

DEM analysis of segregation phenomena in swelling granular media

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Abstract

Swelling granular media can experience size-induced percolation phenomena giving rise to segregation. In this work, the Discrete Element Method is employed to investigate the effects of size ratio and swelling kinetics on the segregation. The numerical analysis was carried out on a binary mixture consisting of coarse and fine particles and several mixing indices found in literature were adapted and tested for evaluating the mixing of expanding systems. Additionally, a relative percolation velocity was employed to quantify the percolation of fine particles. The results show that the percolation of fine particles becomes more significant as the size ratio increases. Additionally, results showed that the swelling kinetics has no impact on the segregation tendency. This research provides valuable insight into the effect of size ratio and swelling kinetics on the segregation behaviour of swelling granular materials, which can contribute to understanding percolation phenomena in various fields.

Keywords: Swelling, segregation, DEM, percolation velocity, mixing

1. Introduction

Swelling is observed in a variety of natural and industrial materials. For example, rice and super absorbent polymers can increase their radius by about 12% [1] and 600% [2], respectively, during soaking in water. The swelling phenomenon can be beneficial, i.e., for super absorbent polymers used to absorb liquids in hygienic products [3], or detrimental, which is the case for grains stored inside silos exposed to high humidity air [4].

The majority of these swelling systems are heterogeneous media, consisting of grains which differ in physical properties, i.e. size or shape. When a polydisperse granular bed expands, the bulk deformation generates large voids through which fine particles can percolate to the bottom. This size-induced percolation phenomenon leads to segregation.

Predicting and controlling segregation phenomena is of great importance in a variety of industries, from pharmaceuticals to agricultural. Hence, segregation has been extensively studied in the past, experimentally and numerically. For example, Volpato *et al.* [5] carried out an experimental investigation of shear-induced percolation in binary systems and proposed a theoretical model to predict the percolation velocity in such systems. Using a binary mixture of spheres differing only in size, they quantified the percolation speed for different shear rates and size ratios. Yan *et al.* [6] employed the Discrete Element Method (DEM) to analyse the mixing and segregation of a binary mixture in the Freeman FT4 powder rheometer. They characterised the system mixing using a segregation index which is based on the coordination number and found that the segregation in the FT4 rheometer is caused by the sifting/sieving mechanism induced by the blade movement. The segregation tendency and kinetic were found to be proportional to the size ratio and blade velocity, respectively.

All these previous studies focused on the segregation induced by an external perturbation which induces the deformation of the granular material, while to the best of our knowledge, no

research can be found for swelling granular media. However, understanding segregation in swelling granular systems is important to ensure stability, mechanical behaviour, material homogeneity and product quality of granular media. In our previous work [7], we employed the Discrete Element Method and the same approach proposed by Yan *et al.* [6] to carry out a preliminary analysis of the segregation as affected by particle numbers in a swelling granular system. It was found that the swelling perturbation increases the segregation and that very loose systems, with low particle numbers, have a high tendency to segregate which could be linked to the initial particle centre position in the system and not to percolation phenomena. Hence, the investigation of the impact of swelling on the segregation must consider a certain initial bed height such that the number of particles in the system does not affect the results. This work aims to extend the numerical segregation analysis to the impact of size ratio, swelling kinetic and system initial configuration on the size-induced percolation and segregation. Therefore, the swelling of systems containing a binary mixture with various size ratios is simulated by employing DEM. Several mixing indices and a relative percolation velocity are used to characterise the mixing of the granular beds. Two types of initial configurations are considered, i.e., initially randomly mixed and layered systems. Moreover, the segregation kinetic and tendency as affected by swelling kinetic is explored by changing the single particle swelling kinetic.

2. Methodology

2.1 DEM swelling model and simulation setup

The DEM software used in this work is the open-source code LIGGGHTS [7] and the Hertz-Mindlin model was used for contact modelling. Only spherical particles were considered and assumed to be made of super absorbent polymer which swell due to water absorption when

soaked in a fully saturated system. The swelling of each particle was simulated using the following model [8]:

$$V' = 1 - \exp(-kt) \quad (1)$$

where V' is a dimensionless swelling parameter defined based on the initial (V_0) and equilibrium (V_{eq}) volumes of each particle, as:

$$V' = \frac{V - V_0}{V_{eq} - V_0} \quad (2)$$

while t is the soaking time and k is the kinetic parameter which is found by fitting experimental data at a given temperature [2]. For more information about the model and its validation, the reader is referred to Ref. [8].

Table 1 lists material properties, and kinetic parameter k (Eq. 1) which, if not otherwise specified, are employed in the simulation. Note that \bar{D} is the ratio between the final and initial diameter of each particle and is fixed at 6.18 based on previous findings for a super absorbent polymer [8].

Table 1 DEM parameters and material properties [8]

	SAP	Wall
Kinetic parameter k (s^{-1})	0.003 [8]	-
Density ρ ($\frac{kg}{m^3}$)	1,600 [2]	-
Young modulus E (Pa)	5×10^4	2.50×10^7 [8]
Poisson ratio ν (-)	0.50 [2]	0.25 [8]
Friction coefficient (-)	0.096 [2]	-
\bar{D} (-)	6.18 [8]	-

The granular material used in the simulations is a binary mixture consisting of coarse particles (c) and fine particles (f), differing only in size. Hence, a size ratio (SR) was defined as:

$$SR = \frac{r_c}{r_f} \quad (3)$$

where r_c and r_f are the radius of coarse and fine particles, respectively. Various initial configurations of the system were considered, and described in the following sections; however, in all cases, particles were randomly generated inside the simulation domain and packed under gravity. All the simulations were run for 20 s because it was observed that the segregation increases significantly only in the first few seconds of swelling.

2.2 Mixing indices and relative percolation velocity

To assess quantitatively the mixing and segregation during swelling, several indices were employed. A segregation index (S) was determined as proposed by Yan *et al.* [6]:

$$S = \frac{C_{cc}}{C_{cc} + C_{cf}} + \frac{C_{ff}}{C_{ff} + C_{cf}} \quad (4)$$

where C_{cc} , C_{ff} and C_{cf} are the total number of contacts between coarse-coarse, fine-fine and coarse-fine, respectively. This parameter ranges from 0, when the system is perfectly mixed, to 2 which corresponds to a perfectly segregated configuration.

Furthermore, two additional mixing indices were evaluated, i.e., Lacey's index [9] and a new mixing index proposed by Govender *et al.* [10]. Lacey's index (M_1) is a statistical parameter determined by dividing the system into a certain number of zones (N_1) and is given by the ratio between the achieved mixing and the possible mixing of the system as:

$$M_1 = \frac{\sigma_0^2 - \sigma^2}{\sigma_0^2 - \sigma_r^2} \quad (5)$$

where σ^2 represents the variance for the concentration of fine particles (chosen as reference component) in each zone, and is determined as:

$$\sigma^2 = \sum_{i=1}^{N_1} \frac{(\Phi_i - \Phi_m)^2}{N_1} \quad (6)$$

where Φ_i indicates the mass concentration of fine particles in the i^{th} zone, while Φ_m is the average mass concentration of fine particles in the entire system [11]. σ_0^2 and σ_r^2 are, respectively, the maximum and minimum of σ^2 and are given by:

$$\sigma_0^2 = \Phi_m(1 - \Phi_m) \quad (7)$$

$$\sigma_r^2 = \frac{\Phi_m(1 - \Phi_m)}{n_p} \quad (8)$$

where n_p is the maximum possible particles in a cell and is evaluated based on the cell and fine particle volumes. In literature, when determining Lacey's index, the system is divided into a fixed number (N_1) of zones having a fixed size [11], [12]. In our case, we deal with a swelling system which changes its size with time, hence it is not possible to employ a fixed cell number and size. Therefore, in our approach, we employ a fixed cell size (S), while the number of cells (N_1) is not constant and is determined as the total number of cells occupied by particles at that swelling time.

Govender *et al.* [10] proposed a mixing index which can be applied to multicomponent systems. This mixing index, indicated as M_2 , for our swelling binary mixture can be determined by dividing the system into a certain number of cells (N_2) having a fixed size (S). The total

mixing index of the system is evaluated as an average of the mixing index of each cell ($M_{2,i}$), i.e.:

$$M_2 = \sum_{i=1}^{N_2} \frac{M_{2,i}}{N_2} \quad (9)$$

Considering $SumProb_i$ in Eq. 10 as the sum of the contribution of each mixture component to the mixing statistics of the i^{th} cell, the mixing index of the i^{th} cell is evaluated with Eq. 11.

$$SumProb_i = \frac{\Phi_{i,f}}{MaxD} + \frac{\Phi_{i,c}}{MaxD} \quad (10)$$

$$M_{2,i} = SumProb_i - 1 \quad (11)$$

where $\Phi_{i,f}$ and $\Phi_{i,c}$ are the mass concentration of fine and coarse particles in the i^{th} cell, while MaxD is the maximum denominator which, for a system containing an equal concentration in the mass of fine and coarse particles, is determined as the maximum among $\Phi_{i,f}$ and $\Phi_{i,c}$. Note that for both M_1 and M_2 , the size of the cell (S) is set as the maximum particle radius at the final simulated time and only cells containing more than 10 particles are considered in the evaluation of the mixing index. The two mixing indices (M_1 and M_2) range from 0, for a completely segregated system, to 1 for a completely mixed configuration.

Along with the above mixing and segregation indices, a relative percolation velocity was employed to characterise the percolation tendency of the systems. Volpato *et al.* [5] proposed a theoretical model to determine the percolation velocity of a sheared system; however, their methodology is difficult to apply to swelling systems where particles are constantly expanding and moving towards the top of the system. Hence, a relative percolation velocity ($\overline{v_p}$) was estimated directly from the simulation outputs, defining a percolation distance (D_p) of the fine particles relative to the coarse ones as:

$$D_p = Cm_f - Cm_c - (Cm_{f0} - Cm_{c0}) \quad (12)$$

where Cm_f and Cm_c are the centre of mass of the fine and coarse particles, respectively, and are defined based on the z position and mass (m) of each particle in the system:

$$Cm_c = \frac{z_{i,c} \cdot m_{i,c} + \dots + z_{n,c} \cdot m_{n,c}}{m_{tot,c}} \quad (13)$$

$$Cm_f = \frac{z_{i,f} \cdot m_{i,f} + \dots + z_{n,f} \cdot m_{n,f}}{m_{tot,f}} \quad (14)$$

the subscripts n_f and n_c are, respectively, the total number of fine and coarse particles, m_{tot} represents the total mass and the subscript 0 (in Eq. 12) represents the initial condition. The percolation distance is divided by time t to evaluate the percolation velocity (v_p):

$$v_p = \frac{D_p}{t} \quad (15)$$

The relative percolation velocity (\bar{v}_p) is evaluated considering the ratio between the percolation velocity (v_p) and the bed expansion velocity (v_b).

$$\bar{v}_p = \frac{v_p}{v_b} \quad (16)$$

Note that the bed expansion velocity is determined based on the time evolution of the height of the granular system. The system scale could affect the segregation index and the relative percolation velocity, hence, before investigating the size ratio effect on the segregation, the effect of system dimension was investigated.

2.3 Effect of the system scale on the segregation index

Scaling up and reducing the cross-sectional area of the system are good strategies to optimise the DEM computational time. To investigate whether the system scale affects the segregation analysis, two preliminary case studies were performed. In the first one, two systems were considered as reported in Table 2, i.e. a “small system” and a “scaled-up system”. Note that the size ratio (SR) in the two systems is kept constant. The DEM time step of the two simulations was evaluated accordingly as 20% of the Rayleigh time. The visual representation of the small (in red) and scaled-up (in purple) systems is shown in Fig. 1.

Table 2 Dimensions and time step employed in investigating the system scale impact on the segregation.

	Small system	Scaled up system
System dimensions (m)	0.01×0.01×0.1	0.1×0.1×1
r_f (m)	2.0×10^{-4}	2.0×10^{-3}
r_c (m)	3.0×10^{-4}	3.0×10^{-3}
time step (s)	4.0×10^{-5}	4.0×10^{-4}
SR	1.5	1.5

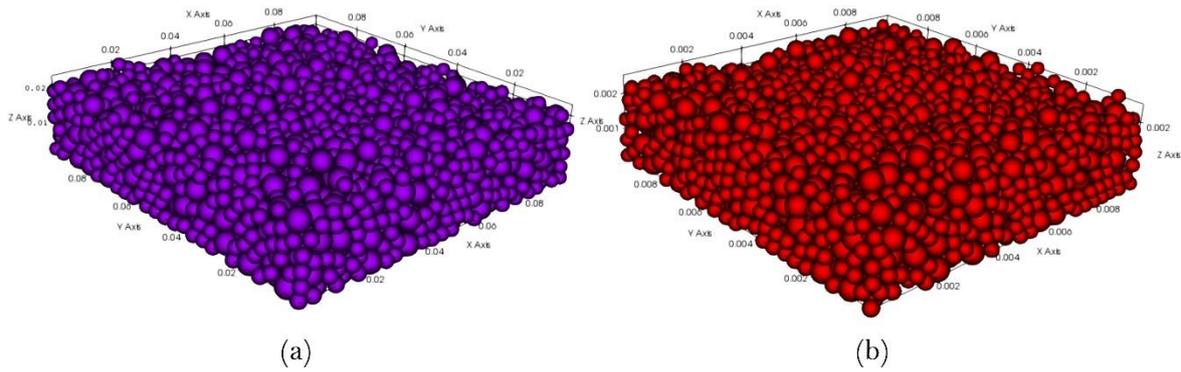


Fig. 1 Particle initial configuration for the (a) scaled-up system (in purple) and (b) small system (in red). Note the different dimensions of the axis grid.

The time evolution of the segregation index (S) as affected by system scale is shown in Fig. 2. The comparison was made between a small system and a scaled-up system containing different numbers of particles (N_p), i.e., 500, 1000, 3000. Results show that the segregation of the system is not affected by its scale, especially for dense granular beds containing more than 1,000 particles.

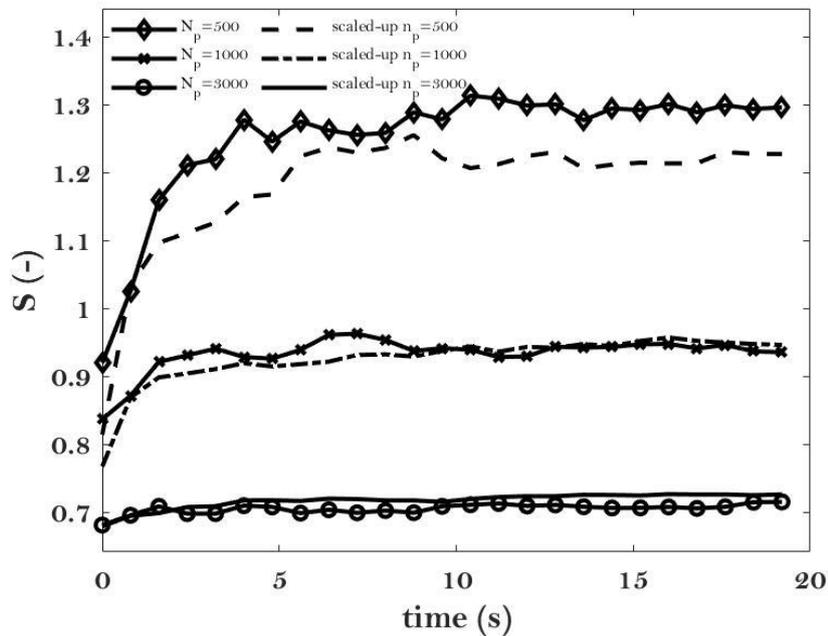


Fig. 2 Time evolution of the segregation index as affected by scaling up the system for granular beds containing different numbers of particles (N_p)

The impact of the system dimensions on the segregation tendency was further investigated by employing the scaled-up system (Table 2) and changing the cross-sectional area of the container while keeping the initial granular bed height (H_0) constant at 0.02 m. Hence, the x and y dimensions of the system were reduced to accommodate 1000, 2000 and 3000 particles keeping the pre-set H_0 constant. The initial configuration of the three systems is shown in Fig. 3, while Fig. 4 shows the evolution of the segregation index (S) as affected by the cross-sectional area. It is shown that by changing the container dimensions (in x and y), the index S has a variation of a maximum of 4% which can be considered negligible.

In our previous study [8], we observed a significant change in segregation with varying particle numbers. However, these results demonstrate that when performing a segregation analysis of swelling systems, an important role is played by the initial height of the granular bed, rather than the particle numbers.

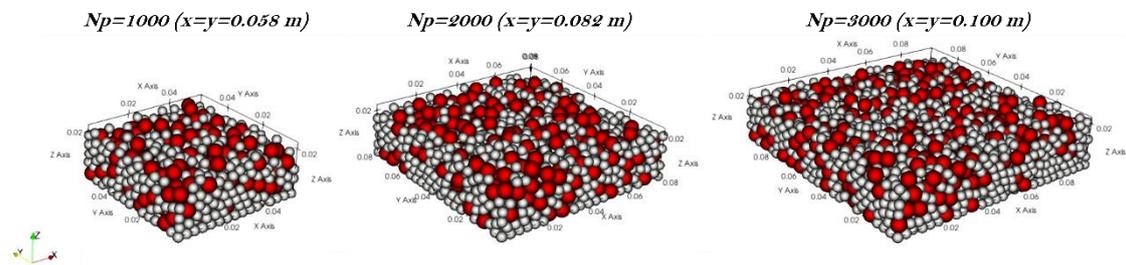


Fig. 3 Initial configuration of the systems with reduced cross-section area having the same initial bed height H_0 and containing different numbers of particles N_p .

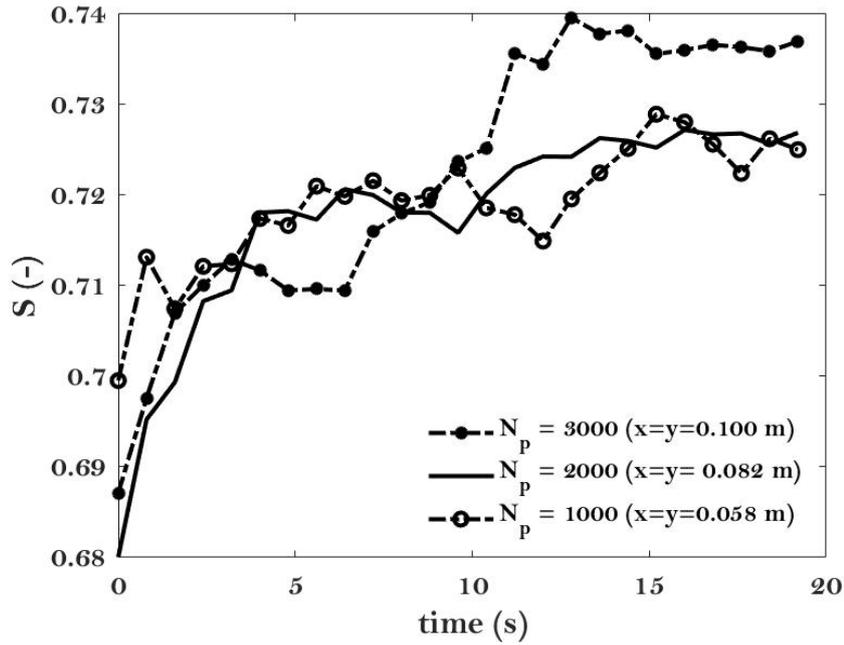


Fig. 4 Time evolution of the segregation index (S) as affected by the system cross-section area

Since scaling up the system does not affect the segregation analysis but significantly reduces the computational time (five times lower), the numerical analysis in the next sections is performed employing the scaled-up dimensions in Table 2. Moreover, the results from this preliminary study showed that to conduct a consistent DEM analysis, the initial bed height (H_0) in all cases must be fixed.

3. Results and discussion

The DEM analysis of segregation in swelling granular beds was performed by changing the size ratio (SR) and employing two different initial configurations, i.e., randomly mixed and segregated. Furthermore, the impact of swelling kinetic was investigated for the initially mixed system.

3.1 Mixed systems

3.3.1 Influence of size ratio

The investigation of the influence of the size ratio on the segregation of swelling granular systems was performed using the case studies reported in Table 3 in which the initial height of the bed (H_0) is kept constant while the size ratio (SR) was changed from 1.5 to 4. Note that the radius of the fine particles (r_f) was fixed at 2.0×10^{-3} m, while the coarse particle size (r_c) was changed based on the size ratio. All the cases consider a volume fraction of fine and coarse particles equal to 50%. Hence, the number of particles (N_p) in the system was evaluated by fixing the initial height and considering a bed porosity of 0.4. The initial configuration of the systems is shown in Fig. 5, while the final swollen configuration of the systems is shown in Fig. 6.

The simulation outputs were then processed to evaluate the time evolution of the segregation index (S) shown in Fig. 7, and the two mixing indices (M_1 and M_2) in Fig. 8 (a) and (b), respectively.

Table 3 Parameters employed in the simulations for investigating the impact of the size ratio on the segregation.

	SR 1.5	SR 2	SR 3	SR 4
H_0 (Initial height of the system, m)	2.23×10^{-2}	2.23×10^{-2}	2.23×10^{-2}	2.23×10^{-2}
r_f (fine particle radius, m)	2.0×10^{-3}	2.0×10^{-3}	2.0×10^{-3}	2.0×10^{-3}
r_c (coarse particle radius, m)	3.0×10^{-3}	4.0×10^{-3}	6.0×10^{-3}	8.0×10^{-3}
$N_{p,f}$ (number of fine particles)	1997	1997	1997	1997

$N_{p,c}$ (number of coarse particles)	592	250	74	32
N_p (total number of particles)	2589	2247	2071	2029

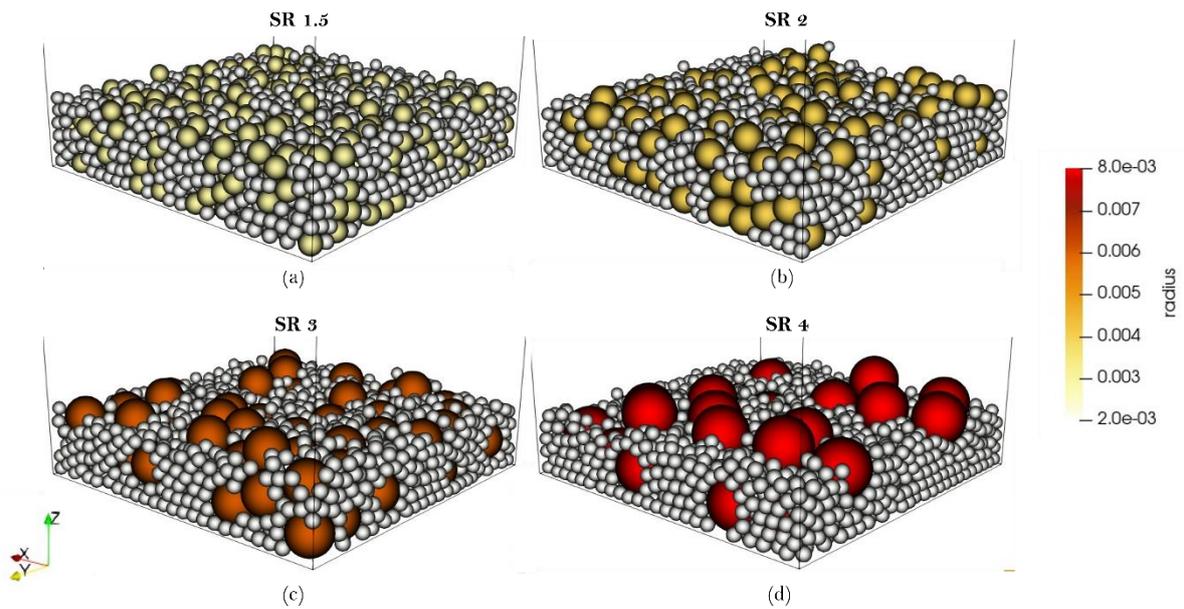


Fig. 5 Initial and unswollen configuration of the system (mixed) with a size ratio (SR) equal to (a) 1.5, (b) 2, (c) 3 and (d) 4

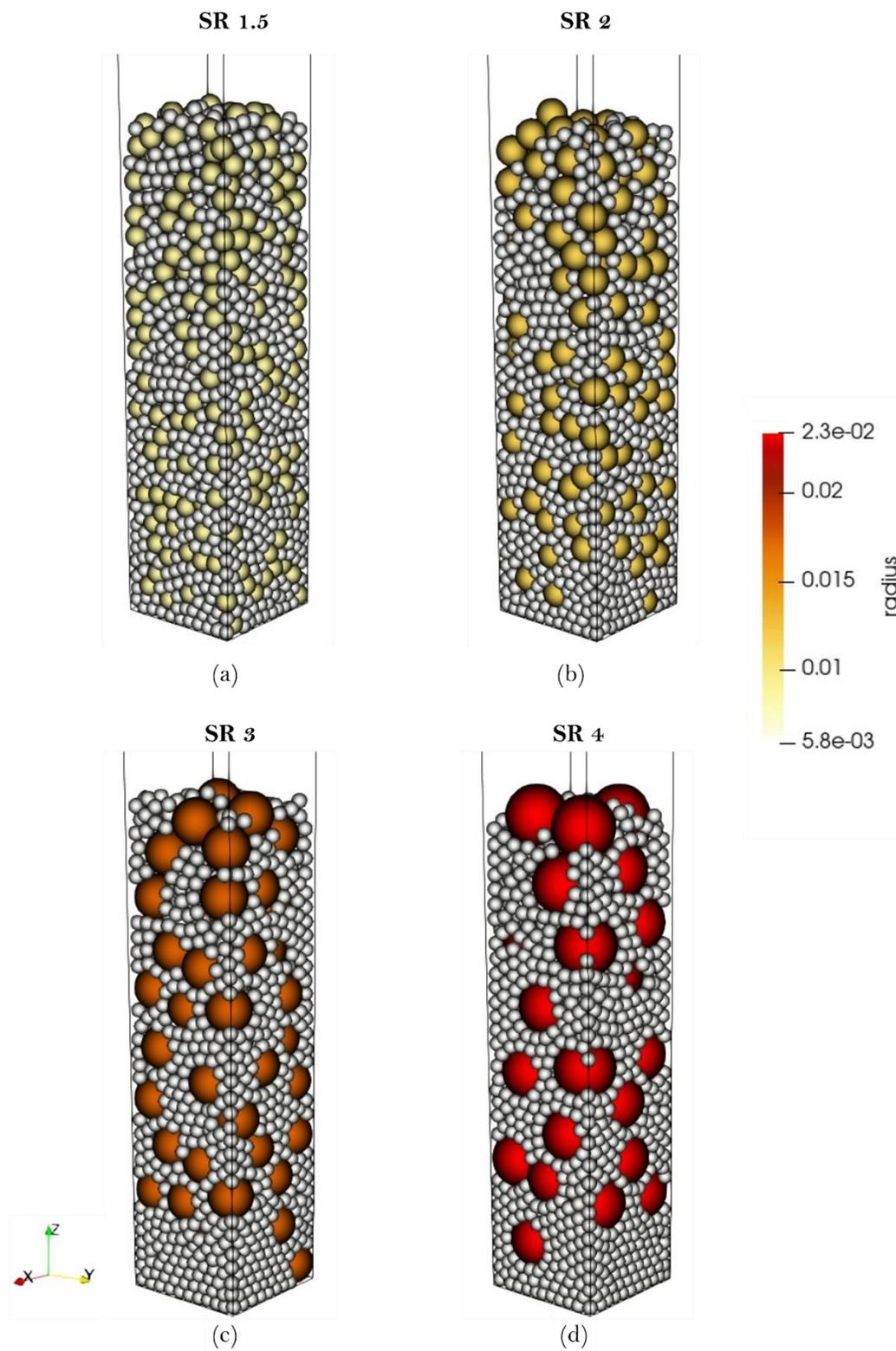


Fig. 6 Final and swollen configuration of the system (mixed) with a size ratio (SR) equal to (a) 1.5, (b) 2, (c) 2.5, (d) 3, (e)3.5 and (f) 4

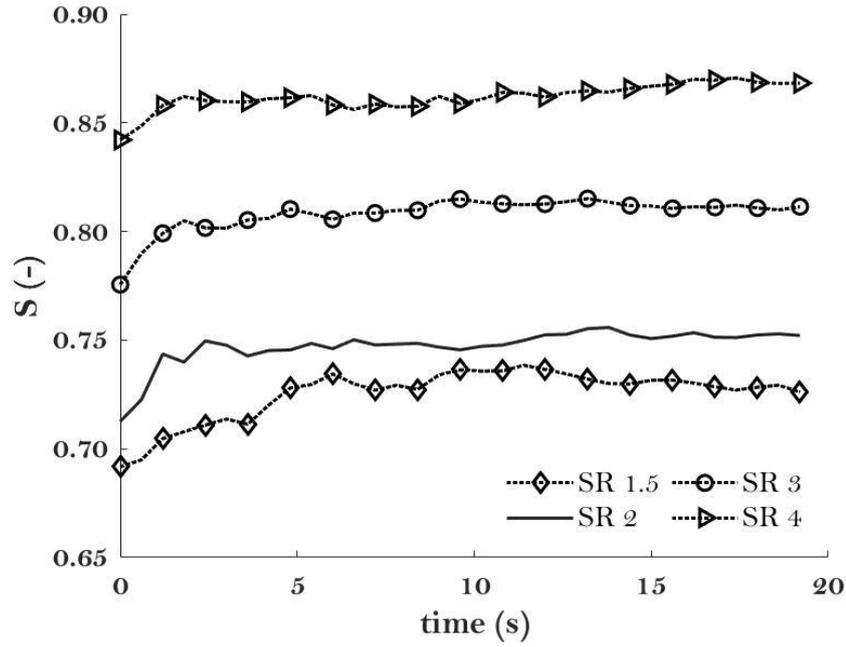


Fig. 7 Time evolution of the segregation index as affected by size ratio for an initially mixed system.

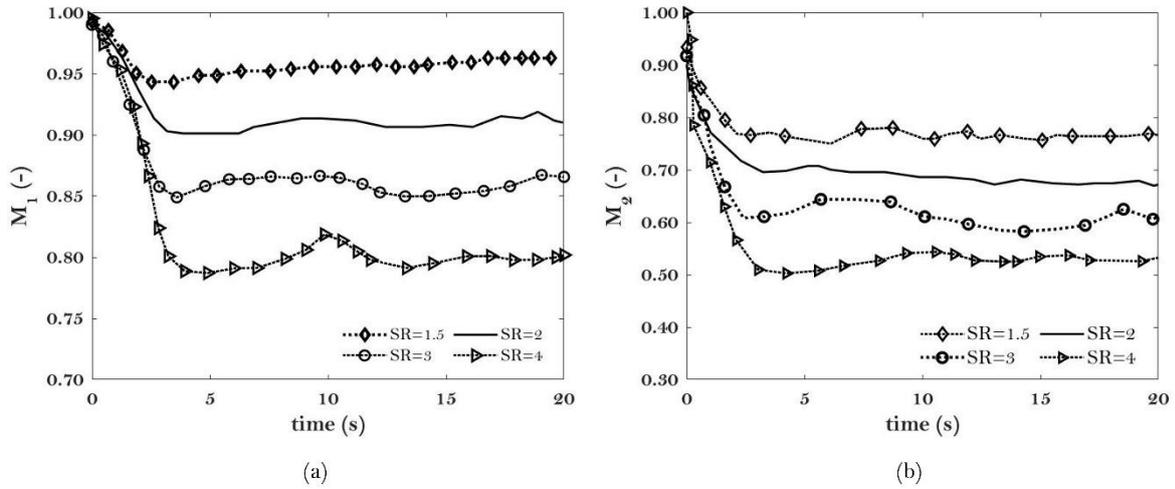


Fig. 8 Time evolution of the mixing indices (a) M_1 (Lacey's index) and (b) M_2 (Govender et al. [10]) as affected by size ratio for an initially mixed configuration.

As shown in Fig. 7, the segregation index increases with time for all size ratios (SR), implying that swelling increases the segregation of the system, as already demonstrated in our previous research work [8]. The same conclusion is reached by examining the mixing indices, M_1 and M_2 in Fig. 8, i.e., the indices decrease with time, indicating that the systems are evolving from a mixed to a segregated state. Interestingly, as shown in Fig. 7, the initial value of the

segregation index ranges from 0.69 for a size ratio of 1.5 to about 0.84 for a size ratio of 4, implying that this index is highly affected by the size ratio (SR). This is because the segregation index is evaluated based on the coordination number, which is affected by the ratio between the number of coarse and fine particles. As shown in Table 3, increasing the size ratio from 1.5 to 4, the number of fine particles remains constant ($N_f = 1997$), while the number of coarse particles (N_c) decreases from 592 to 32. This variation in the ratio between the number of coarse and fine particles affects C_{cf} in Eq. 4, which decrease as the size ratio increases, leading to a higher index (S). Therefore, the segregation index is a reliable indicator only in systems with a constant ratio between coarse and fine particles.

The bar graph in Fig. 9 shows the impact of the size ratio on the variation of the indices, which is evaluated as the percentage of the difference between the final and initial state normalised by the initial state, e.g., for the segregation index:

$$S_{\%} = \left| \frac{S_{fin} - S_0}{S_0} \right| \times 100 \quad (17)$$

As shown in Fig. 9, the segregation index variation with the size ratio shows an inconsistent trend, as its value decreases going from SR=1.5 to SR=2 and then increases from SR=2 to SR=3. This inconsistency was expected, because as already mentioned the segregation index is affected by the variation of particles number in the system. On the other hand, both M_1 and M_2 show that by increasing the size ratio, the segregation of the system increases. Indeed, M_1 shows that after 20 s of swelling the increase in segregation is about 3% at SR =1.5 and 19% at SR=4, and M_2 shows a segregation rise of 18% at SR = 1.5 and 48% at SR = 4.

The fact that a larger size ratio leads to more significant segregation could mean that at large SR, fine particles have larger voids in the expanding granular bed to percolate to the bottom.

This hypothesis is confirmed by the relative percolation velocity ($\overline{v_p}$), in Fig. 10, which is higher (in absolute value) as the size ratio increases. Note that the relative percolation velocity in all four cases exhibits negative values, meaning that the centre of mass of fine compared to coarse particles is moving towards the bottom of the system, which is expected if the system is migrating from an initially mixed to a segregated configuration.

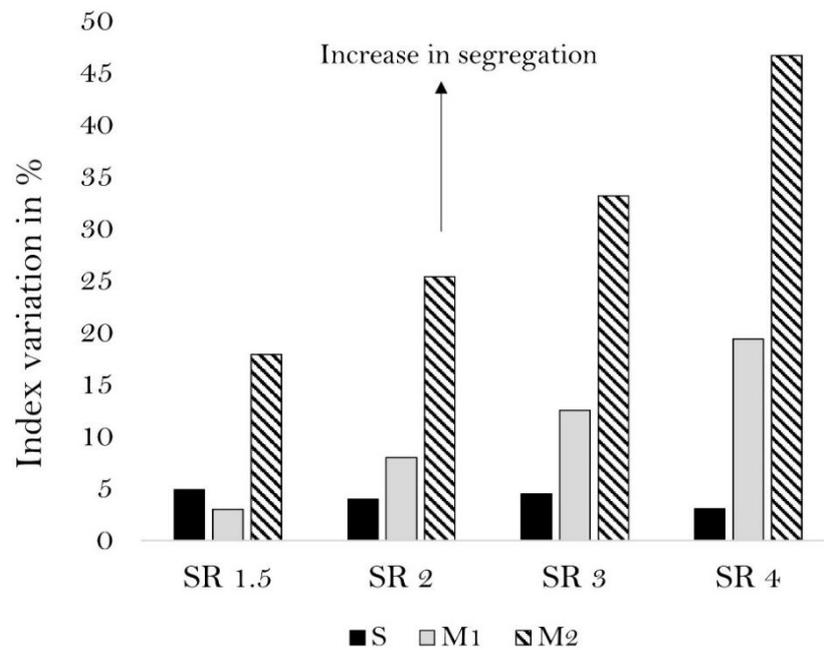


Fig. 9 Impact of the size ratio on the index variation after 20 s of swelling for an initially mixed system

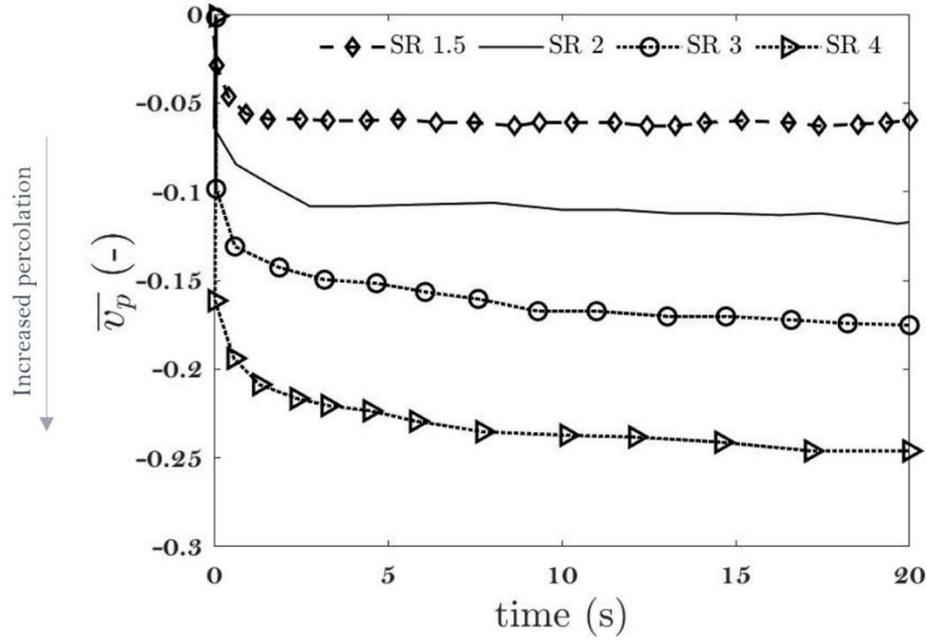


Fig. 10 Relative percolation velocity as affected by size ratio for an initially mixed configuration.

3.3.2 Influence of kinetic parameter

The impact of the swelling kinetic on the segregation was investigated by changing the kinetic parameter k (Eq. 1) while considering the same initial particle configuration with a size ratio equal to 2 and initially mixed system shown in Fig. 5 (b). Three different kinetics parameters (k) were employed, i.e., 0.001, 0.003 and 0.006 s^{-1} and the final configuration (after 20 s of swelling) of the three systems are shown in Fig. 11. At a glance, the three final configurations do not show any difference in terms of segregation. For a quantitative analysis, the time evolution of the segregation index (S) and two mixing indices (M_1 and M_2) is shown in Fig. 12 and Fig. 13, respectively. The segregation index is approximately the same when the kinetic increases from 0.001 s^{-1} to 0.003 s^{-1} , but slightly higher for a kinetic equal to 0.006 s^{-1} . However, note that the index at the highest kinetic is higher even prior to swelling, suggesting that the initial configuration is already slightly more segregated. As shown in Fig. 13, the time

evolution of both the mixing indices appears to be constant with the swelling kinetic: M_1 decreases from 1 to about 0.95 and M_2 from 0.85 to about 0.72. Indeed, the variation of all three indices after swelling in Fig. 14 shows no variation with k . The relative percolation velocity for the three systems, which is plotted in Fig. 15, shows negative values, which as explained previously is an indicator of systems evolving toward segregation through fine particle percolation through the interstitial spaces of the coarser particles. In agreement with the mixing indices, the relative percolation velocity appears to be almost constant when changing the swelling kinetics. The small difference observed for $k_1 = 0.001 \text{ s}^{-1}$ is shown to be statistically insignificant by a t-student test, as the p-value (P) is greater than 0.05 for both k_1 - k_2 ($P = 0.395$) and k_1 - k_3 ($P = 0.12$).

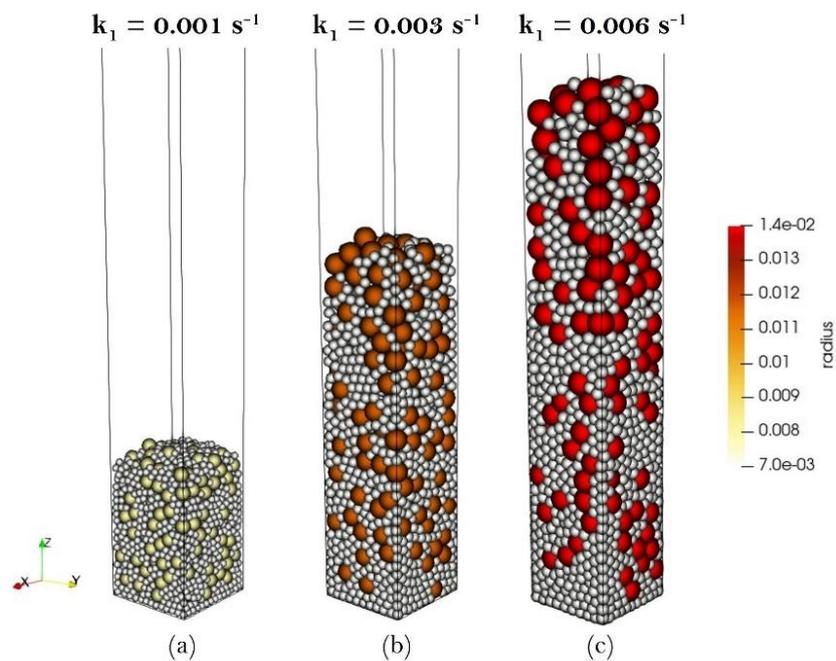


Fig. 11 Final configuration of the three systems which expanded for 20 s with different kinetics.

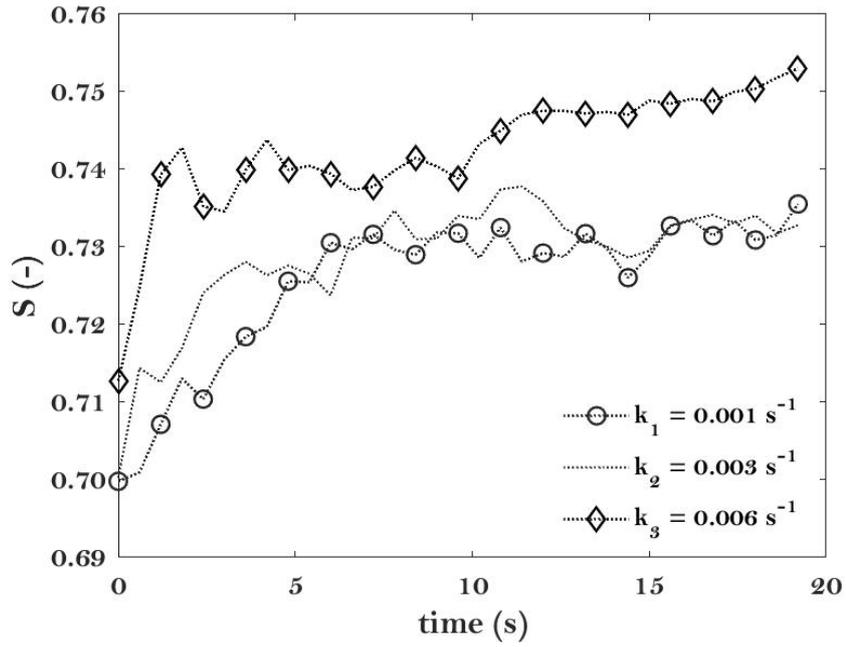


Fig. 12 Time evolution of the segregation index (S) as affected by swelling kinetics for an initially mixed configuration.

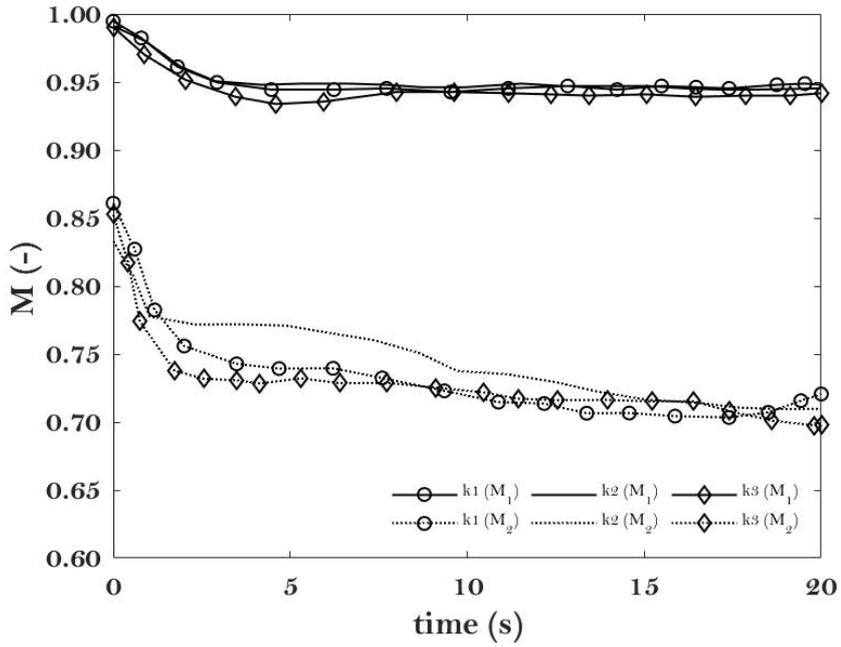


Fig. 13 Time evolution of the two mixing indices (M_1 and M_2) as affected by swelling kinetics for an initially mixed configuration.

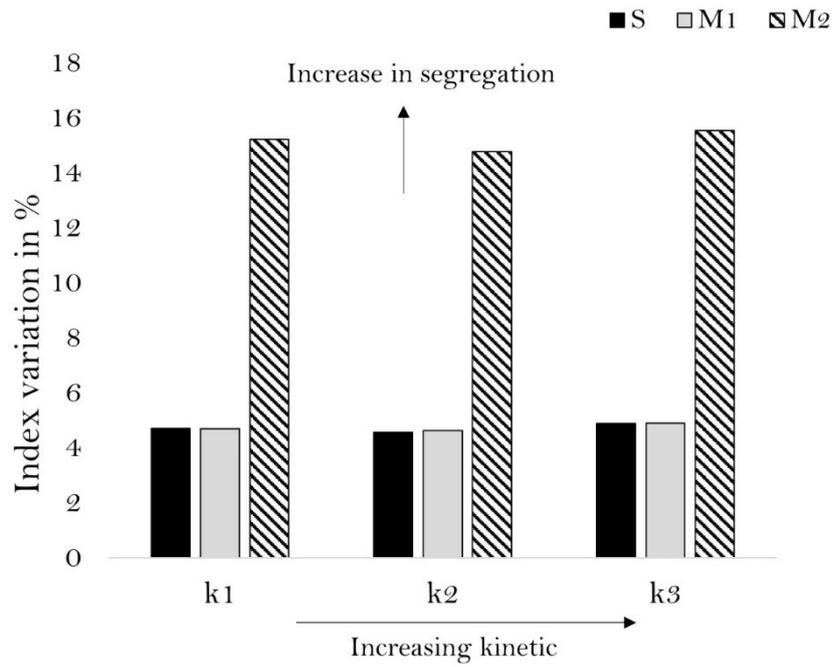


Fig. 14 Impact of the swelling kinetic on the index variation after 20 s of swelling for an initially mixed system

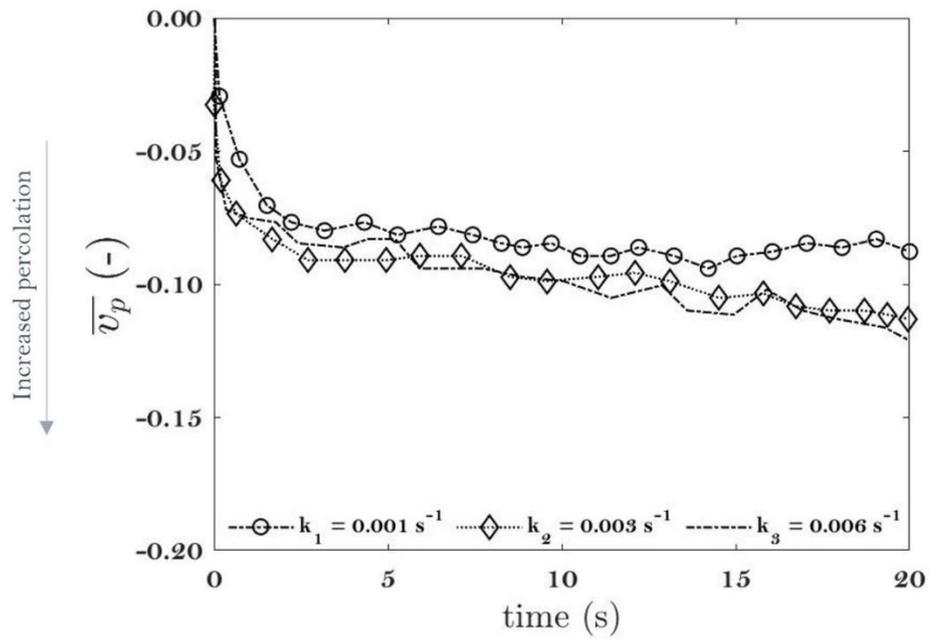


Fig. 15 Relative percolation velocity as affected by the swelling kinetics.

3.2 Layered system

The numerical analysis of the effect of swelling on the segregation was also carried out on initially layered systems, where fine particles were originally located at the top of coarse particles as shown in Fig. 16. The size ratio was changed from 2 to 4, keeping the same conditions and number of particles indicated in Table 3. Swelling was simulated for 20 s and the final configuration is shown in Fig. 17. Comparing the final configurations of the three systems, at glance the system with the highest size ratio ($SR = 4$) in Fig. 17 (c) is significantly more mixed compared to the system in Fig. 17 (a) ($SR = 2$).

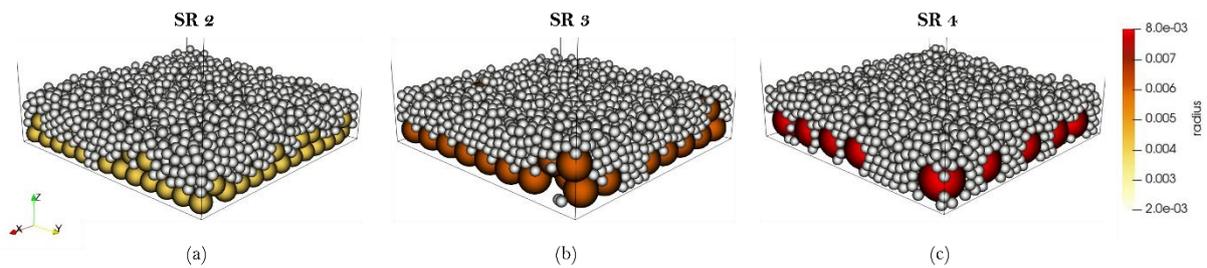


Fig. 16 Initial configuration of the system (layered) with a size ratio (SR) equal to (a) 2, (b) 3 and (c) 4.

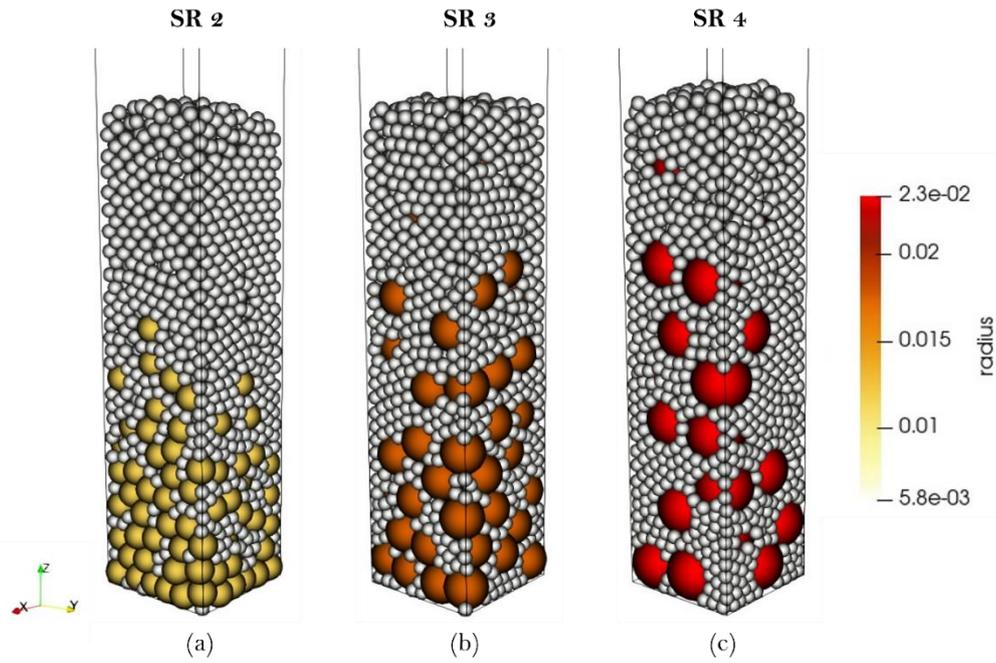


Fig. 17 Final and swollen configuration of the system (layered) with a size ratio (SR) equal to (a) 2, (b) 3 and (c) 4.

The quantitative analysis of the segregation of these three systems, performed through the determination of the segregation index (S) is shown in Fig. 18. It is shown that the segregation index (S) decreases with swelling, indicating that the three systems are evolving towards a mixed configuration. Fig. 19 (a) and (b) report the time evolution of the mixing indices, M_1 and M_2 respectively. Both the mixing indices increase with time, indicating that, as the system swells, fine particles percolate through the coarse particle layer. The mixing magnitude depends on the size ratio, i.e., the larger the difference in size, the greater the achieved mixing. Indeed, as shown in Fig. 20, according to M_1 , the mixing increased by about 120% when the size difference is the lowest (SR=2) and by about 130% when SR is the highest (SR=4). Similarly, according to M_2 , the mixing increased by 120% when SR=2 and by about 140% when SR=4. As found for the initially mixed configuration, the results in terms of the impact of size ratio on the segregation index (S) are inconsistent with the other indices, implying that this parameter is highly affected by the ratio between the number of coarse and fine particles in the system.

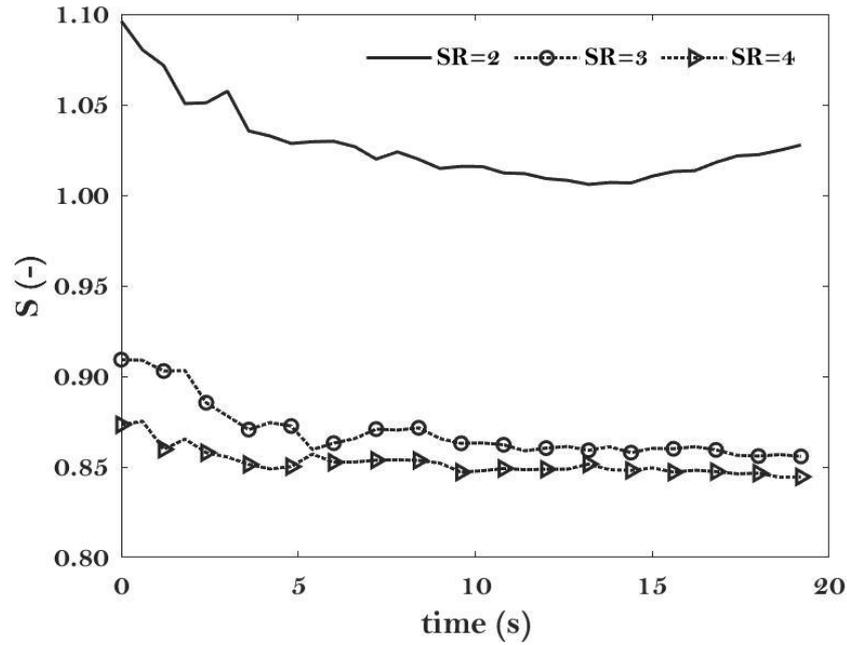


Fig. 18 Time evolution of the segregation index (S) as affected by size ratio for an initially layered configuration.

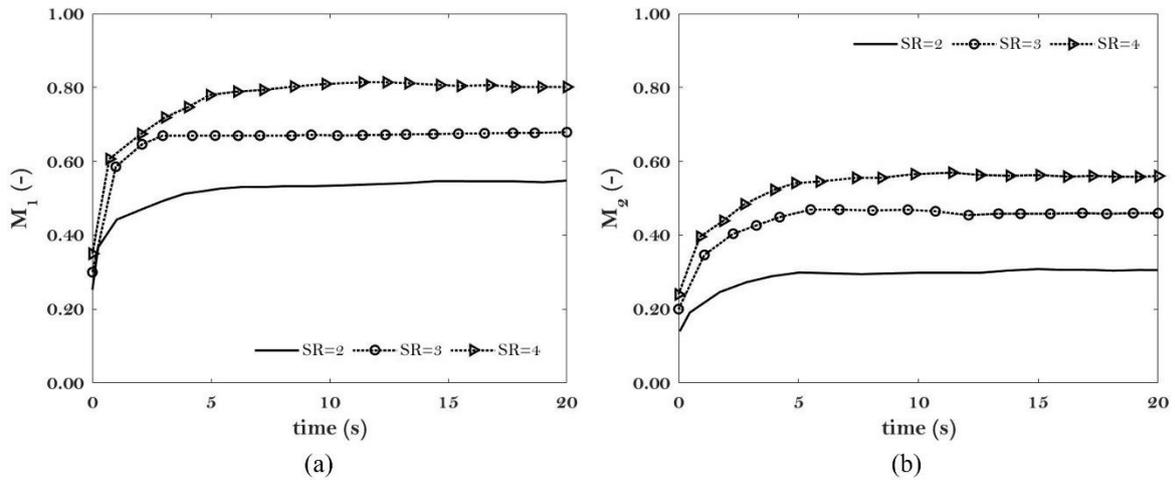


Fig. 19 Time evolution of the mixing indices (a) M_1 (Lacey's index) and (b) M_2 (Govender et al. [10]) as affected by size ratio for an initially layered configuration.

The relative percolation velocity in Eq. 16 is plotted in Fig. 21. In this case the velocity exhibits positive values, meaning that the centre of mass of fine particles compared to the coarse particles and the overall bed expansion is moving towards the top of the system. This is expected in this case because in the initial configuration all the fine particles are located on the top of the coarse and even if there is percolation, the fine centre of mass will always be on top

of the coarse particles. However, it can be observed that the relative velocity is lower as the size ratio increases, meaning that the centre of mass of fine particles is expanding towards the top with a lower velocity when the size ratio is higher, which can only be associated with percolation phenomena since the swelling kinetic is the same. Interestingly, in this case, the relative percolation velocity shows an initial jump, indicating that the centre of mass of fine particles moves to the top faster in the first seconds of swelling, and then it decreases due to percolation phenomena until it reaches a steady state value which is affected by the size ratio.

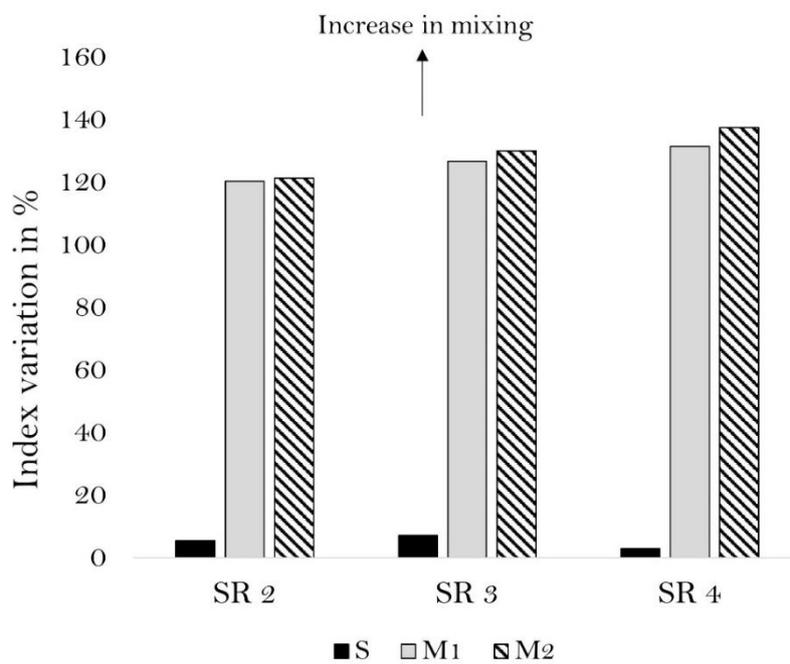


Fig. 20 Impact of the size ratio on the index variation after 20 s of swelling for an initially layered system

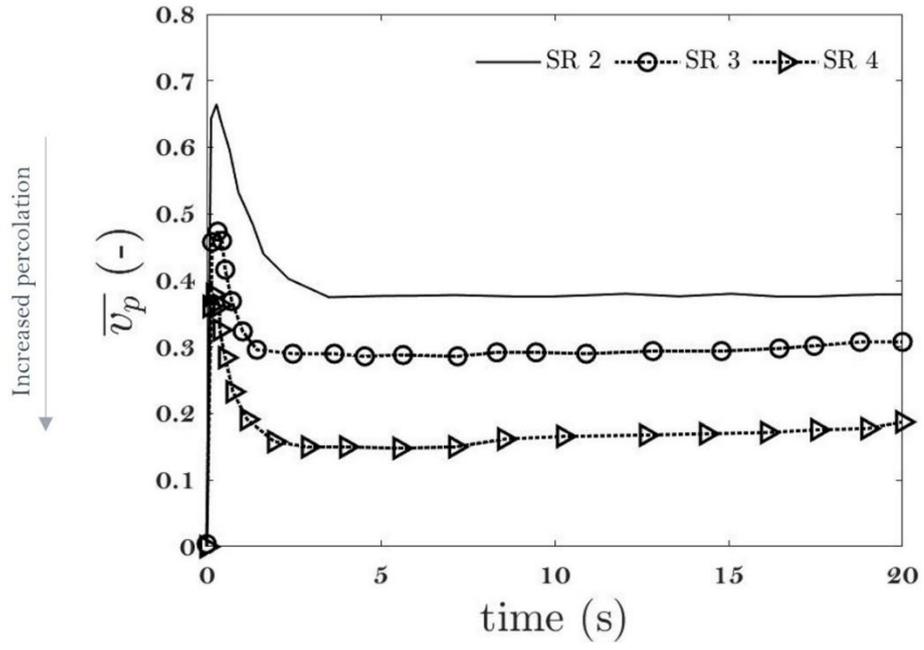


Fig. 21 Relative percolation velocity as affected by size ratio for an initially layered configuration.

4. Conclusions

The numerical analysis of the segregation and mixing tendency during swelling using several indices and a relative percolation velocity showed that the segregation index, which is evaluated based on the coordination number is not a reliable parameter for comparing systems where the ratio between the number of fine and coarse particles changes. For such systems, the mixing indices proposed by Lacey [9] and Govender *et al.* [10] were found suitable to describe the mixing and segregation tendencies. Additionally, the relative percolation velocity was demonstrated to be an effective parameter to examine the percolation of fines during swelling. It is found that when the granular bed swells, fine particles tend to experience percolation phenomena leading to segregation when systems are initially mixed and to improved mixing when systems are initially segregated. It was also found that as the particle size ratio increases, the percolation of particles intensifies. This observation holds true for initially layered systems which experience higher percolation and mixing as the size difference of the binary mixture increases. Swelling kinetics showed no impact on the percolation behaviour of the systems as the simulations indicated that different swelling rates did not significantly affect the segregation of the granular materials. However, in this work, the swelling kinetic was set to the same value for fine and coarse particles, nevertheless, that is not always the case in swelling granular systems. Indeed, several common applications employ swelling mixed with non-swelling components or mixtures in which the components swell with different kinetics. Future research can build upon these findings to explore additional factors, including systems with more than two components or components swelling with different kinetics.

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Data availability statement

All the numerical data from Figures 2, 4, 7-10, 12-15, 18-20 are evaluated based on the simulation outputs contained in the .zip file. The remaining figures are visualisation of the files contained in the same .zip file. This .zip file includes a folder for each of the section of this paper containing particles and contacts information data, and an input file which contains all the information on the swelling implementation and other simulation settings that were used in our LIGGGHTS calculations.

References

- [1] J. H. Perez, F. Tanaka, and T. Uchino, "Comparative 3D simulation on water absorption and hygroscopic swelling in japonica rice grains under various isothermal soaking conditions," *Food Research International*, vol. 44, no. 9, pp. 2615–2623, 2011, doi: 10.1016/j.foodres.2011.05.003.
- [2] T. Sweijen, B. Chareyre, S. M. Hassanizadeh, and N. K. Karadimitriou, "Grain-scale modelling of swelling granular materials; application to super absorbent polymers," *Powder Technol*, vol. 318, pp. 411–422, 2017, doi: 10.1016/j.powtec.2017.06.015.
- [3] T. Santagata, R. Solimene, G. Aprea, and P. Salatino, "A Grain-Scale Study of Swelling Composite Porous Media Made of Fibres and Particles," in *the 30th European Symposium on Computer Aided Process Engineering*, 2020.

- [4] J. Horabik and M. Molenda, "Grain pressure in a model silo as affected by moisture content increase," *Int. Agrophysics*, vol. 14, pp. 385–392, 2000.
- [5] S. Volpato, M. Tirapelle, and A. C. Santomaso, "Modeling and experimental investigation of shear-induced particle percolation in diluted binary mixtures," *Phys Rev E*, vol. 102, no. 1, pp. 1–8, 2020, doi: 10.1103/PhysRevE.102.012902.
- [6] Z. Yan, S. K. Wilkinson, E. H. Stitt, and M. Marigo, "Investigating mixing and segregation using discrete element modelling (DEM) in the Freeman FT4 rheometer," *Int J Pharm*, vol. 513, no. 1–2, pp. 38–48, 2016, doi: 10.1016/j.ijpharm.2016.08.065.
- [7] C. Kloss, C. Goniva, A. Hager, S. Amberger, and S. Pirker, "Models, algorithms and validation for opensource DEM and CFD-DEM," *Progress in Computational Fluid Dynamics*, vol. 12, no. 2–3, pp. 140–152, 2012, doi: 10.1504/PCFD.2012.047457.
- [8] D. Braile, "DEM Analysis of swelling behaviour in granular media," *Advanced Powder Technology*, vol. 33, no. 11, p. 103806, 2022, doi: 10.1016/j.appt.2022.103806.
- [9] P. M. C. Lacey, "Developments in the theory of particle mixing," *Journal of Applied Chemistry*, vol. 4, no. 5, pp. 257–268, 1954, doi: <https://doi.org/10.1002/jctb.5010040504>.
- [10] N. Govender, R. Kobyłka, and J. Khinast, "The influence of cohesion on polyhedral shapes during mixing in a drum," *Chem Eng Sci*, vol. 270, p. 118499, 2023, doi: 10.1016/j.ces.2023.118499.
- [11] M. J. Khala, C. Hare, C. Wu, N. Venugopal, M. J. Murtagh, and T. Freeman, "Density and size-induced mixing and segregation in the FT4 powder rheometer: An

experimental and numerical investigation,” *Powder Technol*, vol. 390, pp. 126–142, 2021, doi: 10.1016/j.powtec.2021.05.027.

- [12] S.-H. Chou, Y.-L. Song, and S.-S. Hsiau, “A Study of the Mixing Index in Solid Particles,” *Kona Powder and Particle Journal*, vol. 34, p. 2017018, 2017, [Online]. Available: <https://api.semanticscholar.org/CorpusID:138186385>