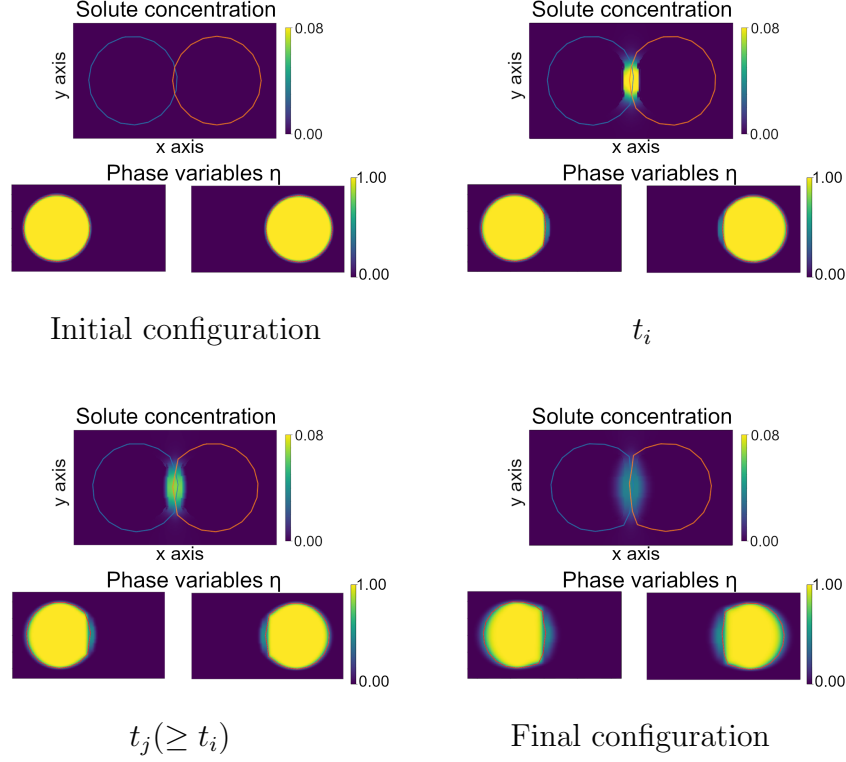


Figure 26: Pressure solution phenomena between two grains. For each time, the upper plot represents the position of the grains and the solute concentration whereas the lower plots represent the phase-field variable of each grain. Between the initial and the final steps, solute has been generated and the shape of the grains have become less and less spherical.

677 Various curves presented in Figures 27, 28, 29, and 30 provide further
678 insights into the simulation. Figure 27 shows the evolution of lateral strain
679 with time, highlighting a creep phenomenon. This behavior is consistent with
680 previous research findings on the topic [60, 35]. The slowdown in the strain
681 evolution can be attributed to two scenarios: (i) the expanding contact area
682 reduces the introduced mechanical energy, and (ii) the pore water becomes
683 saturated with the solute concentration. This creep phenomenon is known as

684 the Andrade creep response, and it has been observed in various geomaterials
 685 [64]. The lateral strain evolves proportionally with the cubic root of time
 686 ($\epsilon = k t^{1/3}$), and the slope of 1/3 is verified for long-term times. Concerning
 687 the short-term times, the result and the Andrade creep law must not be
 688 compared as the validity of the latter is limited to the long term. This short
 689 term is dominated by dissolution only (not enough solute for precipitation).
 690 The lateral strain is computed incrementally, $\epsilon = \sum_i \Delta_i / (2 R)$, where Δ_i is
 691 the rigid body motion applied to verify the targetted overlap and R is the
 692 initial radius of the particles.

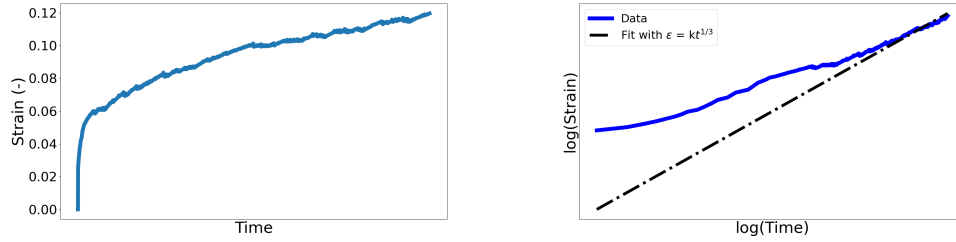


Figure 27: Evolution of the lateral strain with times (left) and fit with the Andrade creep law (right).

693 Figure 28 illustrates the shape evolution of the grains using different
 694 sphericity indices. The shape of a 2D grain is characterized by five spheric-
 695 ities, as defined in Equation 19 [65]. The sphericities decrease over time,
 696 indicating that the shape of grain 1 deviates from a perfect circle as pressure
 697 solution occurs. It is worth noting the evolution of the width-to-length ra-
 698 tio sphericity, which initially decreases, then increases, and finally decreases
 699 again. This suggests that the particle shape tends to transition from a circle
 700 to a square and then to a rectangle.

$$\begin{aligned}
&\text{Area sphericity} \quad \frac{A_s}{A_{cir}} \\
&\text{Diameter sphericity} \quad \frac{D_c}{D_{cir}} \\
&\text{Circle ration sphericity} \quad \frac{D_{ins}}{D_{cir}} \\
&\text{Perimeter sphericity} \quad \frac{P_c}{P_s} \\
&\text{Width to length ratio sphericity} \quad \frac{d_2}{d_1}
\end{aligned} \tag{19}$$

Here, A_s is the surface of the grain, A_{cir} is the surface of the minimum
 circumscribing circle, D_c is the diameter of the perfect circle having the
 same area as the grain $(= 2\sqrt{A_s/\pi})$, D_{cir} is the diameter of the minimum
 circumscribing circle, D_{ins} is the diameter of the largest inscribing circle,
 P_c is the perimeter of the perfect circle having the same area as the grain
 $(= 2\sqrt{A_s\pi})$, P_s is the perimeter of the grain, and d_1 and d_2 are the length
 and the width of the grain.

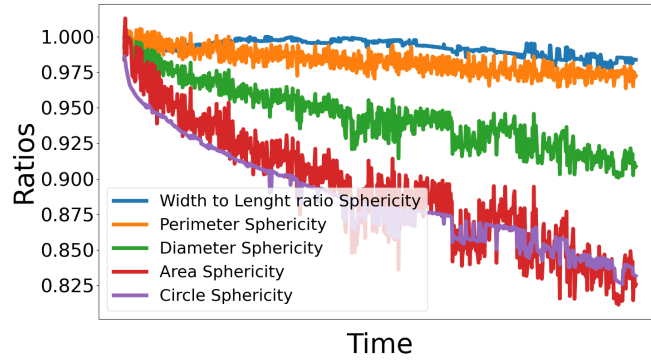


Figure 28: Evolution of different indices to appreciate the sphericity of the grain 1.

The evolution of the chemical ed_{che} , mechanical ed_{mec} , and total energy ed

709 at the center point of the sample $(0, 0)$ is depicted in Figure 29. This point is
 710 selected because it always remains in the contact area during the simulation.
 711 As explained in Equation 17, the chemical and mechanical energy terms are
 712 proportional to the solute concentration at the selected point and the contact
 713 surface, respectively. The simulation starts with significant dissolution of
 714 the grains, followed by solute diffusion and precipitation. These phenomena
 715 increase the size of the contact area, leading to a decrease in the mechanical
 716 energy term. By choosing appropriate parameters, the two limiting scenarios
 717 described by Lu et al. [66] can be reproduced.

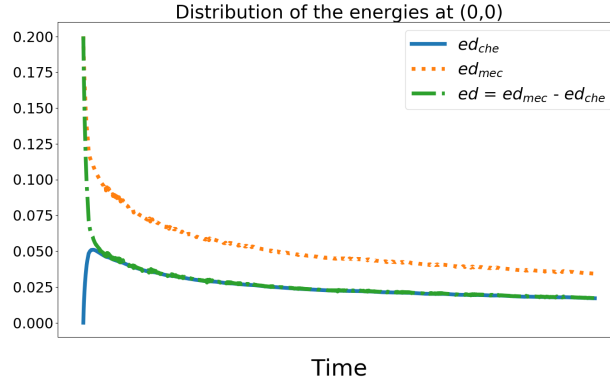


Figure 29: Evolution of the chemical, mechanical and total energy at the center point of the sample (always in the contact area).

718 Figure 30 presents the material distribution between the grains and the
 719 solute. A normalized distribution is shown for a clearer plot. The normal-
 720 ization is presented in Equation 20.

$$v^n(t) = \frac{v(t) - v_{min}}{v_{max} - v_{min}} \quad (20)$$

721 where $v^n(t)$ is the normalized value (grain or solute quantity in the entire

domain) at the time t , $v(t)$ is the value at the time t , v_{min} is the minimum reached of the value and v_{max} is the maximum reached of the value.

The evolution of the grain quantity is computed by summing the phase-field variables over the sample, while the evolution of the solute quantity is computed by summing the solute concentration over the sample. Figure 30b demonstrates the mass conservation, as the sum of the two variables remains constant (with a negligible difference due to mass loss during phase-field generations). In conjunction with Figure 29, it appears that a steady-state is reached. At the center point (in the contact area), material dissolution occurs (indicated by the positive sum of energy), but at the same time, the sum of the solute concentration over the sample remains constant. This indicates that the same quantity of solute generated at the contact is precipitated far from the contact, ensuring mass conservation.

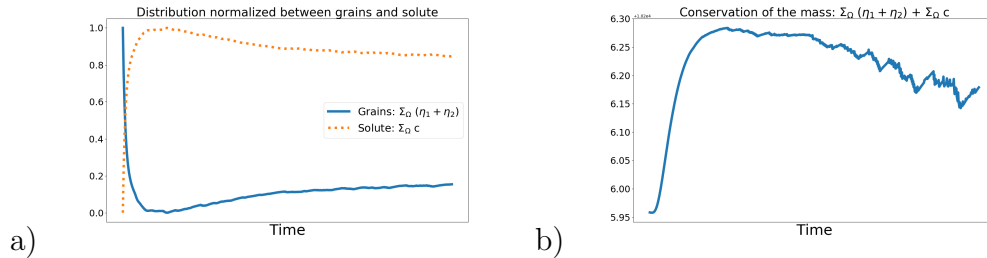


Figure 30: Evolution of the grains and solute quantities (first steps have been removed to facilitate the curve reading): a) A decrease in the grain quantity leads to an increase in the solute one. b) The conservation of the mass is verified.

Figure 31 illustrates the computing time distribution between the four main steps: DEM, DEM to PF, PF and PF to DEM. It appears the DEM part is negligible. Indeed, in this two grains configuration, an analytical solution can be used, decreasing hugely the time cost. The PF part is the most

important cost. Of course, this distribution is here only for example purposes. It depends on multiple parameters such as the mesh size, the number of processors used or the duration of the phase-field simulation (similar to the deformation induced by this step), among others.

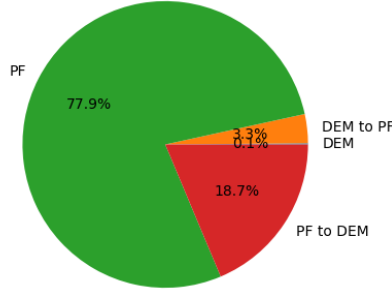


Figure 31: Computing time distribution between the four main steps: DEM, DEM to PF, PF and PF to DEM. The part of the DEM is negligible in this case because an analytical result is possible for the two grains configuration.

4.2. Multiple grains configuration

Once a standard configuration with only two grains has been studied, a multiple grains configuration is considered using the parameters presented in Table 6. In the multiple grains configuration, a new algorithm described in Appendix C is applied to move the solute with the grains.

Parameters	Unit	Value
Number of grains	-	30
Particle Size Distribution for grains (radius and percentage of the number of grains)	μm (%)	420 (17), 385 (33), 315 (33), 280 (17)

Young modulus	GPa	70
Poisson's ratio	-	0.3
Density	kg/m^3	2500
Friction grain-grain	-	0.5
Friction grain-wall	-	0
Restitution coefficient	-	0.2
Time step for discrete element model	sec	$\Delta t_{crit}/8$
Mobility for phase-field	-	1
Gradient coefficient for phase-field κ_j	-	0.01
Gradient coefficient for solute κ_c	-	50
Initial time step for phase-field simulation	s	0.01
Duration of the phase-field simulation	s	$8 \times \text{the time step}$
Linear force applied on the upper wall	N/m	$4 \times Y R_{mean}/1000$
Chemical energy coefficient χ	-	0.5
Mechanical energy coefficient α	-	Calibrated $\sum_{\Omega} min(\eta_i)$

Table 6: Parameters used for multiple grains pressure solution simulation.

748 To define the initial configuration, perfect disks are assumed. They are
749 randomly generated without overlap between them in the sample and then
750 loaded. Once a equilibrium is reached, the disks are discretized. The sample
751 generation aims to achieve a ratio between the sample diameter (D) and the

752 sample height (H) equal to 0.6.

753 The coefficient α is computed from a calibration simulation. Two grains
754 with a radius equal to the mean radius of the particle size distribution are
755 generated. Then, a vertical confinement force is applied to the two grains
756 to obtain an overlap. Finally, the coefficient α is equal to $0.06 \times \sum_{\Omega} \min(\eta_i)$,
757 where $\sum_{\Omega} \min(\eta_i)$ is the sum over all the calibration sample of the minimal
758 value of η_i at each node.

759 Figure 32 shows some pictures of the configuration, and a movie is linked
760 to the article for a more comprehensive visualization. Initially, there is no
761 solute in the sample. After several iterations, some solute is generated and
762 localized at the contact level. Grain reorganization occurs as shape deformation
763 takes place. Strong concentrations of solute appear, and solute leakage
764 can be observed at the top of the sample. In this configuration, the interaction
765 with the top wall is not considered, and nothing forces the generated
766 solute to follow the movement of the grains.

767 The evolution of the shape of the grains is illustrated in Figure 33 by the
768 evolution of the mean sphericity indices. In the case of multiple grains, the
769 mean value over the grains is considered. It can be observed that the grains
770 become less and less similar to a sphere over time.

771 Figure 34 shows the sample's vertical strain and compacity. The evolution
772 of compacity is important to comment on, as it is noisy and does not evolve
773 linearly with the vertical strain. It is computed with the ratio of the surface
774 grains over the sample surface. In this configuration, the confinement force is
775 large, and overlaps are quite large as well. To obtain a corrected compacity
776 index, it becomes important to reduce the contact surface and not count the

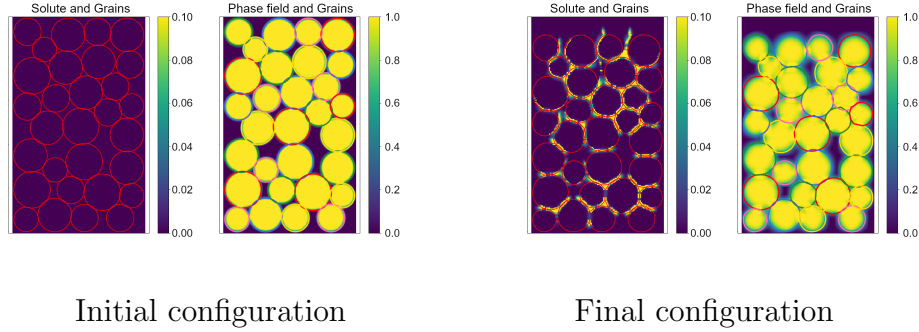


Figure 32: Pressure solution phenomena between multiple grains. For initial and final configurations, the left plot shows the position of the grains and the solute concentration, and the right plot shows the phase variables. Between the initial and the final steps, solute has been generated and the shape of the grains has become less and less spherical. The granular material becomes denser while the pressure-solution phenomenon occurs.

777 material surfaces twice.

778 A fit with an Andrade creep response has been attempted in Figure 35.
 779 The results of the simulation seems to be aiming to reach the tendency of
 780 the Andrade fit.

781 The normalized distribution (presented Equation 20) between grains and
 782 solute is illustrated in Figure 36a. The mass conservation is studied in Figure
 783 36b. It is observed that a small loss of mass occurs, approximately 1% of
 784 the initial mass. This loss is due to the grain displacement and detection
 785 algorithms, as these algorithms use a window around the grain to limit the
 786 computational cost and cut everything outside this window.

787 Figure 37 illustrates the computing time distribution between the four
 788 main steps: DEM, DEM to PF, PF and PF to DEM. The PF part is the most
 789 important cost and the DEM part is quite important also (as no analytical
 790 solution is used here compared to Figure 31). This distribution is here only

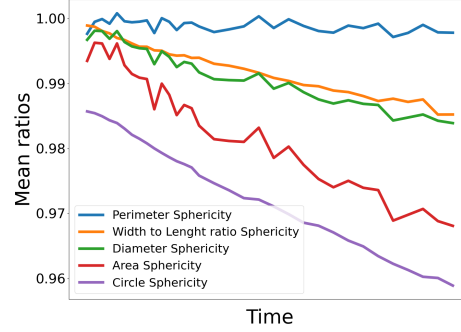


Figure 33: Evolution of different mean indices to appreciate the sphericity of the grains.

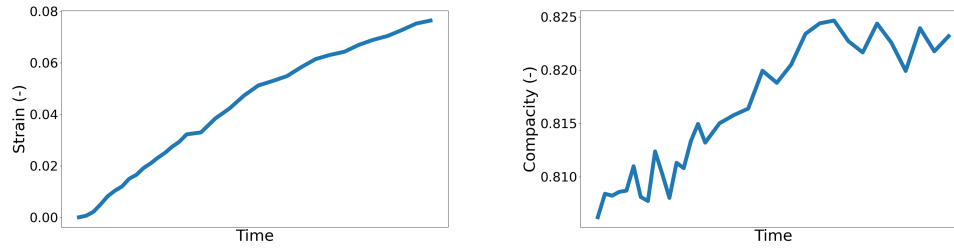


Figure 34: Evolution of the vertical strain and the sample compacity.

791 for example purposes as explained in section 4.1. It depends on multiple
 792 parameters listed above but also in this case, the discretization of the grains,
 793 the criteria to define the DEM steady-state or the duration of the phase-field
 794 simulation (similar to the deformation induced by this step), among others.

795 The goal of this Section was to highlight the PFDEM's capability to han-
 796 dle heterogeneous dissolution/precipitation processes. The observed grain
 797 shape evolution during the pressure solution phenomena is not uniform, as
 798 it varies from the contact point (high-stress area) to the pore surface (low-

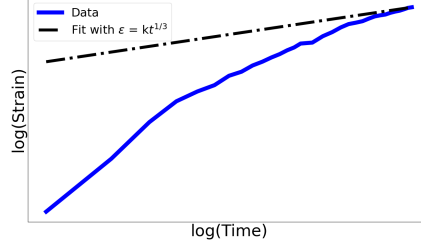


Figure 35: Comparison between the numerical results and the Andrade creep law for the evolution of strain with time.

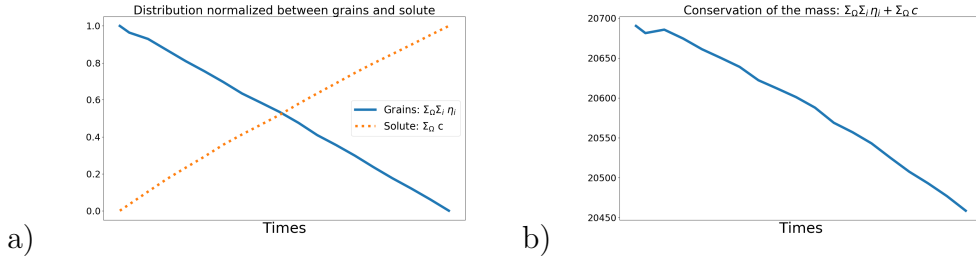


Figure 36: Evolution of the grains and solute quantities. a small loss of mass (because of the grain displacement and detection algorithms) is detected.

799 stress area). To capture this behavior, a solution concentration and a chemo-
800 mechanical formulation have been developed and incorporated into the phase-
801 field equations. The results demonstrate that the sphericities of the grains
802 decrease over time, indicating that the grains become less disk-like. Addi-
803 tionally, the sample tends to an Andrade creep response, which is commonly
804 observed in geomaterials during pressure solution. These findings illustrate
805 the PFDEM's ability to accurately simulate and analyze the complex be-
806 havior of pressure solution phenomena, taking into account heterogeneous
807 dissolution/precipitation effects.

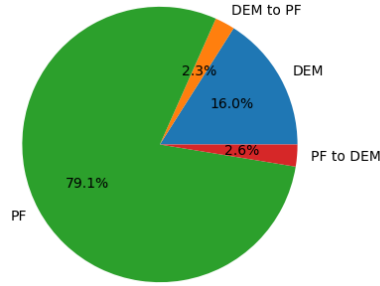


Figure 37: Computing time distribution between the four main steps: DEM, DEM to PF, PF and PF to DEM.

808 5. Conclusions

809 This work presents an extension of the classical discrete element model
810 by coupling it to the phase-field theory, enabling the capture of real grain
811 shape and its evolution based on physical laws. The study was conducted
812 in two steps. Firstly, the effect of grain shape was investigated through a
813 sample subjected to constant stress in oedometric conditions with partial
814 dissolution. The results highlighted the importance of considering the actual
815 grain shape instead of relying solely on modeling with rolling resistance as the
816 increase of grain angularity showed opposite evolution for the lateral pressure
817 coefficient evolution K_0 . Secondly, the pressure solution phenomena was
818 examined, considering a heterogeneous dissolution/precipitation simulation.
819 A comparison with experimental data was performed, demonstrating that
820 the simulated data tends to the Andrade creep response, a characteristic
821 behavior observed in granular materials undergoing pressure solution.

822 The proposed formulation opens up avenues for further exploration into
823 the influence of various parameters on the creep behavior and the effect of

824 grains characteristics. Parameters such as solute diffusivity, chemical energy
825 factor, and mechanical energy factor can now be systematically studied to
826 gain insights into their impact on the overall behavior. This coupling of dis-
827 crete element modeling with phase-field theory provides a useful framework
828 for investigating the complex interplay between grain characteristics and
829 the mechanical response of geomaterials undergoing dissolution/precipitation
830 processes.

831 **6. Software**

832 The PF model is solved using the MOOSE software [49], while the DEM
833 is solved using a custom-developed Python software. The scripts and algo-
834 rithms used in this study are available on GitHub at the following links:

- 835 • https://github.com/AlexSacMorane/PFDEM_AC
- 836 • https://github.com/AlexSacMorane/PFDEM_ACS
- 837 • https://github.com/AlexSacMorane/PFDEM_ACS_MultiGrains

838 **7. Acknowledgements**

839 We would like to thank the reviewers. We sincerely feel that the quality
840 of this article has been increased with their remark and advice.

841 This research has been partially funded by the Fonds Spécial de Recherche
842 (FSR) and by the Wallonia-Bruxelles Federation. The work has also received
843 funding from the NSF project CMMI-2042325.

844 **Appendix A. A Monte Carlo method algorithm to characterize a** 845 **polygonal particle**

846 A polygonal particle is defined by the coordinates of the vertices and
847 some essential information as center, surface and inertia must be known for
848 the DEM algorithm. A Monte Carlo [39] is used to compute them [28]. this
849 method is based on the creation of a box around the grain. Then, a point
850 is generated randomly inside the box. A point-inside-polygon test is used
851 to determine if the point is inside or not the polygonal particle (this test is
852 explained in the following). This step is repeated a large number of time.
853 The mass, the center and the inertia of the particle can be determined with
854 the equations A.1, A.2 and A.3.

$$mass = \frac{A_{box}}{N_p} \sum_{i=1}^{N_p} \chi(\vec{X}_i) \sigma \quad (A.1)$$

$$mass \times \vec{r} = \frac{A_{box}}{N_p} \sum_{i=1}^{N_p} \chi(\vec{X}_i) \sigma \vec{X}_i \quad (A.2)$$

$$I + mass \|\vec{r}\|^2 = \frac{A_{box}}{N_p} \sum_{i=1}^{N_p} \chi(\vec{X}_i) \sigma \|\vec{X}_i\|^2 \quad (A.3)$$

855 with A_{box} is the area of the box, $\vec{X}_i$ the coordinate of the random point,
856 N_p the total number of random point, σ the surface mass, $\vec{r}$ the center of
857 mass and I the moment of inertia. The function $\chi(\vec{X}_i)$ is the characteristic
858 function, which returns 1 if the point is inside the grain and 0 otherwise.

859 This characteristic function follows the point-inside-polygon test devel-
860 oped by Franklin [67]. A semi-infinite ray is generated from the point in the

861 direction of the increasing x . Every time this ray crosses an edge the point
862 switches between inside and outside.

863 **Appendix B. Method to build the field of the solute diffusion co-** 864 **efficient**

865 In the case of pressure solution, the problem asks for a heterogeneous
866 solute diffusion coefficient field. The goal is to have diffusion at the contact
867 area (to evacuate the solute generated) and in the pore area (as some fluid is
868 assumed). However, it should not have diffusion in the grain material. The
869 map of this coefficient is following the different steps :

- 870 1. for each node of the mesh, a Boolean value is attributed, depending on
871 the different phase variable (if $\eta_i > 0.5$ and $\eta_j > 0.5$ returns True; if
872 $\eta_i < 0.5$ and $\eta_j < 0.5$ returns True; else return False). A Boolean map
873 is generated.
- 874 2. a dilation method *scipy.ndimage.binary_dilation* is applied to the Boolean
875 map to generate a dilated map.
- 876 3. The diffusion coefficient map is generated. For each node, if the value
877 of the dilated map is True, κ_c is attributed to the diffusion map; else,
878 0 is attributed to the diffusion map.

879 As Figure B.38 highlights, the dilation step is needed to be sure the
880 contact zone is connected to the pore space. Depending on the configuration,
881 the selection of the morphological operator is the main choice. The larger
882 the size of the operator is, the more diffusion inside the grains occurs.

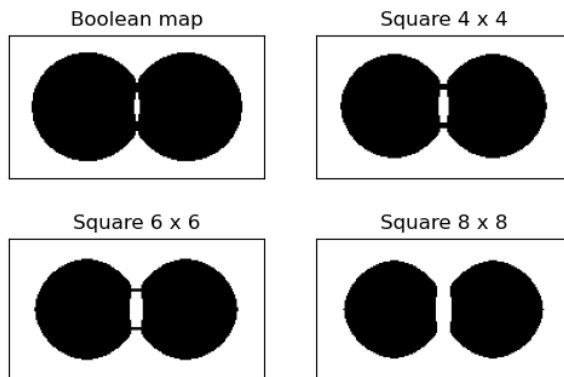


Figure B.38: Diffusion coefficient maps for different morphological operators.

883 **Appendix C. Method to move solute out of a grain**

884 Compare to the two grains pressure solution simulation, the multi-grain
 885 one faces trouble concerning the solute and the grain moves. As illustrated
 886 by Figure C.39, the solute generated does not follow the grain moves and
 887 it produces nonphysical results. It can be noticed the troubles are located
 888 more at the top of the sample. Hence, the simulation is made as the upper
 889 wall moves to verify the confinement pressure. Grains at the top move more
 890 than grains at the bottom. A new algorithm must be designed to move the
 891 solute with the grains.

892 The fundamental assumption is a small displacement of the grains. As
 893 illustrated by Figure C.40, all nodes are said available (in the pore space
 894 or in the contact area) or not (in only one grain) to host solute. Then, for
 895 nodes not available the solute is distributed to the nearest node available as
 896 illustrated in Figure C.40. If there are multiple nodes available at the same

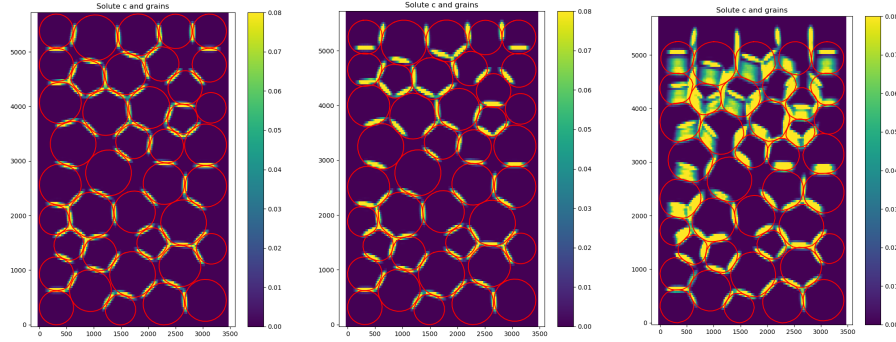


Figure C.39: Solute is generated initially (left). Then, the grains move after the DEM step to find a new equilibrium whereas the solute stays at the same place (center). Finally, it appears after multiple iterations that the solute needs to follow the grains (left).

897 distance, the concentration to move is divided into equivalent parts.

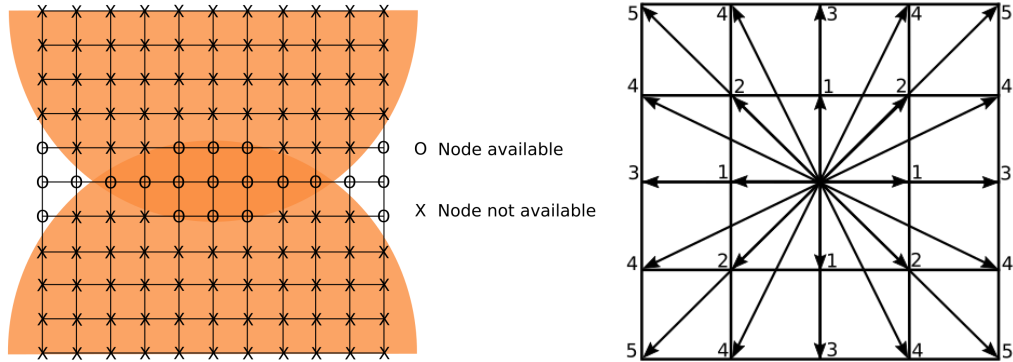


Figure C.40: A node is considered available to host solute if it is a pore space or a contact area (left) and an illustration of the priorities to move the solute to the nearest node (right).

898 The algorithm can be resumed by the following steps :

- 899 1. a Boolean map is generated depending if the node can host or not
900 solute.

- 901 2. a dilation method *scipy.ndimage.binary_dilation* is applied to the Boolean
902 map to generate a dilated map.
- 903 3. For all nodes of the dilated map not available to host the solute, the
904 nearest nodes are searched:
- 905 (a) It is checked if at least one node of the priority i can host solute.
- 906 (b) If no node is available: the previous step is repeated with priority
907 $i + 1$.
- 908 (c) If at least one node is available: the solute is moved with the rela-
909 tion at every available node $c_{available} = c_{available} + c_{not\ available} / n_{available}$,
910 where $c_{available}$ is the solute concentration at the available node,
911 $c_{not\ available}$ is the solute concentration at the not available node
912 and $n_{available}$ the number of available nodes at the priority i . The
913 solute concentration at the not available node is set to 0.

914 **References**

- 915 [1] J. McCartney, M. Sánchez, and I. Tomac, “Energy geotechnics: Ad-
916 vances in subsurface energy recovery, storage, exchange, and waste man-
917 agement,” *Comput. and Geotech.*, vol. 75, pp. 244–256, 2016.
- 918 [2] H. Rattez and M. Veveakis, “Weak phases production and heat genera-
919 tion control fault friction during seismic slip,” *Nat. Commun.*, vol. 11,
920 p. 350, 2020.
- 921 [3] M. Lesueur, T. Poulet, and M. Veveakis, “Three-scale multiphysics fi-
922 nite element framework (FE3) modeling fault reactivation,” *Comput.*
923 *Methods in Appl. Mech. and Eng.*, vol. 365, p. 112988, 2020.
- 924 [4] K. Ramesh Kumar, H. Honorio, D. Chandra, M. Lesueur, and H. Ha-
925 jibeygi, “Comprehensive review of geomechanics of underground hydro-
926 gen storage in depleted reservoirs and salt caverns,” *J. Of Energy Stor-*
927 *age*, vol. 73, p. 108912, 2023.
- 928 [5] R. Brzesowsky, C. Spiers, J. Peach, and S. Hangx, “Time-independent
929 compaction behavior of quartz sands,” *J. Geophys. Res. Solid Earth*,
930 vol. 119, pp. 936–956, 2014.
- 931 [6] J. Rohmer, A. Pluymakers, and F. Renard, “Mechano-chemical interac-
932 tions in sedimentary rocks in the context of CO2 storage: Weak acid,
933 weak effects?,” *Earth-Sci. Rev.*, vol. 157, pp. 86–110, 2016.
- 934 [7] J. Manceau and J. Rohmer, “Post-injection trapping of mobile CO 2
935 in deep aquifers: Assessing the importance of model and parameter
936 uncertainties,” *Comput. Geosci.*, vol. 20, pp. 1251–1267, 2016.

- 937 [8] H. Rattez, F. Disidoro, J. Sulem, and M. Veveakis, “Influence of dis-
938 solution on long-term frictional properties of carbonate fault gouge,”
939 *Geomech. For Energy And The Environ.*, vol. 26, p. 100234, 2021.
- 940 [9] K. Bjørlykke, M. Ramm, and G. Saigal, “Sandstone diagenesis and
941 porosity modification during basin evolution,” *Geol. Rundsch.*, vol. 78,
942 pp. 243–268, 1989.
- 943 [10] B. Bos, C. Peach, and S. CJ, “Slip behavior of simulated gouge-bearing
944 faults under conditions favoring pressure solution,” *J. of Geophys. Res.*,
945 vol. 105, pp. 16,699–16,717, 2000.
- 946 [11] E. Rutter, “Discussion on natural strain and geological structure - The
947 kinetics of rock deformation by pressure solution,” *Philos. Transact. of
948 the R. Soc. of London. Series A, Math. and Phys. Sci.*, vol. 283, pp. 203–
949 219, 1976.
- 950 [12] N. Sleep and M. Blanpied, “Creep, compaction and the weak rheology
951 of major faults,” *Nat.*, vol. 359, pp. 687–692, 1992.
- 952 [13] X. Tang and M. Hu, “A Reactive-Chemo-Mechanical Model for Weak
953 Acid-Assisted Cavity Expansion in Carbonate Rocks,” *Rock Mech. and
954 Rock Eng.*, vol. 56, pp. 515–533, 2023.
- 955 [14] B. Burman, P. Cundall, and O. Strack, “A discrete numerical model for
956 granular assemblies,” *Geotech.*, vol. 30, pp. 331–336, 1980.
- 957 [15] C. O’Sullivan, *Particulate Discrete Element modeling*. CRC Press, 2011.

- 958 [16] M. Cha and J. Santamarina, “Pressure-dependent grain dissolution using
959 discrete element simulations,” *Granul. Matter*, vol. 21, pp. 1–10,
960 2019.
- 961 [17] M. Alam, V. Parol, and A. Das, “A DEM study on microstructural
962 behaviour of soluble granular materials subjected to chemo-mechanical
963 loading,” *Geomech. for Energy and the Environ.*, vol. 32, p. 100390,
964 2022.
- 965 [18] M. van den Ende, G. Marketos, A. Niemeijer, and C. Spiers, “Investigating
966 Compaction by Intergranular Pressure Solution Using the Discrete
967 Element Method,” *J. of Geophys. Res.: Solid Earth*, vol. 123, pp. 107–
968 124, 2018.
- 969 [19] A. Guével, H. Rattez, and M. Veveakis, “Morphometric description of
970 strength and degradation in porous media,” *Int. J. of Solids and Struct.*,
971 vol. 241, p. 111454, 2022.
- 972 [20] T. Binaree, I. Preechawuttipong, and E. Azéma, “Effects of particle
973 shape mixture on strength and structure of sheared granular materials,”
974 *Phys. Rev. E*, vol. 100, p. 012904, 2019.
- 975 [21] G. Mollon, A. Quacquarelli, E. Andò, and G. Viggiani, “Can friction
976 replace roughness in the numerical simulation of granular materials?,”
977 *Granul. Matter*, vol. 22, p. 44, 2020.
- 978 [22] R. Rorato, M. Arroyo, A. Gens, E. Andò, and G. Viggiani, “Image-based
979 calibration of rolling resistance in discrete element models of sand,”
980 *Comput. and Geotech.*, vol. 131, p. 103929, 2021.

- 981 [23] X. Garcia, J. Latham, J. Xiang, and J. Harrison, “A clustered over-
982 lapping sphere algorithm to represent real particles in discrete element
983 modeling,” *Geotech.*, vol. 59, 2009.
- 984 [24] L. Rothenburg and R. Bathurst, “Numerical simulation of idealized
985 granular assemblies with plane elliptical particles,” *Comput. Geotech.*,
986 vol. 11, pp. 315–329, 1991.
- 987 [25] A. Podlozhnyuk, S. Pirker, and C. Kloss, “Efficient implementation of
988 superquadric particles in discrete element method within an open-source
989 framework,” *Comput. Part. Mech.*, vol. 4, pp. 101–118, 2017.
- 990 [26] P. Cundall, “Formulation of a three-dimensional distinct element model-
991 Part I. A scheme to detect and represent contacts in a system composed
992 of many polyhedral blocks.,” *Int. J. Rock Mech. Min. Sci. & Geomech.*,
993 vol. 25, pp. 107–116, 1988.
- 994 [27] E. Nezami, Y. Hashash, D. Zhao, and J. Ghaboussi, “A fast contact
995 detection algorithm for 3-D discrete element method,” *Comput. and*
996 *Geotech.*, vol. 31, pp. 575–587, 2004.
- 997 [28] F. Alonso-Marroquin and Y. Wang, “An efficient algorithm for granular
998 dynamics simulations with complex-shaped objects,” *Granul. Matter*,
999 vol. 11, pp. 317–329, 2009.
- 1000 [29] F. van der Meer and L. Sluys, “The Thick Level Set method: Sliding
1001 deformations and damage initiation,” *Comput. Methods in Appl. Mech.*
1002 *and Eng.*, vol. 285, pp. 64–82, 2015.

- [30] R. Kawamoto, E. Andò, G. Viggiani, and J. Andrade, “All you need is shape: Predicting shear banding in sand with LS-DEM,” *J. of the Mech. and Phys. of Solids*, vol. 111, pp. 375–392, 2018.
- [31] L. Landau, “The Theory of Phase Transitions,” *Nat.*, vol. 138, pp. 840–841, 1936.
- [32] J. Cahn and J. Hilliard, “Free energy of a nonuniform system. I. Interfacial free energy,” *The J. of Chem. Phys.*, vol. 28, pp. 258–267, 1958.
- [33] S. Allen and J. Cahn, “A microscopic theory for antiphase boundary motion and its application to antiphase domain coarsening,” *Acta Metall.*, vol. 27, pp. 1085–1095, 1979.
- [34] A. Wheeler, W. Ltoettinger, and G. Mc Fadden, “A Phase-field model for isothermal phase transitions in binary alloys,” *Phys. Rev. A*, vol. 45, pp. 7424–7439, 1992.
- [35] A. Guével, H. Rattez, and M. Veveakis, “Viscous phase-field modeling for chemo-mechanical microstructural evolution: application to geomaterials and pressure solution,” *Int. J. of Solids and Struct.*, vol. 207, pp. 230–249, 2020.
- [36] A. Munjiza, *The Combined Finite-Discrete Element Method*. Wiley, 2004.
- [37] K. Shinagawa, “Simulation of grain growth and sintering process by combined phase-field/discrete-element method,” *Acta Mater.*, vol. 66, pp. 260–369, 2014.

- 1025 [38] K. Samiei, B. Peters, M. Bolten, and A. Frommer, “Assessment of the
1026 potentials of implicit integration method in discrete element modeling of
1027 granular matter,” *Comput. and Chem. Eng.*, vol. 49, pp. 183–193, 2013.
- 1028 [39] R. Motwani and P. Raghavan, *Randomized Algorithms*. Cambridge Uni-
1029 versity Press, 1995.
- 1030 [40] V. Smilauer and al., *Yade Documentation 3rd ed.* The Yade Project,
1031 2021.
- 1032 [41] F. Dubois and M. Jean, “LMGC90 une plateforme de développement
1033 dédiée à la modélisation des problèmes d’interaction,” *Actes du Sixieme*
1034 *Colloque National en Calcul des Structures*, pp. 111–118, 2003.
- 1035 [42] G. Mollon, “A unified numerical framework for rigid and compliant gran-
1036 ular materials,” *Comput. Part. Mech.*, vol. 5, pp. 517–527, 2018.
- 1037 [43] H. Hertz, “Über die Berührung fester elastischer Körper (On the contact
1038 of elastic solids),” *J. fur die Reine und Angew. Math.*, vol. 92, pp. 156–
1039 171, 1882.
- 1040 [44] K. Johnson, *Contact Mechanics*. Cambridge University Press, 1985.
- 1041 [45] A. Di Renzo and F. Di Maio, “Comparison of contact-force models for
1042 the simulation of collisions in DEM-based granular flow codes,” *Chem.*
1043 *Eng. Sci.*, vol. 59, pp. 525–541, 2004.
- 1044 [46] R. Mindlin and H. Deresiewicz, “Elastic spheres in contact under varying
1045 oblique forces,” *J. Appl. Mech.*, vol. 20, pp. 327–344, 1953.

- [47] Y. Feng, “Thirty years of developments in contact modelling of non-spherical particles in DEM: a selective review,” *Acta Mech. Sin.*, vol. 39, p. 722343, 2023.
- [48] T. Takaki, “Phase-field modeling and simulations of dendrite growth,” *ISIJ Int.*, vol. 54, pp. 437–444, 2014.
- [49] C. Permann, D. Gaston, D. Andrš, R. Carlsen, F. Kong, A. Lindsay, J. Miller, J. Peterson, A. Slaughter, R. Stogner, and R. Martineau, “MOOSE: Enabling massively parallel multiphysics simulation,” *SoftwareX*, vol. 11, p. 100430, 2020.
- [50] N. Moelans, B. Blanpain, and P. Wollants, “An introduction to phase-field modeling of microstructure evolution,” *Calphad: Comput. Coupling of Phase Diagr. and Thermochem.*, vol. 32, pp. 268–294, 2008.
- [51] H. Shin and J. Santamarina, “Mineral Dissolution and the Evolution of k_0 ,” *J. of Geotech. and Geoenviron. Eng.*, vol. 135, pp. 1141–1147, 2009.
- [52] A. Aitken, “XX.—Studies in Practical Mathematics. II. The Evaluation of the Latent Roots and Latent Vectors of a Matrix,” *Proc. of the Royal Soc. of Edinburgh*, vol. 57, pp. 269–304, 1938.
- [53] M. Cha and J. Santamarina, “Dissolution of randomly distributed soluble grains: Post-dissolution k_0 -loading and shear,” *Geotech.*, vol. 64, pp. 828–836, 2014.
- [54] Y. Sheng, C. Lawrence, B. Briscoe, and C. Thornton, “Numerical studies of uniaxial powder compaction process by 3D DEM,” *Eng. Comput.*, vol. 21, pp. 304–317, 2004.

- 1069 [55] D. Potyondy and P. Cundall, “A bonded-particle model for rock,” *Int.*
1070 *J. Rock Mech. Min. Sci.*, vol. 41, pp. 1329–1364, 2004.
- 1071 [56] A. Sac-Morane, H. Rattez, and M. Veveakis, “Discrete Element Mod-
1072 eling of a fault reveals that viscous rolling relaxation controls friction
1073 weakening.” Submitted to *Granul. Matter*.
- 1074 [57] J. Eliáš, “Simulation of railway ballast using crushable polyhedral par-
1075 ticles,” *Powder Technol.*, vol. 264, pp. 548–465, 2014.
- 1076 [58] K. Szarf, G. Combe, and P. Villard, “Polygons vs. clumps of discs:
1077 A numerical study of the influence of grain shape on the mechanical
1078 behaviour of granular materials,” *Powder Technol.*, vol. 208, pp. 279–
1079 288, 2011.
- 1080 [59] J. Kafashan, J. Wiacek, N. Abd Rahman, and J. Gan, “Two-dimensional
1081 particle shapes modelling for DEM simulations in engineering: a re-
1082 view,” *Granul. Matter*, vol. 21, pp. 1–19, 2019.
- 1083 [60] S. De Meer, C. Spiers, and C. Peach, “Pressure solution creep in gypsum:
1084 Evidence for precipitation reaction control,” *Phys. and Chem. of the*
1085 *Earth*, vol. 22, pp. 33–37, 1997.
- 1086 [61] C. Spiers, P. Schutjens, R. Brzesowky, C. Peach, J. Liezenberg, and
1087 H. Zwart, “Experimental determination of constitutive parameters gov-
1088 erning creep of rocksalt by pressure solution,” *Geol. Soc. Spec. Publ.*,
1089 vol. 54, pp. 215–227, 1990.
- 1090 [62] J. Gratier, “Experimental pressure solution of halite by an indenter tech-
1091 nique,” *Geophys. Res. Lett.*, vol. 20, pp. 1647–1650, 1993.

- 1092 [63] J. Urai, Z. Schlöder, C. Spiers, and P. Kukla, *Dynamics of Complex*
1093 *Intracontinental Basins The Central European Basin System*, ch. 5.2
1094 Flow and Transport Properties of Salt Rocks. Springer Berlin, 2008.
- 1095 [64] D. Dysthe, Y. Podladchikov, F. Renard, J. Feder, and B. Jamtveit,
1096 “Universal Scaling in Transient Creep,” *Phys. Rev. Lett.*, vol. 89, pp. 1–
1097 4, 2002=.
- 1098 [65] J. Zheng and R. Hryciw, “Traditional soil particle sphericity, roundness
1099 and surface roughness by computational geometry,” *Geotech.*, vol. 65,
1100 pp. 494–506, 2015.
- 1101 [66] R. Lu, C. Cheng, T. Nagel, H. Milsch, H. Yasuhara, O. Kolditz, and
1102 H. Shao, “What process causes the slowdown of pressure solution creep,”
1103 *Geomech. and Geophys. for Geo-Energy and Geo-Resour.*, vol. 7, pp. 1–
1104 11, 2021.
- 1105 [67] R. Franklin, “How do I find if a point lies within a polygon,” 1994. In:
1106 comp.graphics.algorithms FAQ.