SIMULATION OF POWDER RHEOLOGICAL BEHAVIOUR IN THE CERAMIC TILE PRESSING PROCESS

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1. ABSTRACT

Although the discrete element method (DEM) has been successfully used to simulate the flow of particulate materials in different industrial processes, the die filling procedures used in numerous compaction processes have drawn little attention. The die filling step before compaction by pressing of ceramic tile bodies needs to be appropriately performed to assure the end product's targeted physico-chemical properties. This study fine-tunes a constitutive model, based on the DEM, which allows the spray-dried powder feed procedure into the press die to be studied.

2. INTRODUCTION

In recent years, numerous studies have been conducted on the ceramic tile body pressing process. These have focused fundamentally on the forming process, neglecting the die cavity filling process [1].

At present, all control and optimisation actions performed during die filling are done on the basis of operator experience, there being no systematised knowledge in this regard. This approach entails limitations, which lead to heterogeneous distribution of the quantity of deposited powder both inside a given die cavity and across different cavities in the die. When an axial compression force is applied, these heterogeneities produce a heterogeneous pressure distribution on the powder that, on giving rise to bulk density differences in the freshly pressed bodies, can subsequently lead to defects such as fracture, low mechanical strength, uncontrolled firing shrinkage or deformation, and lack of dimensional stability [2].

Understanding the physical phenomena and mechanisms involved on a microscopic level in the press powder deposition process in the die cavities requires modelling the rheological behaviour of the powder. The methodology used for this purpose in the present study was the discrete element method (DEM). This method stems from Molecular Dynamics, formulated at the end of the 1950s during the development of the theoretical physics involved in studying atomic energy [3]. Years later, the basic scheme of this technique was adapted to the field of soil mechanics in civil engineering, with a view to simulating and analysing the behaviour of rock masses. The adaptation was originally performed in 1971 by Peter A. Cundal [4] who, given the success of his idea, eventually published the formal structure of the DEM and its direct application to real problems in 1979 [5] [6].

Over the last fifteen years, the DEM has been widely used in studying the mechanisms that govern the behaviour of particulate materials, such as powder flow during silo discharge [7], filling and particle packing [8], storage pile formation [9], compression behaviour [10], and mixing and transport [11]. In particular, it may be noted that, in the pharmaceutical industry, the DEM has led to a precise understanding of the influence of the manufacturing process parameters that directly affect tablet quality, such as the influence of the filling rate and the geometrical shape of the die on granule size segregation.

3. DEM SIMULATION MODEL

The DEM approach describes a system of N particles that represent each of the constituent bodies of the granular medium. In the case at issue here, the medium corresponds to the press powder, and the constituent bodies are the granules. The dynamic evolution of the whole takes place by the simultaneous resolution of every Newtonian equation of motion associated with each granule, providing complete information on the state of the powder at each moment and in any region of the spatial domain through which it extends. The overall mechanical and rheological behaviour of the powder is mainly determined by the intergranular interaction mechanisms and the interaction between the granules and the medium in which they are immersed. The main implications of the fine-tuned physical simulation model and the physical parameters involved are described below.

3.1. DEVELOPMENT AND IMPLEMENTATION OF THE PHYSICAL MODEL

The interaction between any two bodies is formulated from a so-called constituent model. Such a model is a mathematical representation that contains all the physical phenomena determining the values of the forces and the mechanical moments that will appear in both bodies owing to the interaction. Although there is no conceptual restriction when it comes to formulating the constituent model in the DEM, it is common practice to use a series of approximations, combining the following micro-rheological contributions:

- Elastic contribution. This provides the elastic repulsion that appears between two bodies when they enter into contact. A perpendicular contribution with respect to the plane defined by the contact area is distinguished, associated with the compression strain, in addition to a tangential contribution, associated with the shear. Although both reactions can be formulated in a non-linear way (Hertzian elastic contact), for the sake of simplicity, in this study, a linear model based on Hooke's law, known as the Linear Spring-Dashpot (LSD) was used [12].
- Friction contribution. This describes the level of friction that appears when two granules rub against each other. The friction associated with the translation displacements (slip resistance) and the friction associated with the relative rotation of one granule with relation to the other (rotation resistance) are differentiated. This microscopic element is one of the factors that determine the macroscopic yield capacity of a powder [13].
- Adhesive contribution. This describes the tendency of the granules to join together spontaneously, the intensity of which depends on the distance between their surfaces. In press powder, the maximum adhesive contribution is due to water. In this study, to facilitate the obtainment of the parameters, dry powder was used so that intergranular adhesion was disregarded.
- Viscous contribution. This allows the physical processes involving energy dissipation on a microscopic level to be incorporated. In the proposed model, only the viscosity stemming from intergranular collisions in the normal direction was considered.

The scheme in Figure 1 shows the geometric magnitudes involved in the constituent model used. Given the high sphericity of the spray-dried powder granules, these were assumed to be spherical. When two neighbouring granules i and j, of diameters d_i and d_j, respectively, came sufficiently close to each other, they were able to interact producing a force $\vec{F}_{ij}$ and a moment $\vec{\Gamma}_{ij}$ at an initial contact point. In this type of simulation, material deformation is not modelled as such; rather, the magnitude of the interpenetration produced between both geometries, h_{ij}, is a quantity from which granule deformation can be determined, which is used to define the constituent model of interaction.

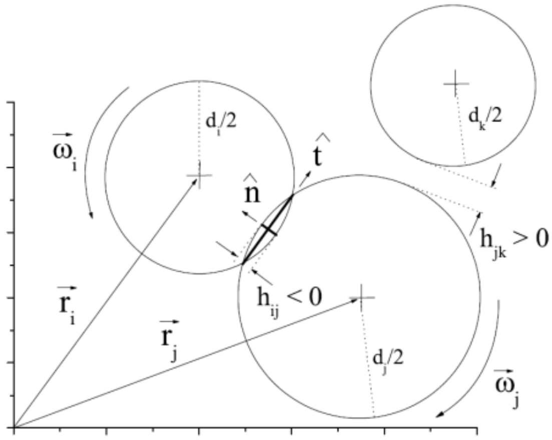

Figure 1. *Geometric magnitudes of the constituent model of interaction in the DEM.*

The state of the powder in a certain region of the space and at a certain moment of time is given by the set of spatial coordinates $\vec{r}_i$ and angular coordinates $\vec{\theta}_i$ with $i = 1,\ldots,N$, where N is the total number of granules in the system. The temporal evolution of these quantities is governed by Equations 1 and 2. In these equations, with relation to granule i, the sum extends to all granules j with which this interacts, and where the quantities m_i and I_i correspond to mass and moment of inertia, respectively, and $\vec{F}_i^S$ and $\vec{\Gamma}_i^S$ to force and moment owing to the interaction with the powder's confining context.

Equation 1. $$m_i \frac{d^2\vec{r}_i}{dt^2} = \sum_j \vec{F}_{ij} + \vec{F}_i^S$$

Equation 2. $$I_i \frac{d^2\vec{\theta}_i}{dt^2} = \sum_j \vec{\Gamma}_{ij} + \vec{\Gamma}_i^S$$

From a mathematical viewpoint, as the analytical and simultaneous solution of the strongly coupled N equations of motion is impracticable, the evolution of the system was solved by means of an integrating algorithm, specially developed in C++. In addition, given the great number of equations that needed to be solved in each time step, a parallel calculation strategy was envisaged.

3.2. PHYSICAL PARAMETERS OF THE MODEL

For the sake of simplicity, the equations constituting the LSD model used in implementing the simulation are not described here. For further details, these equations can be consulted in the literature [14]. The main physical parameters associated with the rheological behaviour of the powder were as follows:

- Stiffness coefficients for contacts in the normal and tangential direction, k_n and k_t, respectively. At a physical level, these parameters indicate how hard or soft the granules were in the powder constituent model when they hit each other during the material flow. High values of K_i (N/m) corresponded to materials made up of hard particles, which displayed little interpenetration at contact. Very low stiffness coefficient values were associated with extremely soft particles that exhibited large deformation upon contact and, therefore, a high degree of overlapping.
- Damping coefficient, η_n. Expressed as (N.s/m), this indicates the degree of conservation of energy that the granules displayed when they hit each other. It is usually expressed as a fraction of the so-called coefficient of critical restitution. For the sake of simplicity, the model was only applied in the normal direction.
- Intergranular friction coefficient, μ_{gg}. This is directly related to the internal friction angle of the material, this being a measure of its flowability. Highly cohesive powders exhibit high internal friction angles and, therefore, high inter-particle friction coefficients.
- Friction coefficient between granules and surfaces, μ_{gs}. This is equivalent to the friction angle of the material with the elements it is in contact with, and which consequently depends on the properties of these elements. In general, materials such as Teflon or stainless steel on which powders tend to exhibit little adhesion have small friction angles.
- Rolling resistence coefficient, μ_r. This describes the friction that appears when two granules enter into contact with a relative rotation different from zero.

4. MATERIALS AND EXPERIMENTAL PROCEDURE

Once the DEM simulation model had been developed, it was necessary to conduct a series of experiments to determine the physical parameters that would allow the rheological behaviour of the spray-dried powder to be reproduced. The study was conducted with a spray-dried powder of the type customarily used in porcelain tile manufacture. The powder was dry and its granule size distribution (GSD), obtained by dry sieving, is shown in Figure 2. The experimental procedure used in the different tests and the way in which the test results were related to the physical parameters of the model are briefly described below.

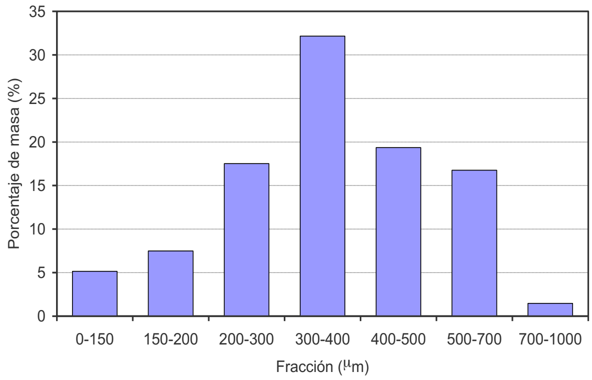

Figure 2. *Granule size distribution of the spray-dried powder used.*

4.1. COMPRESSION TESTS

To determine the stiffness coefficient of the spray-dried powder granules, compression tests were conducted on the spray-dried powder beds at low pressures [15]-[17]. To avoid the possible influence of humidity on the results, the powder was dried before testing in an oven at 120 ºC to constant weight. The tests consisted of filling a metal die like the one shown in Figure 3 with the test powder. Once the surface of the resulting bed had been levelled flush with the top of the die, it was subjected to a total of eight compression–decompression cycles with a metal punch coupled to the crosspiece of a universal testing machine.

Several compression–decompression cycles were performed to minimise the effect of the initial conditions resulting from manual filling of the die on the mechanical properties. Thus, the stress–strain curves corresponding to the last cycles conducted enabled the effective Young's modulus of the powder bed to be calculated as the quotient of the difference in the recorded pressure to the deformation undergone by the bed between the ends of each cycle. Using the DEM model, the stress–strain simulation curves at different values of the stiffness coefficient were then reproduced, with a view to determining which stiffness value provided a Young's modulus equal to the experimentally obtained one.

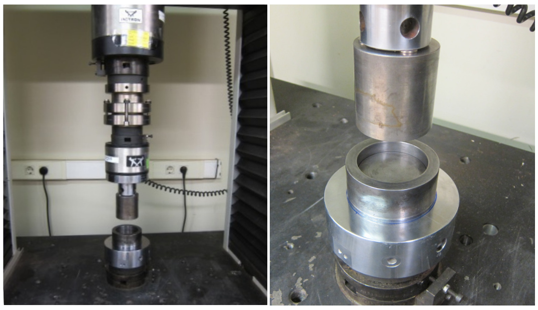

Figure 3. *General view and detail of the die in the experimental set-up used in determining the stiffness coefficient of the spray-dried powder.*

The pressure limits of the compression cycles were selected in a series of preliminary trials such that, at maximum pressure, no deformation of the spray-dried powder granules occurred, while at minimum pressure the granules were still in direct contact with each other. The load application rate was very low (1 mm/min). Consequently, the test was conducted in a quasi-steady state in which the powder constituent model was exclusively governed by the intergranular elastic contacts.

4.2. DETERMINATION OF THE FLOW CURVES AND FRICTION ANGLES

To determine the intergranular friction angles and friction angle between the granules and the surfaces, a rotational shear cell was used. Basically, the tests consisted of subjecting the powder bed to different compression pressures, while applying a shear stress to cause the material to flow. The plot of the shear–normal stress pairs of values, at which powder flow started, at different maximum consolidation stresses, constitutes the so-called flow curves of the material from which the internal angle of friction was obtained.

Following a similar procedure, it was possible to measure the angle of friction between the granules and the surface of a given material, putting the powder bed in contact with small plates of the material to be studied (Figure 4). For further information on these procedures, please see reference [18].

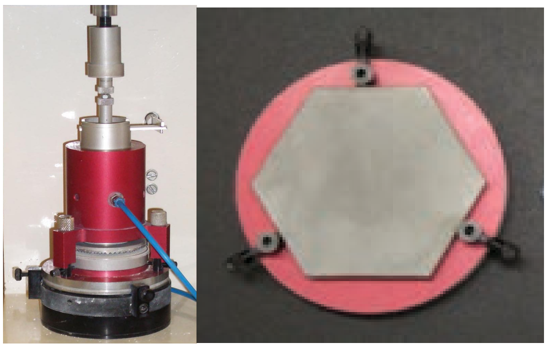

Figure 4. *Rotational shear cell and cell base with a steel plate for measurement of the friction angle with a surface.*

Figure 5. *Set-up used to measure the angle of repose.*

4.3. MEASUREMENT OF THE ANGLE OF REPOSE

To optimise the coefficients of friction and to evaluate the performance of the simulation model under defined conditions of flow, tests were carried out to measure the spray-dried powder angle of repose. The set-up shown in Figure 5 was used for this purpose. The assembly consisted of a top glass funnel into which the material to be characterised was poured to make it pass through a manually operated ball valve to a second bottom glass funnel from which the powder was emptied through a hatch on a flat surface set on the base. The assembly also had a video graphic system for recording the formation of the little heap of powder in order subsequently to determine the angle of repose by image analysis.

5. RESULTS AND DISCUSSION

5.1. PARAMETRIC FITTING OF THE PHYSICAL MODEL

The results obtained in the tests and simulations that allowed the parameters of the implemented physical model to be fitted are described below.

5.1.1. DETERMINATION OF STIFFNESS

Figure 6 shows the experimental pressure–strain curve corresponding to a compression test. As may be observed, in the first compression cycle, owing to granule reordering in the die, bed deformation was much greater than in the other test cycles. As the test proceeded, the deformation increased slightly; however, in the last cycles, it remained practically constant. This confirmed that in the last compression cycle, the resulting stress–strain curve was representative of the interaction between the granules, and the calculated Young's modulus was associated with their elastic deformation. After performing this experiment in triplicate, and determining the Young's modulus of the last compression cycle, an average Young's modulus of 45,4 ±1,5 MPa was obtained for the dry spray-dried powder.

Knowing the Young's modulus of the spray-dried powder, the described compression experiment was simulated on a computer, using the DEM model, to obtain the theoretical stress–strain curves corresponding to the system's stiffness values from 250 N/m to 10 000 N/m. Figure 7 shows, from left to right, the evolution of one of the simulations.

By way of example, Figure 8 shows the curves resulting from the simulations made at two extreme stiffnesses, which were in agreement with what was observed in [17]. It shows that granule degree of deformation decreased significantly as the simulated stiffness value increased. To save calculation time, not all experimental cycles were simulated. This did not affect the results as the simulations were based on a random granule distribution, which was in itself already more ordered than a distribution that could be achieved in the experiments performed.

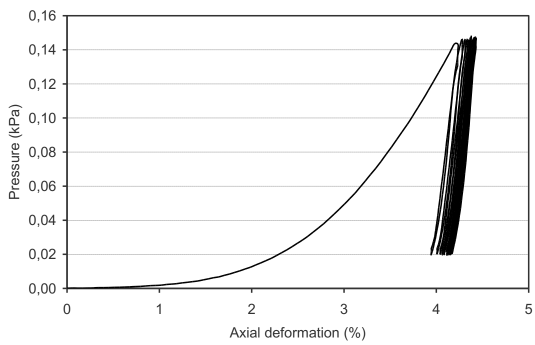

Figure 6. *Stress–strain curve for determining system stiffness.*

Calculating the Young's modulus from each simulation enabled a graph to be made like the one shown in Figure 9, which plots the variation of the simulated Young's modulus as a function of the stiffness of the spray-dried powder introduced in the model.

Although the data obtained fitted quite well to a power curve, it was observed that, at the stiffness values used in the calculations, the experimentally determined Young's modulus was not reached. That is, in practice the spray-dried powder granules were even harder than in the simulation made with a stiffness of 10 000 N/m.

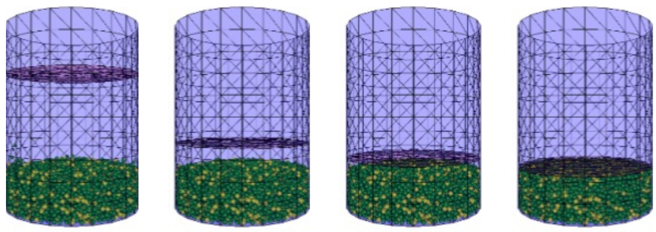

Figure 7. *DEM simulation of a compression test.*

This entailed a setback in the development of the model, to the degree in which system stiffness enormously limited simulation speed. Indeed, extrapolating the curve shown in Figure 9 to the value of the experimentally obtained Young's modulus required a system stiffness of 87.10^3 N/m to enable powder behaviour to be reproduced.

To work with such high stiffness involves integration time steps of 10^{-7}s, which make use of the model unfeasible, even when using equipment with a high calculation capability. It was therefore decided to adopt a different strategy, setting the system stiffness value at the stiffness value above which the number of contacts during a compression test, expressed as a coordination number, remained constant. DEM simulations were thus made modifying the stiffness and determining the evolution of the average coordination number of the granules, as the compression test progressed.

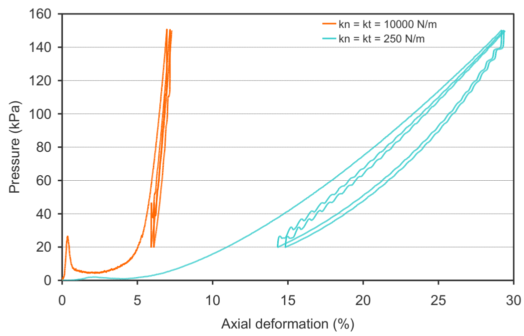

Figure 8. *Simulated stress–strain curves.*

Figure 10 shows how the coordination number of the granules in the bed varied in the simulations, as granule stiffness increased. It was observed that at stiffness values above 400 N/m the coordination number remained constant.

This indicated that, above this value, the number of granules with which a given granule interacted was not affected by the deformation they underwent. Finally, this stiffness limit value was the value kept to perform the calculations..

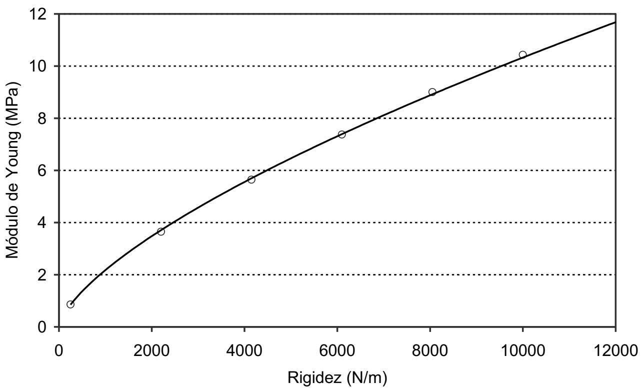

Figure 9. *Young's modulus calculated in the simulations, as a function of stiffness.*

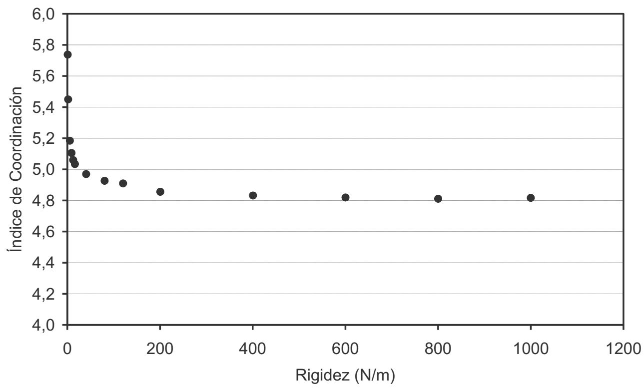

Figure 10. *Variation of the coordination number of the granules with stiffness.*

5.1.2. DETERMINATION OF THE FRICTION ANGLES

With relation to the characteristic friction angles, Figure 11 shows the flow curves obtained for the dry spray-dried powder, from which an internal spray-dried powder friction angle of 31,5º was determined. With regard to the powder angle of friction, this was evaluated for two types of surfaces: on the one hand for an AISI 304 steel, which displayed an angle of 26º, and on the other for a float glass, which exhibited an angle of 25º.

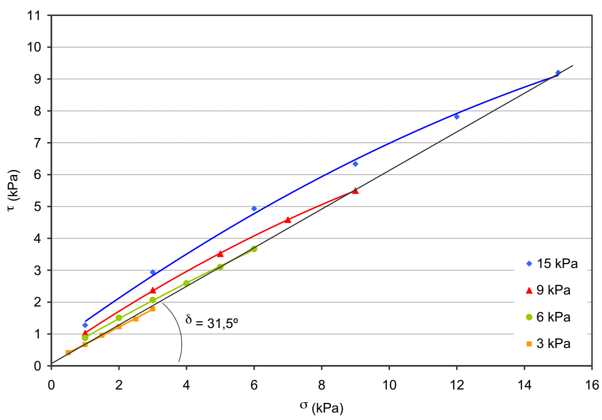

Figure 11. *Flow curves of the dry spray-dried powder.*

5.1.3. FITTING ROLLING RESISTENCE COEFFICIENT

Finally, having established system stiffness and the characteristic friction angles, several tests were performed of the angle of repose to determine the resistance to rotation in the model. Following the procedure described in point 0, images like the one shown in Figure 12 were obtained, from which the material's angle of repose was calculated. The test was then simulated at different rolling resistence values of the physical model. A plot is shown in Figure 13 of the variation of the angle of repose calculated with the model as a function of the rolling resistence coefficient used.

The fitting curve shows that the obtainment of an angle of repose of 32º, just as the one determined experimentally, was satisfied with a rolling resistence coefficient of 0,19.

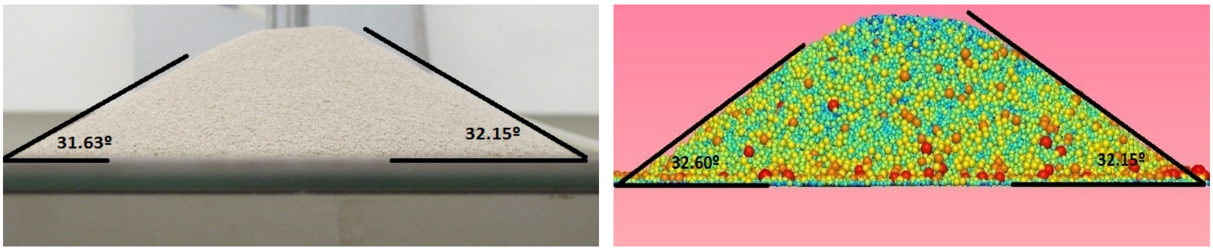

Figure 12. *Angle of repose obtained experimentally and by DEM simulation.*

It may be noted that, given the small size of the granules making up the spray-dried powder, it was impracticable to simulate, in reasonable times, the number of grains involved in any test or industrial process. For that reason, the model was completed with a series of scale laws that kept the properties of the simulated bed constant despite using fewer granules [19]. The appropriateness of this scale-up was reflected, for example, in the presence of granule size segregations in the simulated little heap or in the reproduction of the pattern of tubular discharge in the experimental set-up funnel.

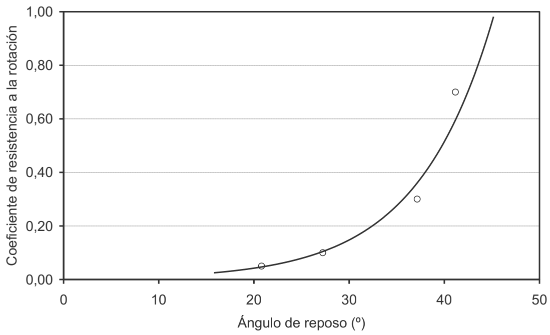

Figure 13. *Variation of the rolling resistence coefficient with the angle of repose..*

5.2. PRACTICAL EXAMPLE

Figure 14 shows powder behaviour during the filling of a metal cavity measuring 30 cm x 60 cm. The granules are coloured as a function of their size, which allows the appearance of the filling patterns at the charge end to be qualitatively observed. Studying these patterns together with the distribution of the densities in the bed is of interest in order to identify the most important parameters that influence the filling process, as well as to see how system geometry affects the properties of the resulting bed.

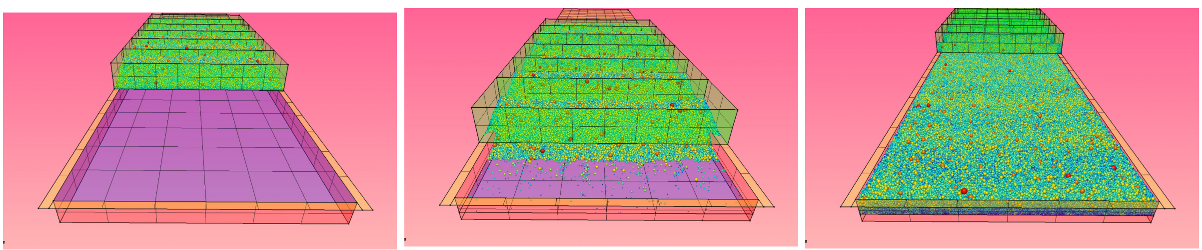

Figure 14. *Press cavity filling sequence simulated with the DEM model.*

6. CONCLUSIONS

This paper describes the experiments conducted to determine the physical parameters of a DEM model specially developed to reproduce the rheological behaviour of press powder. Although further experiments are required to confirm the satisfactory operation of the model, its use opens the way to a systematic study of the die feed operation. In future studies it is intended to pay special attention to the effect of the different operating variables and of powder properties on powder filling patterns and distribution in the die. This will require fine-tuning a method for analysing the density distributions that allow the properties of the resulting beds to be related to the operating conditions and construction form of the feed system.

REFERENCES

[1] Mallol, G. Control y automatización en la industria cerámica. Evolución y perspectivas. Cerámica Información, 347, 63-80, 2007

[2] Amorós, J.L.; Mallol, J.G.; Mezquita, A.; Llorens, D.; Castro-Lopes, F.; Cerisuelo, J.A.; Vargas, M. Mejora de la estabilidad dimensional de piezas de gres porcelánico a través de la medida en contínuo de la humedad de los soportes prensados. *Cerámica Información,* 311, 117-126, 2004

[3] Alder, B.J. and Wainwright, T.E.: Studies in Molecular Dynamics. I. General Method. Journal of Chemical Physics, Vol. 31(2), pp.459-466 (1959).

[4] Cundall, P.A.: A computer model for simulating progressive, large-scale movements in blocky rock systems. Proc. of the Symposium of the International Society of Rock Mechanics, Vol. 1, paper No II-8, pp.132-150 (1971).

[5] Cundall, P.A. and Strack, O.D.L.: A discrete numerical method for granular assemblies. Géotechnique, Vol. 29(1), pp.47–65 (1979).

[6] Cundall, P.A., Drescher, A. and Strack, O.D.L.: Numerical experiments on granular assemblies, measurements and observations. P.A. Vermeer, H.J. Luger (Eds.), Deformation and failure of granular materials, Balkema, Rotterdam, pp.355–370 (1982).

[7] Balevîcius, R., Kâcianauskas, R., Mroz, Z., Sielamowicz, I.: Discrete element method applied to multiobjective optimization of discharge flow parameters in hoppers. In: Structural and Multidisciplinary Optimization, pp. 163–175. Springer, London (2006).

[8] Zhang, Z.P., Liu, L.F., Yuan, Y.D., Yu, A.B: A simulation study of the effects of dynamic variables on the packing of spheres. Powder Technol. 116, 23–32 (2001)

[9] Buchholtz, V. (Doctoral dissertation): Molekulardynamische Untersuchungen granularer Stoffe. Humboldt-University zu Berlin (1994).

[10] Odagi, K., Tanaka, T., Yamane, K.: DEM simulation of compression test of particles. In: Proceedings of World Congress on Particle Technology, Vol.4, Sydney (2002).

[11] Buchholtz, V., Pöschel, T., Tillemans, H.J.: Simulation of rotating drum experiments using noncircular particles. Physica A, Vol.216, pp.199–212 (1995).

[12] Cummins et al.: Contact force models in inelastic collisions. Ninth International Conference on CFD in the Minerals and Process Industries, Australia (2012).

[13] Zhang, Z.P., Liu, L.F., Yuan, Y. D. and Yu, A. B.: A simulation study of the effects of dynamic variables on the packing of spheres. Powder Technology, Vol.116, pp.23-32 (2001).

[14] Luding, S. Introduction to Discrete Element Methods. Basics of Contact Force Models and how to perform the Micro-Macro Transition to Continuum Theory. EJECE. Discrete modelling of geomaterials (2008) 785-826

[15] C.J Coetzee, D.N.J. Els*. Calibration of discrete element parameters and the modelling of silo discharge and bucket filling*. Computers and Electronics in Agriculture 65 (2009) 198-212

[16] Johanstone, M.W. Calibration of DEM models for granular materials using bulk physical tests. Thesis. University of Edinburgh (2010)

[17] Horn, E. The Calibration of Material Properties for Use in Discrete Elements Models. Thesis, University of Stellenbosch (2012)

[18] Amorós, J.L.: Study of the rheological behaviour of different ceramic powder materials, Part 1. Interceram, 57 (4), 236-239, 2008

[19] Sakai, M. et al. Verification and validation of a coarse grain model of the DEM in a bubbling fluidized bed. Chemical Engineering Journal 244 (2014) 33-43