

UNIVERSITY OF DEBRECEN
Faculty of Engineering
Department of Mechanical Engineering



PROCEEDINGS OF THE
5th INTERNATIONAL SCIENTIFIC CONFERENCE ON
ADVANCES IN MECHANICAL ENGINEERING
(ISCAME 2017)

12-13 October, 2017

Debrecen, Hungary

organized by

Department of Mechanical Engineering
Faculty of Engineering, University of Debrecen

and

Working Commission in Mechanical Engineering
Specialized Committee in Engineering
Regional Committee in Debrecen, Hungarian Academy of Sciences



INTRODUCTION OF FREEZE GRANULATION TECHNOLOGY

POÓS Tibor PhD, SZABÓ Viktor, VARJU Evelin, HORVÁTH Dániel

Department of Building Services and Process Engineering, Budapest University of Technology and Economics

E-mail: poos@mail.bme.hu, szabo.viktor@mail.bme.hu, varjuevelin@mail.bme.hu, daniel.horvath.nk@gmail.com

Abstract

Granulation is a widely used process in diverse sectors of the industry, one of them is freeze granulation. Besides pharmaceutical applications (eg. vitamins, vaccines, inhalers), this technology is often used in the chemical industry (eg. environmentally friendly batteries), the ceramic industry (powder sintering) and the food industry (eg. coffee powder). The aim of our work is gathering knowledge about the process and the equipment of freeze granulation and to compare it with other granulator processes on the basis of literature. Subsequently, we will propose a design of a freeze-granulator measuring station (main elements and instruments) at the Department of Building Services and Process Engineering of the Budapest University of Technology and Economics, which could be used for researches and student measurements. The spray-drying equipment in the departmental laboratory can be used later to compare its results to our measuring station's.

Keywords: *freeze granulation, spray dryer, literature review, measurement*

1. INTRODUCTION

Granulation may be defined as a size enlargement process which converts fine or coarse particles into physically stronger and larger agglomerates having good flow property, better compressibility of powders, improve content uniformity and eliminate excessive amounts of fine particles. Granulation is one of the most important unit operation in the production of pharmaceutical granules, ceramics, and foodstuffs. Particle size of the granules is mainly affected by the quantity and feeding rate of granulating liquid. Pharmaceutical granules typically have a size range between 0.2 and 4.0 mm, depending on their subsequent use [1].

Generally, there are three different method of granulation:

- Dry granulation.
- Direct compression.
- Wet granulation.

Wet granulation that utilize a liquid in the process and dry granulation that requires no liquid. The type of process selection requires thorough knowledge of physicochemical properties of the material, and required flow and release properties. Among currently available technologies, spray drying, freeze granulation, and fluid bed granulation are worth of note [2]. During spray drying, a powder suspension is sprayed into a hot air drying chamber. During drying, the droplets shrink to form powder granules. These granules may be more or less spherical and/or hollow depending on the slurry formulation (i.e. powder load and additives) and drying temperature [3]. Binder migration during the drying can additionally reduce the homogeneity and thereby quality of the granules, leading to inhomogeneities of the particles [4].

Freeze granulation is an alternative to spray drying, solving many of its limitations. During freeze granulation, droplets are rapidly frozen in liquid nitrogen before being recovered and subsequently



freeze-dried. Freezing being a fast process, diffusion dynamics are insufficient to induce any binder migration, thereby better preserving the granule homogeneity [5]. Additionally, the granules do not shrink during the whole process and therefore maintain their shape and powder density — consequently allowing higher granule deformability compared to spray drying.

Besides the high degree of granule homogeneity, freeze granulation technology offers several other advantages [6]:

- Control of granule density by the solids content of the suspension.
- Mild drying prevents serious oxidation of non-oxides and metals.
- No cavities in the granules.
- Low material waste, high yield.
- Small (50 – 100 ml suspension) as well as large granule quantities can be produced to equal quantity.
- Easy clean of the equipment (latex binder can be used).
- Possibility to recycle organic solvents.

2. FREEZE GRANULATION TECHNOLOGY

Freeze granulation technology is the combination of spray freezing and freeze drying. *Figure 1* shows an illustration of the process. Spray freezing is the first step in the freeze granulation process where a suspension is atomized, sprayed with a nozzle into a chamber with liquid nitrogen, with temperature around -196°C . The small droplets are then instantaneously frozen with a minimum of ice crystal growth. The frozen material (granules) are then transferred to a freeze dryer where the ice is removed by sublimation, i.e. never go through a liquid state that ensure homogeneity preservation without any segregation effects as in the case of conventional drying with air. The temperature in the freeze dryer is about -30°C [7].

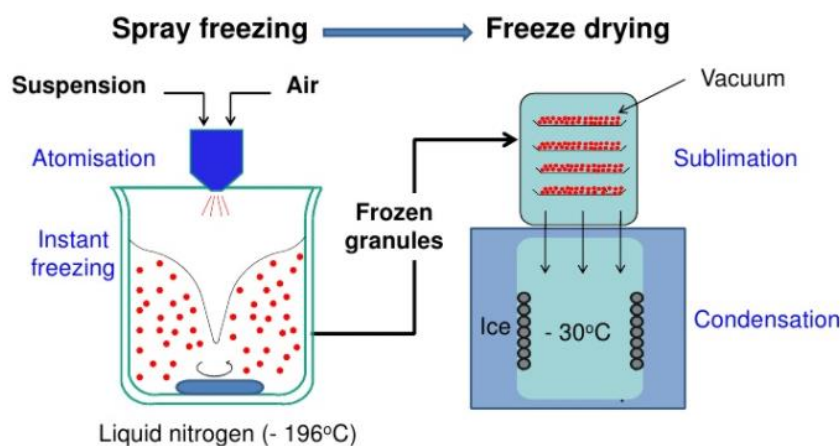


Figure 1 Illustration of the freeze-granulation process [8]

The granule size distribution will be controlled by the suspension rheology (flow properties) and the process parameters (pump speed and air pressure). Normally, a certain size distribution-width is achieved with an average size, typically, around $100\text{--}200\text{ }\mu\text{m}$. In a comparison between the sintering performance of pressed specimens using granules obtained by freeze granulation or spray-drying the effect of homogeneity is clearly illustrated in *Figure 2*.

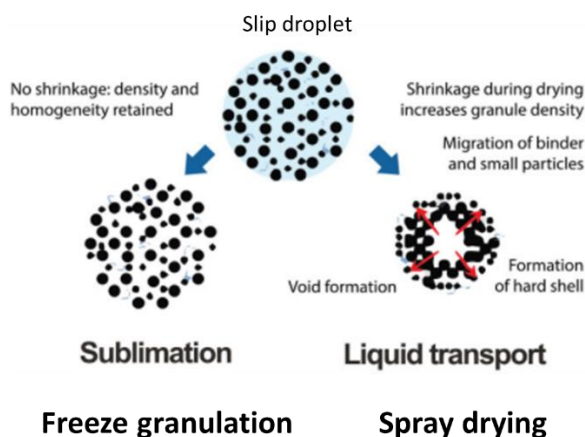


Figure 2 Comparison of freeze granulation and spray drying [8]

Several companies and research labs around the world have applied the freeze granulation process. Typical ceramic powders are oxides (Al_2O_3 , ZrO_2 , and SiO_2), nitrides (Si_3N_4) and carbides (SiC), but also nanopowders, diamonds and pharmaceuticals like proteins and enzymes [9].

3. METHODS

A full-scale plant spray dryer device is available in the laboratory of Department of Building Services and Process Engineering. Figure 3 shows a photo of the apparatus.



Figure 3 Spray dryer equipment

The spray dryer device is used for laboratory measurements. During the measurements, coffee suspension is atomized to drops, than dried in a drying chamber. The apparatus is equipped with instruments, e.g. thermometers, air humidity meters and pressure-difference meters. The atomizer rotates with 36000 1/min, the diameters of the coffee droplets are between 1 – 600 μm . The quality of the drops made with the spray dryer and with the planned freeze granulator, and their operational parameters will be compared. Our work is still in the literature review state. Our work is still in the literature review state.



CONCLUSIONS

Freeze granulation gives spherical, free-flowing granules. The good microstructure and homogeneous distribution of the granules enhance the sintering performance. The method should be well suited for composite systems, pharmaceutical use, or other fields of consumption where a uniform distribution and high quality of components required. The purpose of our work is to research the actual literature of this technology, than install a pilot-plant apparatus, which is suitable for pharmaceutical application. Our future plan is to compare the freeze granulation with the spray drying manufacturing the same type of material, which can be the basis of the scaling up this technology. The impact of operational parameters will be examined. Developing a mathematical model provides better economical operations and higher productivity. As an additional benefit, freeze granulation technology can be integrated to the education, which field is still less elaborated.

ACKNOWLEDGEMENTS

This paper was supported by Gedeon Richter's Talentum Foundation (H-1103 Budapest, Gyömrői str. 19-21, Hungary), and by the Hungarian Scientific Research Fund (NKFIH/PD-116326).

REFERENCES

- [1] Shinde, N., Aloorkar, N., Kulkarni, A., Bangar, B., Sulake, S., Kumbhar, P.: *Recent Advances in Granulation Techniques*. Asian Journal of Research in Pharmaceutical Science, 4 38–47., 2014.
- [2] Shanmugam, S.: *Granulation techniques and technologies: recent progresses*. BioImpacts : BI, 5 (1), 55–63., 2015.
- [3] Walker, W.J.J., Reed, J.S., Verma, S.K.: *Influence of Slurry Parameters on the Characteristics of Spray-Dried Granules*. Journal of the American Ceramic Society, 82 (7), 1711–1719., 1999.
- [4] Hidber, P., Graule, T.J., Gauckler, L.J.: *Competitive Adsorption of Citric Acid and Poly(vinyl alcohol) onto Alumina and Its Influence on the Binder Migration during Drying*. Journal of the American Ceramic Society, 78 (7), 1775–1780., 1995.
- [5] Lyckfeldt, O., Rundgren, K., Sjöstedt, M.: *Freeze Granulation for the Processing of Silicon Nitride Ceramics*. Key Engineering Materials - KEY ENG MAT, 264 281–284., 2004.
- [6] Solanki, H.K., Basuri, T., H.Thakkar, J., Patel, C.A.: *Recent Advances in Granulation Technology*. International Journal of Pharmaceutical Sciences Review and Research, 5 (3), 48–54., 2010.
- [7] Rundgren, K., Lyckfeldt, O.: *Development of Water Based Processing of Silicon Nitride Materials*. 26th Annual Conference on Composites, Advanced Ceramics, Materials, and Structures: A: Ceramic Engineering and Science Proceedings, 23 2–10., 2008.
- [8] *Test-granulation-arkiv, PowderPro*, <http://powderpro.se/category/test-granulation-en/>, 2017.
- [9] Rundgren, K., Lyckfeldt, O., Sjöstedt, M.: *Improving Powders with Freeze Granulation*. Ceramic Industry, 40–44., 2003.



MODELING THE MOVEMENT OF THE GRANULAR MATERIAL IN A STATIC EQUIPMENT WITH DISCRETE ELEMENT METHOD

¹POÓS Tibor PhD, ¹HORVÁTH Dániel, ²TAMÁS Kornél PhD

¹Department of Building Services and Process Engineering, Budapest University of Technology and Economics,

E-mail: poos@mail.bme.hu, daniel.horvath.nk@gmail.com

²Department of Machine- and Product Design, Budapest University of Technology and Economics,

E-mail: tamas.kornel@gt3.bme.hu

Abstract

In this study the measurements of direct shear box test were done on the assembly of hulled millet where different moisture contents were set. The moisture content dependent internal friction angle and cohesion of the aggregation were determined. With the use of a discrete element software the model of the direct shear box test was improved and simulations were done. The results of the simulation and measurement were compared. The aims of this study were to develop the DEM model of the laboratory direct shear box test and find appropriate parameter settings which will be used in further studies.

Keywords: direct shear box, internal friction angle, cohesion, discrete element method

1. INTRODUCTION

There are several technologies in the food and pharmaceutical industry which include mixing, drying and the transportation of granular materials. For design and maintenance, it is necessary to know the physical and mechanical properties of the chosen material.

Due to the rapid technological advances of our day, simulation is the preferred method to provide fast, cost-efficient approximation of the kinetic and thermodynamic processes in granular materials with sufficient accuracy. The aim of this research is to modelling the internal friction angles and cohesions of granular materials with different moisture contents. We have created the model of the direct shear box in the applied Yade discrete element software (DEM) which is suitable for the calibration of the appropriate parameter set of the examined granular material [1].

There are numerous and different types of shear boxes [2, 3] for different sizes of granular materials to determine the internal friction angle and cohesion of the particle assembly. The laboratory direct shear box test presented in our research is suitable for geotechnics, polymer technology, chemical and food industries.

2. MATERIAL AND METHOD

The evaluation method of the selected shear box test is provided by MSZE CEN ISO / TS 17892-10 [4]. The design of the instrument, the evaluation of the measurement results and the presentation of the granular material used are described below.

2.1. The examined material

During the research, the examined granular material was hulled millet which physical and mechanical properties were investigated with different moisture content. Spherical elements were



utilized in the DEM simulation, because this simple particle shape was so close to the geometry of the real hulled millet. The equivalent particle diameter of the aggregate was 1.7 ± 0.1 mm. Before the measurements began, the aggregate was wind classified so as to remove the dust, other contaminants and ensure the nearly same size. Subsequently, water was added to the granular material and stirring occasionally for ten hours to allow the water to enter inside the material and not just into the surface capillaries. We have produced six different moisture contents (wet basis, $x=11.2\%$, 16.1% , 18.7% , 23.6% , 24.1% , 28.2%).

2.2. The laboratory measuring equipment

The actual design of the device is shown on the left side of *Figure 1*, while the schematic [4] is shown on the right side of *Figure 1*, where '1' indicates the load plate, '2' the porous plate, '3' the examined sample, '4' the load cell.

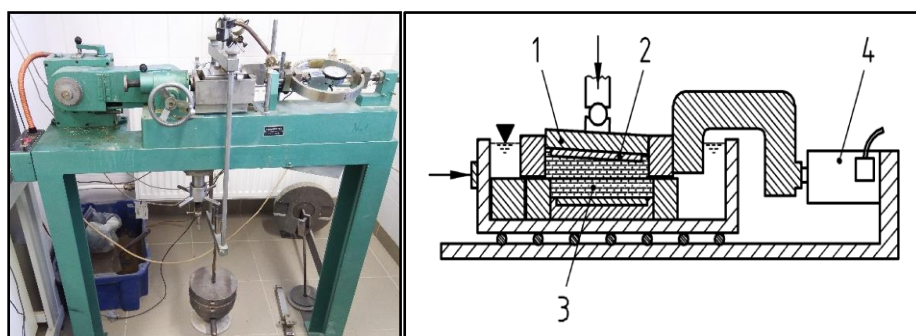


Figure 1 Real (left) and shematic (right) illustration of the laboratory shear box device [4]

The selected standard laboratory direct shear box equipment had a granular material with an inner dimension of $60 \times 60 \times 30$ mm, a shear speed of 0.02 mm/s and a shear length of 7.5 mm. The tests were performed using three different normal stress 11.96 kPa, 19.61 kPa and 29.42 kPa, respectively. While higher preloads were utilized the granules began to crumble, which was not enable to increase the pre-load sufficiency.

2.3. Materials and methods

The evaluation was performed on the basis of MSZE CEN ISO / TS 17892-10 standard [4]. The shear force was measured during the shear test using different normal loads and moisture content.

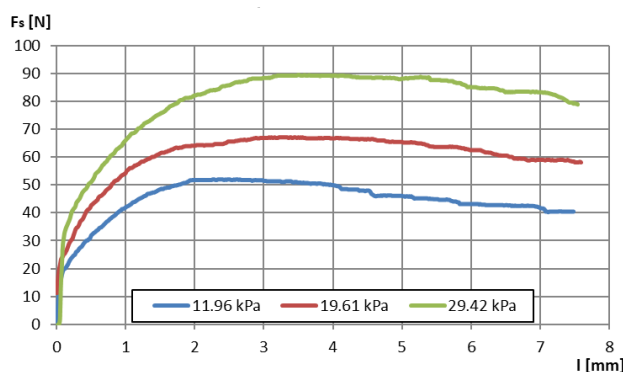


Figure 2 The shear force measured by the laboratory direct shear box test with 16.1% moisture content and three pre-loads



The maximum shear stress can be calculated from the maximum shear force (F_{max}) in *Figure 2* and the shear cross section (A):

$$\tau_{max} = \frac{F_{max}}{A} \quad (1)$$

The maximal shear stress and the stress resulting from the normal load are represented by a pair of points, and then aligned with a linear trend line, the graph of *Figure 3* was obtained:

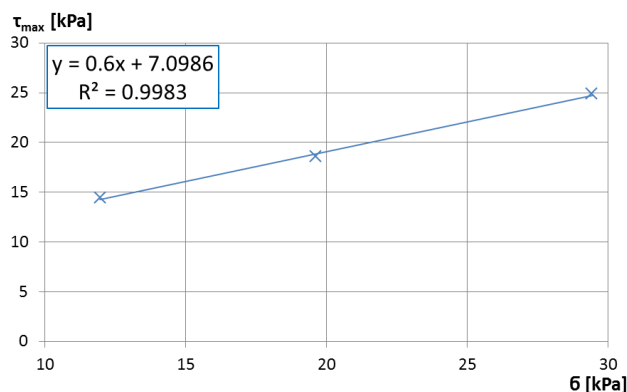


Figure 3 Maximum shear stress as a function of normal stress

The steepness of the aligned trend line equals the internal friction angle of the aggregate. The straight line and the y-axis engagement give the cohesion of the aggregate [4].

3. THE DISCRETE ELEMENT MODEL

In this study, the Yade open source DEM software was used [5]. Several contact models are available to describe the rheological processes between granules in this DEM program (e.g. friction, cohesive, capillary). The frictional cohesive contact model (CohFrictMat) were utilized, because of the nature of the investigated granular material [8]. The Yade DEM software uses an explicit integral system with respect to time, which iterates between the movement and the force-displacement law [6]. The first step in explicit time integral is to set the value of forces to default, then refresh the bounds. Afterwards the collisions are detected. The software then calculates the forces from displacements based on the engine settings. From these forces the acceleration is calculated, the velocity is refreshed, then these data (forces, displacements) are recorded. Finally, the time step is increased by Δt_i and the bodies on the program display are refreshed, then the cycle starts again.

3.1. The setup of the DEM simulation

In order to take into account the moisture content, the CohFrictMat contact model were utilized, which enable the simulation of adhesion [7, 8]. *Figure 4* shows the model of the direct shear box device under the simulation process which was improved in the Yade DEM software.

The model of the direct shear box was created from rigid wall elements called 'facetBoxes'. The bottom box (Number 2 in *Figure 4*) was opened on the upper side and upper box (Number 1 in *Figure 4*) was opened below and above. We created two more box sides to prevent the particles falling out during shearing.

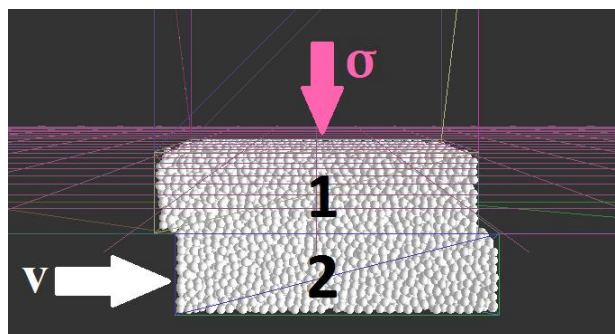


Figure 4 The simulation process of the direct shear box test in Yade DEM software (1: upper box, 2: lower box, σ : normal stress, v : shear direction)

The first stage of the simulation was the gravitational deposition, i.e. the filling of the shear box with particles, until the unbalanced force [6] decreased to less than 0.001. Then we turned on the cohesions between the particles with the 'setCohesionOnNewContacts' command and then using a wall element to provide the normal constant preload (marked with σ on Figure 4) on the assembly. As a result of the movement of the particles during shearing, the particle assembly modifies the displacement of the plate utilized for preloading even in the opposite direction. Therefore, this plate used for preload was controlled by the upward and downward movement as a function of the force exerted on it, so that the applied plate as wall element provided a constant pre-load with this servo mechanism. The shearing speed was set to 1 mm/s and the shear length was 8 mm. During the simulation, the shear force acting on the shear direction side of the upper box, the change of the unbalanced force, the position of the wall element providing the normal load and the force applied to it were recorded. Table 1 summarizes the material parameters of the direct shear box model.

Table 1 Material properties of the measuring equipment in the DEM software [9]

Measured particle density [kg/m ³]	Selected Young modulus [GPa]	Selected poisson ratio [1]	Measured friction angle [°]
7750	200	0.3	40.1

In the simulations, the global damping which create an opposite force against the accelerating particles was 0.5 and the time step was $4.94 \cdot 10^{-6}$ s. The mechanical parameters of the modelled hulled millet are summarized in Table 2, which was taken from porosity measurement, the laboratory direct shear box test, and preliminary simulation experiments [9, 10].

Table 2 Material properties of the hulled millet in the DEM method

Moisture content [%]	Measured particle diameter [mm]	Measured particle density [kg/m ³]	Selected Young modulus [MPa]	Selected poisson ratio [1]	Measured friction angle [°]	Calibrated cohesion strengths [kPa]	Calibrated eta [1]
23.6	1.7±0.1	1402	20 [9]	0.2	40.1	60	0.05

The shape of the hulled millet grains was modelled by spherical particles. During the simulations 21,000 particles were used to fill the DEM model of the shear box. The shear and normal strengths (τ , σ), which simulates the strength of the bonds between the particles (occurred by liquid bridges) were the same. Also the value of eta as the dimensional rolling and twisting strengths were the



same, which could be used for increasing the internal friction of the particle set with the interference of particles.

4. RESULTS

The method of evaluation is described in *Section 2.3*. The obtained internal friction angles and cohesions from laboratory direct shear test are shown in *Figure 5*.

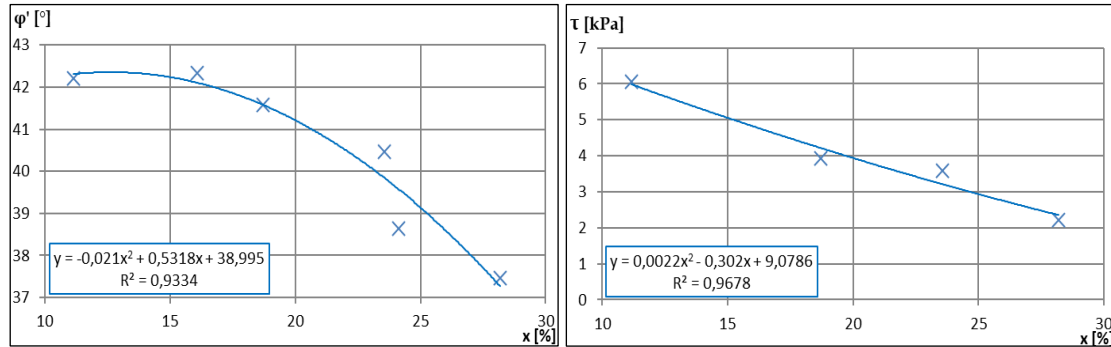


Figure 5 The measurement results of the laboratory direct shear box test. On the left side, the internal friction angle, on the right side the cohesion dependence on the moisture content (wet basis) for hulled millet

Both the internal friction angle and the cohesion were polynomially decreased depending on the moisture content.

During the DEM modeling, the simulations were performed with the moisture content of 23.6%. We compared the simulated and the measured results in *Figure 6*, where the maximum shear stress was represented as a function of normal stress.

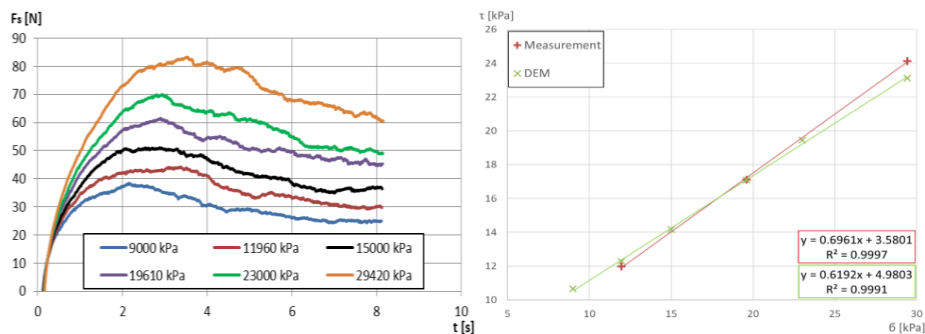


Figure 6 On the left side, the simulated shear force for hulled millet of 23.6% moisture content by the time, and on the right side the maximum shear strength dependence on the normal stress by measurement and DEM simulation

During the DEM modeling, we performed simulations with six pre-loads in order to obtain more accurate results. It has been known that the model yielded the expected results with the parameter setting for a moisture content (*Table 2*), as the points were on a straight line (*Figure 6* on the right). The highest difference between the measured and the discrete elemental method was 4.1%. Based on the setting parameters (*Table 2*) and the results from the measurements (*Figure 6* on the left), it can be determined that the difference between the set (macromechanical) and the particles (micromechanical) cohesions are one order of magnitude higher. Furthermore, during the simulations, it was found that the steepness of the trend line increased by increasing the eta



parameter in the shear strength normal stress diagram (*Figure 6 right*), while the axial section decreased considerably by decreasing the cohesions.

CONCLUSIONS

The standard laboratory direct shear box tests with hulled millet was used with different moisture contents in this study. The effect the proportion of moisture content on cohesion and internal friction angle of the aggregate were investigated. We found that both parameters were polynomially decreased with the increase of the moisture content. Using DEM we created the model of the direct shear box and then simulated with a given moisture content. The geometry of the particles were approximated by sphere, and a cohesive contact model (CohFrictMat) was used to take into account the effect of moisture. The results of measurements and simulations were compared with a maximum deviation of 4.1%. Based on this experience, the steepness of the failure envelope (maximum shear strength - normal stress diagram) can be adjusted by increasing the eta parameter and the "shifting" by increasing the micro cohesion values (σ, τ). The results showed that the improved DEM model will be appropriate for further researches to determine additional moisture dependent micro-mechanical parameters.

ACKNOWLEDGEMENTS

This paper was supported by Hungarian Scientific Research Fund (NKFIH/PD-116326).

REFERENCES

- [1] Cundall, P.A., Strack, O.D.L.: *Discrete numerical model for granular assemblies*. Geotechnique, 29 (1), 47–65., 1979.
- [2] Suhr, B., Six, K.: *On the effect of stress dependent interparticle friction in direct shear tests*. Powder Technology, 294 (Supplement C), 211–220., 2016.
- [3] Yang, H., Xu, W.-J., Sun, Q.-C., Feng, Y.: *Study on the meso-structure development in direct shear tests of a granular material*. Powder Technology, 314 (Supplement C), 129–139., 2017.
- [4] MSZE CEN ISO/TS 17892-10: *Geotechnikai vizsgálatok. Talajok laboratóriumi vizsgálata. 10. rész: Közvetlen nyíróvizsgálat (ISO/TS 17892-10:2004)*, 2004.
- [5] Šmilauer, V., Catalano, E., Chareyre, B., Dorofeenko, S., Duriez, J., Gladky, A., Kozicki, J., Modenese, C., Scholtès, L., Sibille, L., Stránský, J., Thoeni, K.: *Dem formulation*. In *Yade Documentation 2nd ed*. The Yade Project, 37., 2015.
- [6] Šmilauer, V., Gladky, A., Kozicki, J., Modenese, C., Stránský, J.: *Using and programming*. In *Yade Documentation 2nd ed*. The Yade Project, 149., 2015.
- [7] He, Y., Peng, W., Tang, T., Yan, S., Zhao, Y.: *DEM numerical simulation of wet cohesive particles in a spout fluid bed*. Advanced Powder Technology, 27 (1), 93–104., 2016.
- [8] Bourrier, F., Kneib, F., Chareyre, B., Fourcaud, T.: *Discrete modeling of granular soils reinforcement by plant roots*. Ecological Engineering, 61 (Part C), 646–657., 2013.
- [9] Horabik, J., Molenda, M.: *Parameters and contact models for DEM simulations of agricultural granular materials: A review*. Biosystems Engineering, 147 (Supplement C), 206–225., 2016.
- [10] Poós, T., Horváth, D., Tamás, K.: *The compare of angles of repose with discrete element method and measurement*. 8th International Scientific Conference, 65–70., 2017.09.07-09.