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1. Introduction

Among the wide range of granular materials, products manufactured in powder form are present in many industries such as chemical, pharmaceutical, cosmetic, food and polymers industries [1]. A powder is a set of heterogeneous particles dispersed in a continuous gas phase.

Powders processing involves different unit operations such as aeration, drying, milling, compaction, handling, storage and conveying [2]. In order to insure the feasibility of the process chain, good powders flowability is required.

Nevertheless, powders flow behavior is complex in nature because is not an inherent property of the material. In fact, it depends on: the intrinsic and extrinsic properties of the grains such as size, shape, surface rugosity, porosity and crystal chemistry [3]; the bulk powder properties such as size distribution, bulk density, interaction forces between particles; the equipment design; the processing conditions such as stress levels; the processing environment such as temperature and humidity.

The complexity of the link between local particles interactions and their global mechanical behavior has undermined flowability understanding, remaining essentially empirical.

Among the greatest challenges in powder technology, the main trend in industrial applications is to develop an effective device allowing to predict powder flowability by reproducing the mechanical conditions encountered during processing.

In order to allow a better understanding of the powder/process relation, flow properties of powders can be categorized in groups corresponding to the stress levels applied to the powder: over packed bed, free surface and aerated conditions [4]. Different methods can be used in order to reproduce these stress levels:

- Packed bed conditions: consolidation tester (Jenike's cell, FT4, RST-XS…), uniaxial compression (Instron 4505, Stylcam® 100R rotary press, Gerristen test, Johanson test);
- Free surface conditions: angle repose (Granuheap®, BEP2® …), vibratory compaction (Densitap®,Hosokawa®, Granupack®), flow rate through apertures

(Gardco®, Granuflow®, BEP2®, Flodex®…), rotating drum (Aero-Flow®, Granudrum®);

- Aerated conditions: rotating drum-fluidization test (Granudrum®, The Revolution®…), fluidization bed (FT4- aeration test, mini-Glatt).

Different parameters or index can be obtained from these testers such as angle of repose (AoR), Hausner ratio (HR), Carr's index (i.e., compressibility) (CI), basic flow energy (BFE), flowability index (ff_c), flow rate index (FRI), fluidization quality (FQ) and aeration index (AI). Each one of these allows to "estimate/quantify" flow behavior under different mechanical situations.

Hausner ratio and Carr's index are two closely related empirically derived methods that allow to assess the flow behavior from bulk densities. Quite popular in industry and academia, because of the simplicity and the rapidity of the measurement, both compression ratios where obtained under quite different reflections. Carr suggested in 1965: "*it is obvious that compressibility is a very important flow characteristic… the more compressible a material is, the less flowabable it will be"* [5]. Introduced by Hausner in 1967 while studying copper powders, HR was described as "*an indicator of the friction condition between powder particles*" [6].

In this study, *powder flowability under free surface conditions is determined by using HR*. The flow properties of powders, according to HR, were assessed using three vibratory compaction devices: the well-known DensiTap®, GranuPack® which is an automated version of DensiTap® from Granutools® and a homemade Vibratory device initially developed for granular rheology, allowing to control the dynamic parameters of the motion [7].

The main difference between these devices is the way of shaking, in DensiTap® and GranuPack® the vibration is generated by free-falls performances, commonly denoted as taps. The tapping amplitude, or intensity, corresponds to the free-fall height. In the case of our Vibratory device, the vibration is harmonically driven by an electromagnetic shaker excited with a sinusoidal signal.

The aim of this work is to get a deeper understanding of HR by confronting these compaction tests, pointing out their relevance considering the physical sense of the measurement. To do so, it is suitable to approach flowability from a physical vibrated granular materials point of view. Granular compaction takes place when a pile of grains is submitted to a series of taps and the packing fraction of the pile, defined as the ratio of the volume occupied by all the

grains divided by the volume of the assembly, slowly increases. The compaction and flowability of granular materials are still a serious challenge for physicist, and so, most efforts have been made to relate thermodynamic approaches for understanding granular media. Most of these studies are carried out with ideal or model granular materials such as glass beads. Among these works, the ones on compaction dynamics agreed that the granular media subjected to shocks, or taps, slowly compact to a stationary state [8, 9, 10]. Ribière et al. 2007 showed that this stationary state is independent of the initial state of the powder, slightly or not dependent of the vibration frequency but is strongly dependent of the vibration amplitude [11]. Ludewig et al. (2008), proposed an energetic dimensionless parameter for investigating the physical properties of dense granular systems which seems to capture the entire dynamics of compaction [12]. The energy injected by a tap or sinusoidal signal seems to be a key parameter for describing the compaction dynamics. From this observation, also validated in this work, we propose, that, the energy injected to the granular media is also a key parameter for describing powders flowability through HR assessment.

Here we experimentally study granular materials submitted to vibration under free surface conditions. The mainly focus is on the properties of the stationary state for determining HR as a flowability index and discuss the validity of such configurations in function of the vibration energy supplied to the granular system. For this, when possible, the vibration will be described as a function of the energy injected to the system. The aim is to develop an energetical point of view for describing flowability by the use of HR.

In order to illustrate the consistency of the results, a variety of non-ideal, or more complicated, granular materials notably different from glass beads in terms of nature, shape, polydispersity, roughness, even cohesion behavior, have been selected to cover a large range of applications from pharmaceutical to food industries.

2. Materials and methods

2.1. Materials

For the current study, nine powders samples have been selected according to their size distribution, particles shape and their flow behavior observed with naked eye. Among them, two grades of lactose (L): Tablettose 70 and Retalac; two grades of microcrystalline cellulose (MCC): Avicel ph 102 and Avicel ph 105, and five commercial food powders (FP) denoted

Food A, Food B, Food C, Food D and Food E respectively. The commercial name of the food powders is kept confidential.

- Tablettose 70 is the trade name for α-lactose monohydrate produced by MEGGLE (Germany), which particles are characterized by a narrow size distribution. According to the technical brochure, Tablettose 70 is especially designed for direct-compression and exhibits a good flowability.
- Retalac is lactose powder produced by MEGGLE (Germany), composed by equal parts of hypromellose polymer and α −lactose monohydrate. Retalac powders are characterized by excellent flow and compaction properties.
- The two grades of Avicel, ph 102 and ph 105, produced by FMC Biopolymer, are high purity microcrystalline cellulose particles. Both powders differ only by their particles size distribution. Avicel ph grades are well known in the pharmaceutical industry for direct compression tableting applications.
- Food A and C are vegetal powders, Food B is a mineral powder, Food D is a dietary fiber, and Food E is a protein. Food A, Food B and Food C are characterized by having a poor flowability observed with naked eye. All food powders particles differ from each other in size and shape.

It should be noted that, since water vapor can introduce cohesion between the particles and change their frictional properties, at the beginning of the experiments, all powders samples were conditioned at a relative humidity (RH) of 30%. During analysis, samples were keep at laboratory environmental conditions, between 30 and 40 % RH.

2.2. Powders physical characterization techniques

2.2.1. Particle size analysis

Particle size distributions were measured by laser diffraction in liquid media with a Mastersizer 2000 from Malvern Instruments. Depending on the nature of the particles, water or ethanol were used in order to improve their dispersion avoiding dissolution. All samples were analyzed in triplicate to ensure reproducibility.

2.2.2. Particle morphology analysis

Particles morphology analyses were assessed with a Scanning Electron Microscopy (Joel JSM T330A), and a field emission gun operating between 5kV to 15 kV. Samples were placed onto

carbon tapes and coated with gold during 5 min using an Ion SPUTTER JFC-1100 Jeol under argon gas purge.

2.2.3. True particles density analysis

Powder true density analysis was performed in an Helium pycnometer Accupyc 1330 from Micromeritics. All data measurements are the average of ten measurements obtained from one powder sample.

2.3. Hausner ratio determination

Hausner ratio can be defined as the ratio of the tapped bulk density to the loose bulk density as follows:

$$HR_{\infty} = \frac{\rho_{\infty}}{\rho_0} \quad (1)$$

where ρ_0 is the aerated density obtained after freely pouring the powder in to the vessel and ρ_{∞} is the asymptotic constant density obtained during tapping until no further volumes changes occurs, also described as the stationary state [6, 13, 14].

A more practical equation widely used to evaluate flow properties is given by volume changes in a graduated cylinder after certain number of taps, N:

$$HR = \frac{\rho_N}{\rho_0} = \frac{V_0}{V_N} = \frac{C_N}{C_0} \quad (2)$$

where ρ_0 and ρ_N are the aerated and tapped density, V_0 and V_N are the initial and the tapped volume, C_0 and C_N are the initial and tapped compactness ($C_0 = 1 - \varepsilon$, with ε the porosity of the aerated bed).

HR according to Carr's classification, as indicator of the flowability characteristics of the powder, has been reported elsewhere [5, 15].

It should be noted that in the following the accuracy of HR is calculated using instrumental and random errors analysis as follows:

$$E_T = \sqrt{{E_R}^2 + {E_I}^2} \quad (3)$$

$$E_I = \frac{V_0}{V_N}\sqrt{\left(\frac{\Delta V_0}{V_N}\right)^2 + \left(\frac{\Delta V_N}{V_N}\right)^2} \quad (4)$$

$$E_R = \sqrt{\frac{1}{n-1}\sum_{k=1}^{n}(HR_k - \overline{HR})^2} \quad \text{with} \quad \overline{HR} = \frac{1}{n}\sum_{k=1}^{n} HR_k \quad (5)$$

where E_T, E_R and E_I are respectively the total, random and instrumental errors, n is the number of repetitions and $\overline{HR}$ the mean value of Hausner ratio.

2.4. Flowability testers: vibratory compaction under free surface conditions

2.4.1. DensiTap®

This device allows determining aerated and tapped densities by mechanically tapping a graduated glass cylinder. A rotating cam provides the tapping action by raising the cylinder platform through a fixed distance of A = 3 mm at a frequency of 4.16 Hz.

As described in the European Pharmacopeia, about 150 mL of powder are freely poured by using a funnel into the vessel allowing to determine aerated density while tapped density is determined systematically after 500 taps [15]. This number of taps, the most used in scientific literature, is proposed as the number of taps suitable to reach the steady state.

2.4.2. Granupack®

Developed by Granutools®, this device automatically measures the position of a hollow cylinder placed on the top of the powder column. From this distance, the height and the volume of the powder bed are computed allowing to determine the evolution of the packing fraction as function of the number of taps.

The metal cylindrical enclosure containing the powder sample performs free falls with an amplitude of 1 or 3 mm while the tap frequency can be tuned from 0,1 to 2 Hz.

The Granupack® software allows to plot the compaction kinetic curve by monitoring the intermediate states. Other dynamical parameters can be extrapolated from the compaction curves, using for example Bideau's model [9]:

$$C(N) = C_\infty - (C_\infty - C_0)exp\left[-\left(\frac{N}{\tau}\right)^\beta\right] \quad (6)$$

where C_∞ is the extrapolated compactness when N tends to infinity, τ is the characteristic tap number related to the compaction dynamics and β is a stretching exponent.

The kinetic parameters τ and β have been used in others studies to differentiate powders having similar HR, with β related to powder cohesiveness [9, 16]. In this work, we are only interested on the determination of C_∞. The aim is to compare HR from HR_∞ values.

As suggested by Granutools®, to determine HR, a powder sample of 35 mL is poured in the metallic cylinder. Free fall events are carried out at a fixed frequency of 1 Hz and the two tapping amplitudes, 1 and 3 mm, allowed by the device have been tested. The compaction curves are obtained from 0 to 500 taps.

2.4.3. Vibratory system

This homemade device is a particle damper where a cylindrical vessel in borosilicate glass is harmonically driven by an electromagnetic shaker (Fig.1). In this case, each tap is defined by a sinusoidal motion of the container. The motion of the system is given as:

$$x(t) = A_0 \cdot sin(2\pi f \cdot t) \quad (7)$$

Where A_0 and f correspond to the amplitude and frequency of the signal and characterize the motion.

The dynamic of the system is experimentally obtained by an accelerometer glued to the vessel. Thus, the amplitude of the motion, A_0, can be determined by using the root mean square acceleration, a_{RMS}, as follows:

$$A_0 = \frac{a_{RMS} \cdot \sqrt{2}}{(2\pi f)^2} \quad (8)$$

Finally, the mechanical vibration energy, E_v, injected to the powder bed can be calculated:

$$E_v = \frac{1}{2} m (2\pi f)^2 {A_0}^2 \quad (9)$$

where m is the powder mass.

This device allows to measure what we define as HR_∞. Indeed the final compaction state is determined after no further volume changes occur, also known as the stationary state (ρ_∞).

As the stationary state is slightly or not dependent on the vibration frequency but is strongly dependent on the vibration amplitude [11] a frequency of 30 Hz was chosen because it allowed the wider amplitude scanning, from 10 to 110 $m.s^{-2}$.

It should be noted that most studies in granular compaction when working with harmonically driven vibrations use a frequency of 30 Hz [8, 10, 12] or even 60 or 90 Hz [11] but discretize the movement by adding regular intervals of 1 s. The basic idea of using discrete "shakes" is to allow sufficient time between excitations so that all motion in the column from one shake ceased before the next starts. In our case, when working with continuous signals, we assume, as the typical time scale of rearrangements of rigid particles, also called the *confinement time*, is highly inferior ($T_{p,max} \sim$ 0,2 ms) to one period (33 ms), that each successive vertical shake are separated by a time delay that allows the total relaxation of the granular assembly [17]. This can also be validated by the fact that each HR value is obtained at one steady-state density, and so the granular system reaches a stationary state during continuous vibration.

In terms of tapped density, at a vibration frequency of 30 Hz, each HR_∞ value was obtained after at least 5000 periods (comparable to 5000 taps).

The experimental procedure is the following: 30 g of powder are systematically poured into the container to determine ρ_0. Once the frequency of the excitation signal is fixed at 30 Hz the amplitude of the signal is progressively increased, resulting in an increase of the acceleration (measured a_{RMS}). It should be noted that each amplitude scanning experiment takes around 45 min per sample.

In order to allow a better comparison between the devices, their characteristics and operating conditions are summarized in Table 1.

Table 1
Devices characteristics

Characteristics / Device	Densitap®	Granupack®	Vibratory system
Type of vibration	shock wave	shock wave	sinusoidal
Vessel dimensions [mm] (diameter / height)	34.7 / 335	26 / 155	26 / 300
Quantity of powder [mL]	150	35 *	~ 80 (30 g)
Vessel material	borosilicate glass	stainless steel	borosilicate glass
Amplitude of vibration [mm]	1 *	1 ou 3 *	0 to 3
Frequency of vibration [Hz]	4.16 *	1	30 **
HR determination	500 taps*	500 taps*	Stationary state

* Fixed or proposed by the supplier.
** Chosen to obtain a maximal amplitude scanning.

3. Results and discussion

3.1. Physical characterization results

Powders physical properties are reported in Table 2. Particles shape are displayed in Fig .2 (Food Powders), Fig. 3 (MCC powders) and Fig. 4 (Lactose powders). All SEM micrographs are presented with a bar scale of 20 µm, 50 µm and 100 µm.

Table 2
Physical properties of the samples

Powder	Particle size distribution ± 1 [µm]				True density ± 0.01 [g/mL]	Shape [-]
	d_{10}	d_{50}	d_{90}	Span[-]		
Food A	7	15	38	2.2	1.46	Isometric rough in agglomerate state
Avicel ph 105	4	18	40	2.0	1.56	Rod Isometric rough
Food B	4	29	115	3.8	2.28	Isometric smooth
Food C	29	104	155	1.9	1.47	Irregular

Avicel ph 102	29	104	248	2.0	1.57	Rod Isometric rough
Food D	65	120	221	1.3	1.45	Agglomerates pop corn like
Retalac	89	203	408	1.6	1.42	Agglomeration of amorphous-like particles
Food E	123	267	472	1.3	1.23	Isometric rough
Tablettose 70	118	204	380	1.3	1.54	Individual agglomerates with a rough structured surface

281
282
283 SEM micrographs of Food samples are shown in Fig. 2. Food A and Food D are formed by
284 agglomerates of amorphous-like particles and pop-corn like particles respectively. Food B is
285 composed by oval shape particles were smaller particles get stuck on the surface of the largest
286 ones. Food C is composed by bigger irregular particles with rough surfaces. Food E is
287 composed by the most irregular particles, closer zooms exhibit a porous surface.
288
289 Pharmaceutical excipients, Avicel ph 102 and Avicel ph 105, are composed by rough rod
290 particles (Fig. 3). Avicel ph 105 particles are smaller than Avicel ph 102.
291
292 Lactose samples are shown in Fig. 4. Tablettose 70 powder exhibit better-defined particles
293 with a rough structured surface. Retalac particles are the agglomeration of crystalline alpha
294 lactose and hypromellose fibers resulting in amorphous-like particles.
295
296 Particles size distribution are presented in Fig. 5 (Food powders), Fig. 6 (MCC powders) and
297 Fig. 7 (Lactose powders). As depicted from Table 2 and Fig. 5, Food A particles are the
298 thinnest ones, Food B present the widest span, Food C present a bimodal distribution, Food D
299 show a narrow size distribution while Food E is composed of the biggest particles.
300
301 Particle size distribution of MCC powders show that particles of Avicel ph105 are smaller
302 than of Avicel ph 102 particles (Figure 6) while the two lactose samples have a similar
303 particle size distribution (Figure 7).
304
305
306 *3.2.Flowability results*
307
308 *3.2.1. Assessment of powder flowability with DensiTap® device*
309

In order to study the sensitivity of Hausner ratio to the initial state of the powder samples, two batches were selected for their quite different flow behavior: Tablettose 70 (good flowability) and Food A (bad flowability). For each powder, the initial and final volumes were measured by testing different filling methods: gently, rapid, with or without funnel, changing the operator, etc. All data, presented in Table 3, are the average of 15 measurements on 15 different samples from the same batch.

Table 3

Accuracy of the measurements for 15 and 3 repetitions for Tablettose 70 and Food A.

Powder	HR (15 repetitions)	HR (3 repetitions)
Tablettose 70	1.15 ± 0.01	1.14 ± 0.01
Food A	1.26 ± 0.02	1.25 ± 0.01

The good repeatability of the results, according to the reading of initial and final volume after tapping, showed an easily reproducible initial state (Table 3). It should be noted that we do not know if this aerated state obtained after pouring the powder is the loosest packing that could be achieved, but proved to be a rather constant value so that repeated measurements, starting from the same initial conditions, could be made. These results are in disagreement to those obtained by several authors, suggesting that HR values are strongly affected by the highly irreproducible initial pouring of the powder [4, 16].

Moreover, as depicted from Table 3, the use of 3 measurements seems to be enough to obtain an acceptable accuracy to determine HR. Thus, in the following, all HR values calculations are obtained as the average of 3 measurements.

The results obtained for all powders are gathered in Table 4 according to increasing HR.

Table 4

Powders flowability analysis derived from Hausner ratio using a DensiTap® device with f_t = 4.16 Hz, A = 3 mm and N = 500 taps.

Powder	d_{32} (µm)	d_{43} (µm)	HR (A = 3 mm)	Flow behavior
Tablettose 70	184	244	1.14 ± 0.01	Good flow
Food D	102	133	1.15 ± 0.02	Good flow
Food E	178	283	1.21 ± 0.01	Fair flow
Food C	24	81	1.22 ± 0.02	Fair flow

Food A	8	26	1.25 ± 0.01	Fair flow
Retalac	159	229	1.26 ± 0.02	Passable flow
Food B	10	49	1.28 ± 0.03	Passable flow
Avicel ph 102	40	126	1.31 ± 0.04	Passable flow
Avicel ph 105	8	20	1.44 ± 0.04	Poor flow

Several authors have shown that powders ability to flow increases as particles size increases [18, 19]. Generally this is related to an increase of the gravitational forces which become preponderant compared to interparticle forces due to an increase of particles individual mass. In our case, HR analysis with $f_t = 4.16$ Hz and $A = 3$ mm, non-clear conclusions can be drawn by size distribution analysis. For example, lactose powders, Tablettose 70 and Retalac, composed by the biggest particles, exhibit quite a different flow behavior mainly attributed to the highly heterogenous shape of Retalac powders. Nevertheless, this statement based in particles morphology and size doesn't explain flowability differences in the case of food powders.

3.2.2. Flowability assessment with Granupack® device

The evolution of powders compactness as a function of the number of taps for an amplitude of 1 and 3 mm are presented, respectively, in linear and logarithmic scales in Fig. 8, Fig. 9, Fig.10 and Fig.11 (Error bars are shown for each experimental point).

As can be seen from Fig. 8 and Fig.10, the compactness increases with the number of taps. The monitoring of compaction as function of number of tap allows to visualize more easily if the steady state is reached. But, the representation in logarithmic scale (Fig. 9 and Fig. 11) shows that the steady state is not always established as suggested by the representation in linear scale (case of Avicel ph 102, Retalac and Food D).

In Tables 5 and 6 are gathered the HR values obtained with both tapping amplitudes, 1 and 3 mm respectively. According to the index classification from Table 5, it can be depicted that most powders exhibit a fair ability to flow. In difference, from Table 6, most of the samples, exhibit another classification. Thus, HR assessment seems to be strongly dependent on the free-fall amplitude.

Table 5

Flow behavior under Granupack® deduced by Hausner ratio (f_t = 1 Hz, A = 3 mm, N = 500)

Powder	d_{32} (µm)	d_{43} (µm)	HR (A = 3 mm)	Flow behavior
Food E	178	283	1.15 ± 0.02	Good flow
Food C	24	81	1.22 ± 0.01	Fair flow
Retalac	159	229	1.24 ± 0.01	Fair flow
Avicel ph 102	40	126	1.24 ± 0.01	Fair flow
Tablettose 70	184	244	1.24 ± 0.03	Fair flow
Food D	102	133	1.25 ± 0.01	Fair flow
Food B	10	49	1.28 ± 0.02	Passable flow
Food A	8	26	1.31 ± 0.01	Passable flow
Avicel ph 105	8	20	1.42 ± 0.01	Poor flow

Table 6

Flow behavior under Granupack® deduced by Hausner ratio (f_t = 1 Hz, A = 1 mm, N = 500).

Powder	d_{32} (µm)	d_{43} (µm)	HR (A = 1 mm)	Flow behavior
Food E	178	283	1.13 ± 0.01	Good flow
Food C	24	81	1.16 ± 0.01	Good flow
Retalac	159	229	1.16 ± 0.01	Good flow
Food A	8	26	1.17 ± 0.03	Good flow
Food D	102	133	1.19 ± 0.02	Fair flow
Food B	10	49	1.20 ± 0.03	Fair flow
Avicel ph 102	40	126	1.21 ± 0.01	Fair flow
Tablettose 70	184	244	1.25 ± 0.01	Fair flow
Avicel ph 105	8	20	1.29 ± 0.02	Passable flow

To obtain C_∞ values and so HR_∞, all compaction curves were fitted using Eq. (6). For all samples, Bideau's model was found to be in good agreement with the kinetics of compaction ($R^2 \approx 0.99$) as shown in Fig. 12.

Fig. 12 and Fig. 13 show the compaction curve and the compaction model, for Avicel ph 102, in linear and logarithmic scales respectively. The logarithmic representation of the data, Fig.13, shows a slight deviation from the model for $N < 10$. The compaction model chosen for this study has been established using glass beads [9] thus, the discrepancy observed can be explained by the use, in our case, of more complex samples.

The comparison between HR and HR_∞values is presented in Fig. 14. For most powders, statistically similar HR values were obtained from $C_{N=500}$ and C_∞ for a fixed amplitude, showing that the use of 500 taps to evaluate HR, is a quite good choice as a standard procedure and can explain why this number of taps is the most used in scientific literature [13]. Only for Retalac samples HR and HR_∞, obtained with A = 1 mm, are statistically different. This exception can be explained by being too far from the steady state with N = 500 taps, as can be depicted from Fig. 11.

Fig. 14 allows to better apprehend at which point HR is strongly dependent on the tapping amplitude. Indeed, most of the samples present a different flowability classification when changing the free-fall amplitude. Only Food E and Tablettose 70 powders possess statistically similar HR values.

3.2.3. Flowability assessment with a vibratory device

The evolution of powders compactness as function of the vibration amplitude at a fixed frequency of 30 Hz are presented in Fig. 15. The reorganization of all powders studied seems to be similar with a variation of compactness between 0.06 and 0.11. MCC samples present a similar ability to compact while Lactose samples, having similar size distributions, present a quite different compaction behavior. Retalac sample, a co-processed dry binder, exhibit a lower packing density than Tablettose 70, generally attributed to the presence of Hypromellose and particles irregular shape. Food C and Food D have the higher packing density. But, no general conclusions can be drawn on the compaction behavior of food samples mostly attributed to the different nature of the particles.

From a global perspective as Ribiere et al., whose work focuses on a granular system composed of glass beads of 1 mm diameter [11], our experiments have established that all 9 powders studied, when subjected to a tapping dynamics, compact and reach a stationary state

that depends on the tapping intensity (Figure 15). Moreover, all samples seem to reach a maximal compaction state during vibration. Powders such as Food D and Tablettose 70, after a maximal state of compaction, when increasing the vibration amplitude, the powder bed undergoes decompaction in a quite slow manner. This "slow decompaction process" allows to see the convection dynamic taking place at vicinity of the lateral walls and so, to follow progressively the decompaction process. For samples such as Food B and Food E, decompaction takes place in a quite abrupt way, generating a powder cloud making impossible to measure volume changes. Decompaction dynamics seems to be time dependent, even more powder-cohesiveness dependent. More studies should be carry-out to fully fill this statement.

Powders compaction behavior under free surface conditions can be schematized as presented in Fig.16, were three regions can be observed:

- Compaction (region I): the powder bed volume decreases sharply as the vibration amplitude increases. During this first step of compaction, a rearrangement of the powder particles occurs when the energy supplied to the bed powder overcomes interparticle forces.
- Maximal compaction (region II): no evolution of the powder bed volume is observed, and so a constant compactness state is observed. This region, could be interpreted as the maximal/densest compaction state of the powder bed, also known as the random close packing limit, where the particles are touching and packed in as tightly as possible. In this region, the powder has a solid-like behavior.
- Decompaction (region III): corresponds to the transition between solid-like and fluid-like behaviors. It begins with a dynamics of convection generating the auto-aspiration of air into the powder bed, and so inducing decompaction.

The HR was plotted as a function of the vibration amplitude for MCC, Lactose and Food powders as shown respectively in Fig.17, Fig. 18 and Fig. 19. For all figures, HR is only determined until the powder bed undergoes decompaction.

According to HR classification, MCC powders can exhibit fair, passable up to poor flow behavior depending on the vibration energy imposed to the sample (Fig. 17). The flowability of Avicel ph 105 is poorest than Avicel ph 102 ascribed to differences in moisture content or particle size distribution [20]. Our experiences validated this statement but showed an important dependency with the amplitude of vibration. Indeed, Avicel ph 105 has a better

flow behavior at low vibration amplitudes than Avicel ph 102 powders (A_0 < 1.6 mm). When increasing the amplitude of vibration, A_0 > 1.6 mm, Avicel ph 102 powders undergo decompaction while Avicel ph 105 samples continues to compact. According to Geldart's classification, cohesive materials have HR values greater than 1.4 [18]. Thus, Avicel ph 105 cohesiveness could explain why the powder does not undergo decompaction even at high vibration amplitude/energy, as depicted in Fig. 15.

For lactose powders HR classification shows that Tablettose 70 samples exhibit a better ability to flow than Retalac (Fig. 18). The vibration amplitude range to obtain a steady HR value differs for Tablettose 70 ($0.8 < A_0$ (mm) < 1.4) and Retalac ($1.5 < A_0$ (mm) < 3). Moreover, as depicted from Fig. 18, Tablettose 70 undergoes decompaction at smaller amplitudes than Retalac, suggesting that Retalac particles possess stronger interparticle forces interaction.

Food Powders flow behavior from HR classification shows that before decompaction Food E and Food C samples exhibit a fair ability to flow while Food A, Food B and Food D samples exhibit a fair ability to flow (Fig.19).

The vibration amplitude, at a fixed frequency, is responsible for the amount of energy supplied to the powder bed (calculated from Eq. 9, shown in Fig. 20). *This vibration energy will allow the particles to jump and to reorganize, or not.*

When the powder bed is at its maximum compaction state, a maximal HR value is obtained. This maximal HR value, obtained under free surface conditions, is named in this work "**Ultimate Hausner ratio** (HR_U)". Corresponding to the maximum packing fraction of the powder, HR_U, is indeed a topological intrinsic characteristic of a powder, which can be used to understand, or even more to quantify, in a quite simple way, the role of powder interparticle forces in flowability.

In industrial applications, vibrations have been used together with fluidization in order to overcome cohesion problems [21, 22]. The quantification of the vibration energy needed to compact the powder bed, or to undergo decompaction, under free surface conditions, is of great interest in powders handling, filling and transport operations. Thus, the assessment of HR_U could result in significant time and resource saving in industrial technology.

3.2.4. Flowability results comparison

A compilation of HR values obtained with the 3 devices used in this work, Densitap®, Granupack® and our Vibration device, is gathered in Table 7. For simplicity, we chose to compare between HR values obtained by free-falls performances at 3 mm, for Densitap® and Granupack® devices, with the values obtained with our Vibratory device, using as reference the Ultimate Hausner ratio (HR_U).

Table 7

Hausner ratio according to Densitap®, GranuPack® (A = 3 mm) and Vibratory device

Powders	HR (-) $_{DensiTap®}$	HR_∞ (-)$_{GranuPack®}$	HR_U (-) $_{Vibratory\ device}$
Tablettose70	1.14 ± 0.01	1.25 ± 0.03	1.21 ± 0.02
Food D	1.15 ± 0.02	1.27 ± 0.01	1.27 ± 0.01
Food E	1.21 ± 0.01	1.14 ± 0.02	1.19 ± 0.01
Food C	1.22 ± 0.02	1.20 ± 0.01	1.18 ± 0.01
Food A	1.25 ± 0.01	1.31 ± 0.02	1.27 ± 0.01
Retalac	1.26 ± 0.02	1.24 ± 0.01	1.31 ± 0.02
Food B	1.28 ± 0.03	1.28 ± 0.02	1.29 ± 0.02
Avicel ph 102	1.31 ± 0.04	1.27 ± 0.01	1.35 ± 0.01
Avicel ph 105	1.44 ± 0.04	1.41 ± 0.01	1.44 ± 0.01

To facilitate the analysis two figures are presented. In Fig.21 are compare the HR values obtained with DensiTap® and GranuPack®. As depicted from the figure, from nine powder samples studied only five possess statistically similar values. Indeed, samples such as Tablettose 70, Food A, Food D and Food E presented a quite different flowability classification.

No correlation between particle's size, shape and true density were found that allowed to explain all HR differences obtained. For example, for the heaviest particles, Food B

(2.28 g/ml), HR values obtained with both devices are statistically similar, while for the lightest particles, Food E (1.23 g/ml), this correlation is no longer valid, suggesting a density dependency. Nevertheless, for the other seven powder samples with true density values between 1.42 and 1.57 g/ml non correlation can be done. Similarly for size- and shape-based analysis, no correlation has been found.

Upon impact, during tapping, a serial of physical phenomena takes place allowing wave propagation into the powder bed. Thus, despite similar free-falls height performances it is possible, even logical, to assume that the energy provided to the powder bed is device dependent, mostly attributed to materials properties. Under this perspective, from Fig 21. it seems that GranuPack® device supplies more energy to the system, but even here, no general conclusions can be drawn (Food E). Without any measurements allowing to quantify the amount of energy supplied to the powder samples, one point that could perhaps explain this HR differences is the presence of a "diabolo" in GranuPack® device. The "diabolo", deposited on the surface of the powder bed, is used in order to measure, by means of a sensor, the height of the powder bed after each tap. Diabolo's presence could change, mostly for low density particles, the stress patterns into the powder column and so could impact powders reorganization and finally the HR values obtained. Furthermore, it doesn't allow free surface reorganization. Thus, to avoid any misconclusion caused by diabolo's impact no other comparison using GranuPack® HR values is presented.

In Fig. 22, HR values obtained with our Vibratory device are compared with those obtained with DensiTap®. As depicted in the figure, HR values obtained with the Vibratory device are higher than those obtained with DensiTap®. These results validate our statement relating HR_U to a maximal state of compaction energy-dependent. Thus, the energy supplied to the powder bed during harmonically driven motion, as in particle dampers, can reach higher values than those obtained in fixed free-fall height devices. Other experiments allowing to measure the energy supplied to the system in this kind of devices should be carried out for validation.

In the case of Food E and Food C samples, $HR_{DensiTap®}$ values are higher than HR_U values. These two exceptions can be attributed to powders good ability to flow (Fig 20). Indeed, the assessment of powders flowability by compaction techniques is not suitable for good flowability powders because of their difficulties to compact. During vigorous vibration, at

high energy, just before an abrupt decompaction, the aeration of the powder bed can take place, inhibiting the system to reach a maximal compaction state.

3.3. ***General discussion: a critical view***

Non general-conclusions can be drawn relating flowability to the size distribution or the morphology (shape) of the powders. This is mostly attributed to the complexity of granular materials used in our work. The heterogeneity of our powder samples in terms of nature, shape, size distribution and physical properties (crystallinity, density, roughness, porosity…) makes quite difficult the analysis enabling to draw any general statement.

As Ribiere et al., 2007, we have stablished that non-ideal powders (non-spherical, large size distribution, cohesive …) subjected to tapping compact (packing fraction increases) and reach a stationary state that depends on the tapping intensity.

The study of metal powders or glass beads flowability using bulk densities ratio has been successfully achieved [6, 23, 24]. When referring to more complex samples, such as organic powders, numerous limitations has been highlighted when using HR to evaluated flow behavior such as low reproducibility, user-dependency, scattered results, amount others [4, 16]. Determining powders flow behavior using criteria such as HR, implies mastering many parameters, among them the powder mechanical history, that is to say the different mechanical stresses supplied to the powder. In this study, we have shown that HR is strongly related to the energy injected to the powder bed. The system approaches its optimally packed state for a given vibration intensity but not seems to be user-dependent or to present low reproducibility (Fig. 14 and 20). Classic vibration compaction devices such as Densitap (A = 3 mm and f_t = 4.16 Hz) and Granupack (A = 1 or 3 mm) supplied a quite fixed vibration energy to the powder samples under study. If this energy is not enough to allow the particles to reorganize, the HR value obtained will not be an accurate representation of the powder ability to flow.

Over 50 years later, Hausner reflection is still valid. During vibration the powder particles are forced to jump, and those to lose contact with each other for a moment. During this time there is no friction, between the particles. During this frictionless moment, the particles are able to rearrange and thus to compact [6]. **What has not been taken into account, is the *amount of energy needed to actually allow the particles to jump and rearrange***. What we define as the **HR_U**, is indeed, the bulk density ratio allowing the maximal powder rearrangement/compaction; refers to the elastic deformation of the particles and therefore **is an intrinsic characteristic of the powder** i.e. **independent of the tester used**. In fact, the maximal reorganization/rearrangement state of a powder, without any particles deformation can be achieved, or not when using a vibratory device under free surface conditions. The maximal reorganization state of a powder in a vessel is intrinsic to the powder. If the device used to estimate flowability does not allow to supply to the powder bed the energy needed to compact until its maximal compaction state, the system will not achieve the maximal reorganization but it will achieve a given compaction state. The higher the injected energy is, the more important the structural modifications are, in agreement with compacity fluctuations. Indeed, this input energy allows to move the system from one equilibrium state to another one. Thus, the powder compaction state depends on the amount of energy supplied to the system while the amount of energy will depend on the device physical and operating characteristics (amplitude and frequency of the vibration, system materials, …) but the maximal compaction state is not device dependent.

The HR_U, obtained at the maximum packing fraction of the powder bed, allows to relate HR to an intrinsic parameter of the system. Thus, HR_U will provide to some extent a better assessment of a powder flow behavior during granular compaction.

The data presented in this article are mainly given in order to stimulate additional work on the subject which will result in a better understanding of the flowability of powders.

4. Conclusions

Over the last several years, Hausner Ratio, and the closely related Carr Index, have become quite popular to evaluate powders flowability, despite the several limitations involved because of the simplicity and rapidity of the measure.

In order to study the consistency of HR with flow, three vibratory compaction devices were used. Different types of powders were examined, used in pharmaceutical and food industries, to cover a large range of applications.

As expected, the assessment of HR with DensiTap®, GranuPack® and our vibratory device, revealed at which point HR values can be modified as function of the dynamic parameters used. The amplitude and the frequency of the vibration, and thus the energy supplied to the system seems to be the major responsible of these fluctuations. A new approach is proposed to determine flow behavior by using HR based in the study of the energy supplied to the powder bed.

A number of observations can be reported regarding to the obtained results:

(a) The stationary state does not depend on the initial conditions of the powder bed;

(b) Non-ideal granular packings, as the ones used in this work, submitted to mechanical taps or sinusoidal cycles, can reach a stationary state that depends on the tapping intensity at a given frequency;

(c) HR obtained at the stationary state showed to be strongly dependent on the energy applied to the system. Indeed the input energy is related to the device used and so, on the experimental conditions such as the vibration amplitude;

(d) The energy supplied to the system will allow, or not, the reorganization of the grains (packing fraction increases);

(e) To obtain a HR value in consistency with the powder flow behavior, it seems necessary to determine bulk densities at the stationary state but also under a maximal compaction state. This value is named "**ultimate HR, (HR_U)**";

(f) Classical devices, fixed free-fall height, do not always allow to determine **HR_U**, because of energetic limitations**;**

(g) The **HR_U** value is an intrinsic property of the material and could be related the powder cohesiveness.

Finally, there is no doubt that HR, as a factor so simple to measure, has a great potential to determine flow behavior when taking into account the energy supplied to the system. The understanding of the effects of physical and mechanical properties on powders behavior can decrease the need to perform powder flowability analysis, resulting in significant time and resource saving.

The relation between **Ultimate HR** and interparticle forces interaction is the subject of a separate paper currently in preparation.

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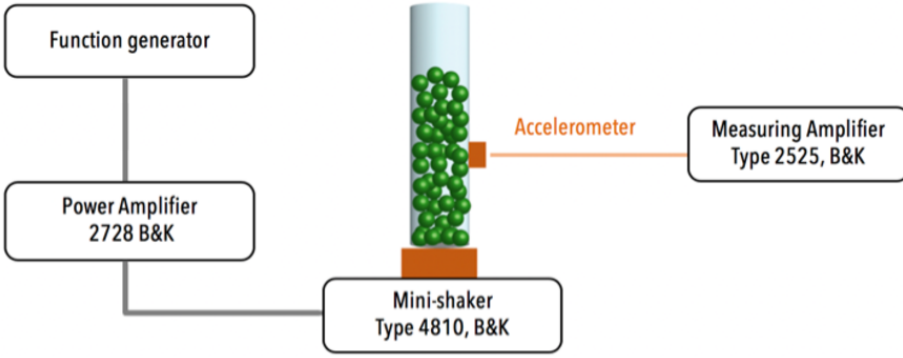
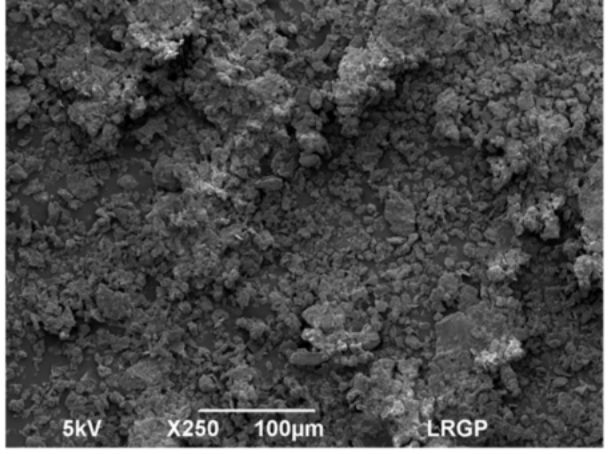
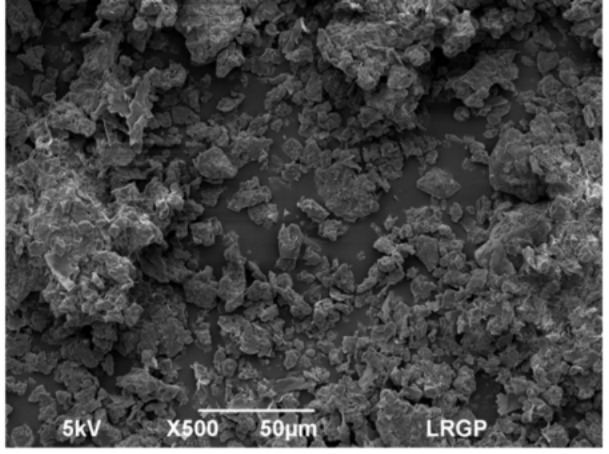
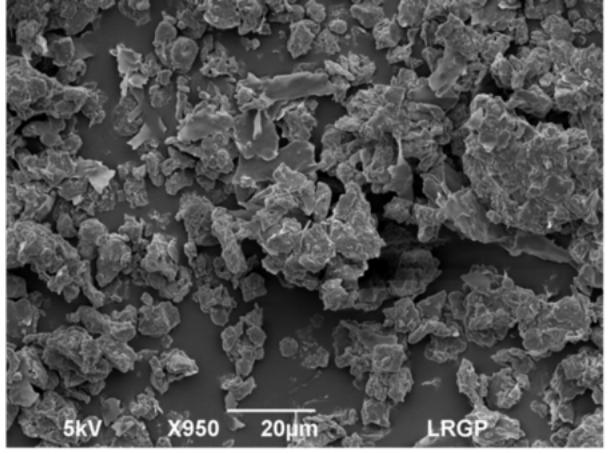

Function generator
Power Amplifier
2728 B&K
Mini-shaker
Type 4810, B&K
Accelerometer
Measuring Amplifier
Type 2525, B&K

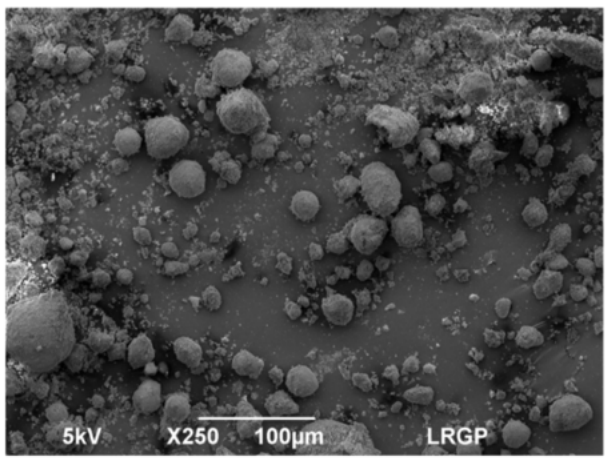
Food A

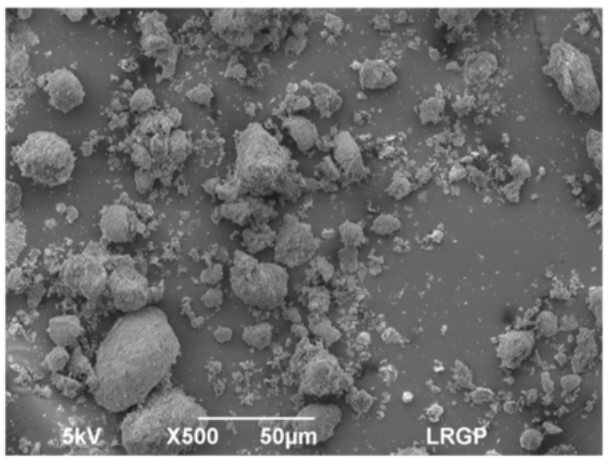


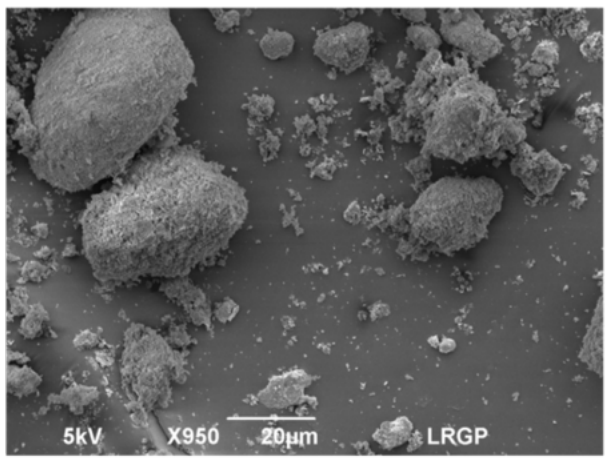




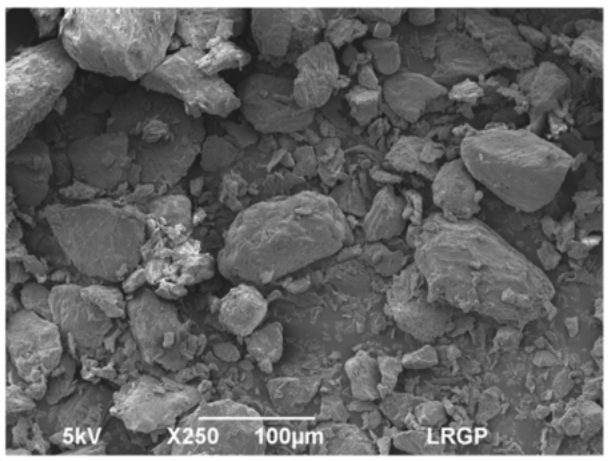
Food B

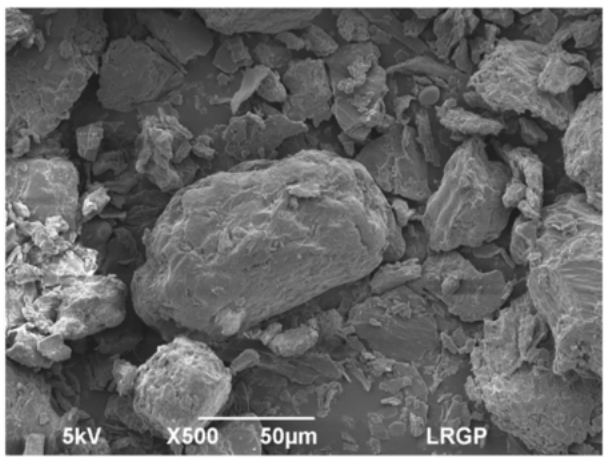


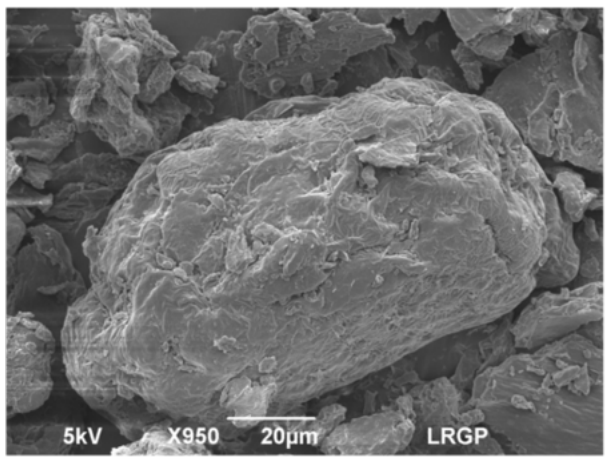




Food C







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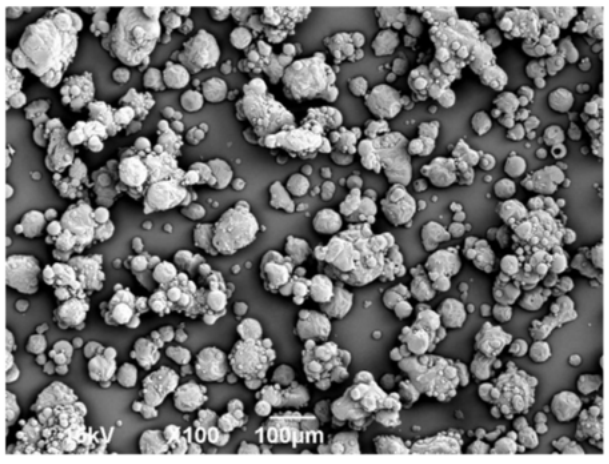
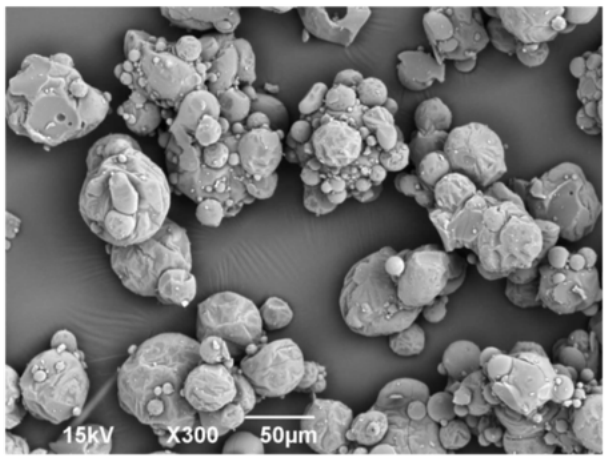
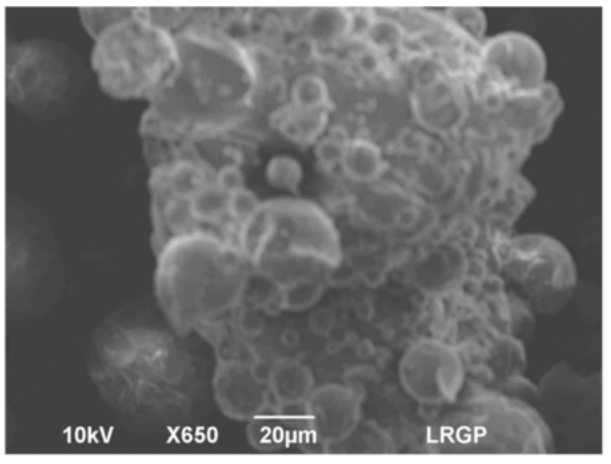
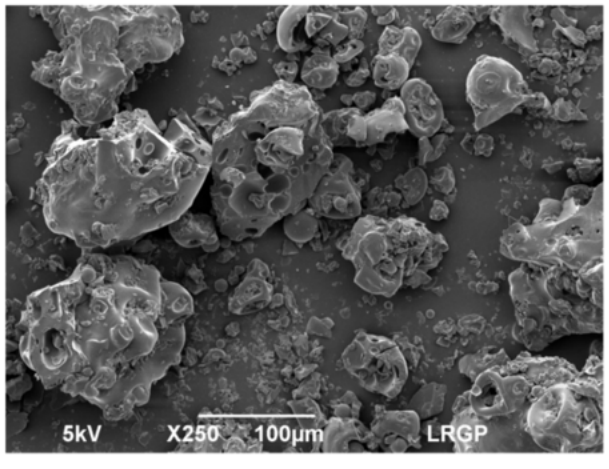
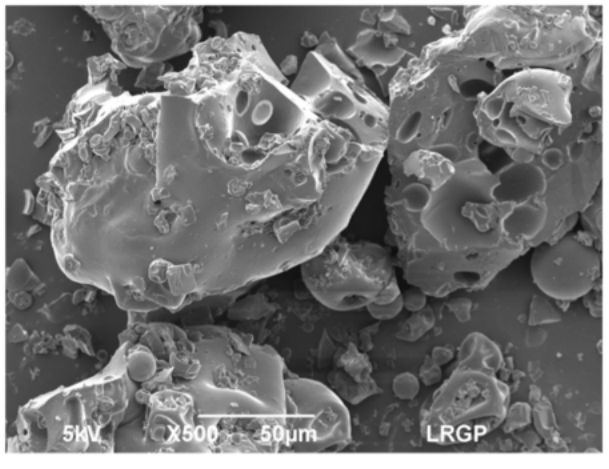


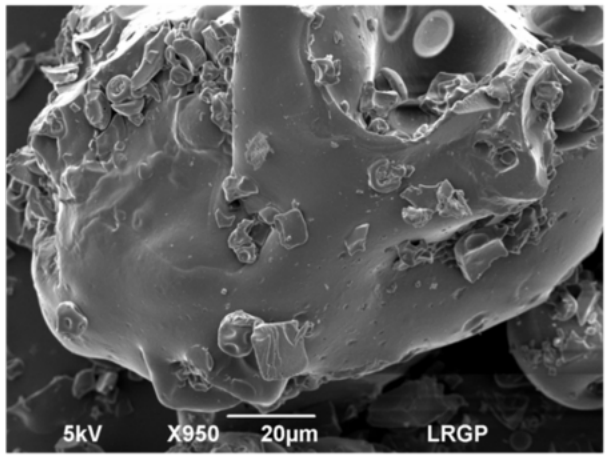




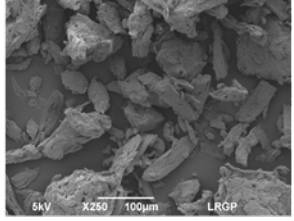
Food E

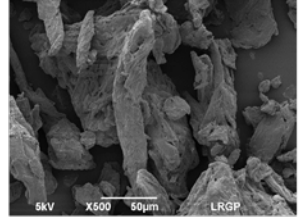


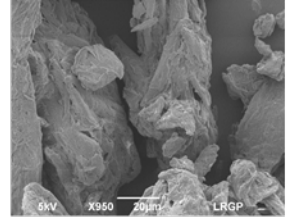




Avicel ph 102







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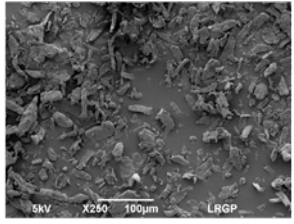
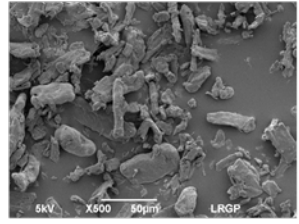
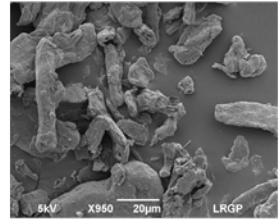
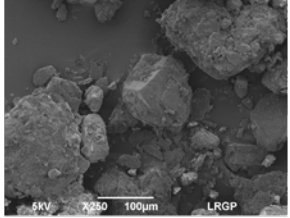
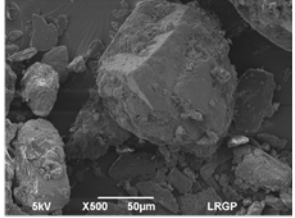


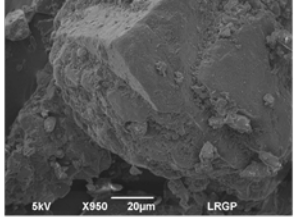




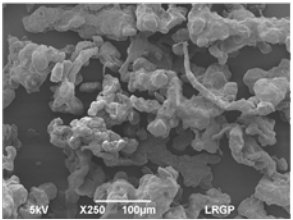
Tablettose 70

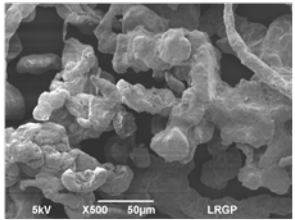


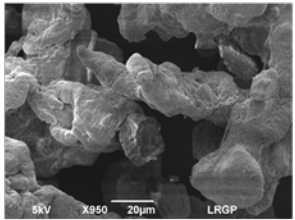


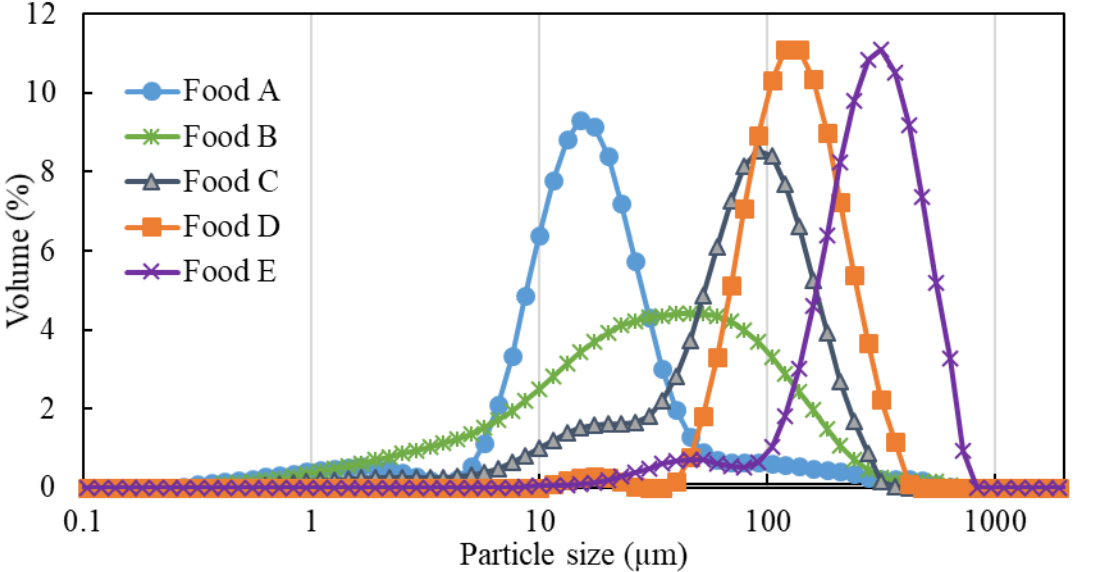


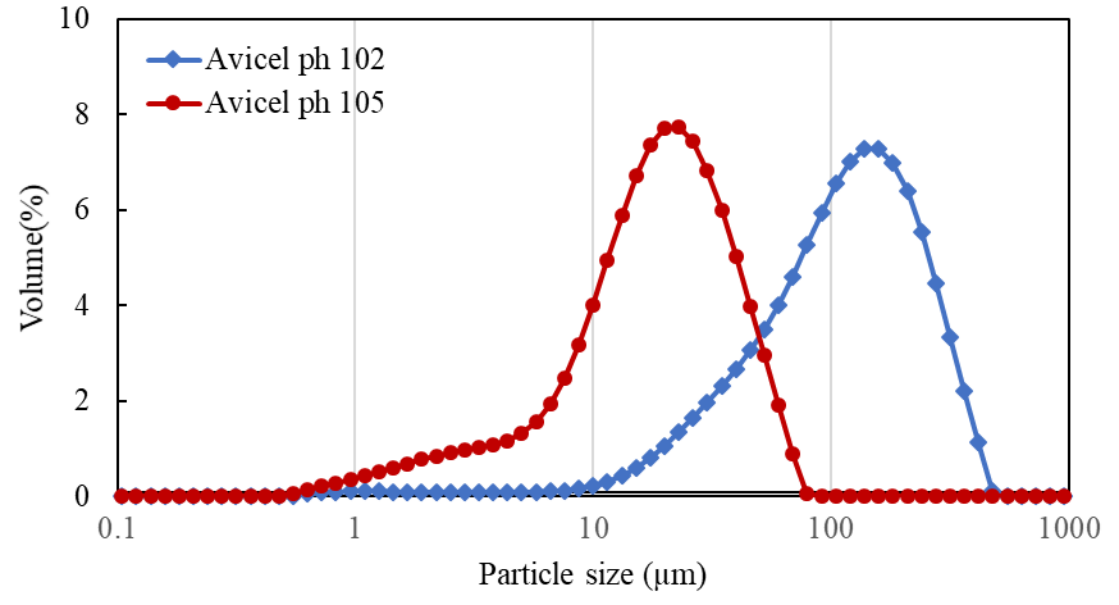
Retalac

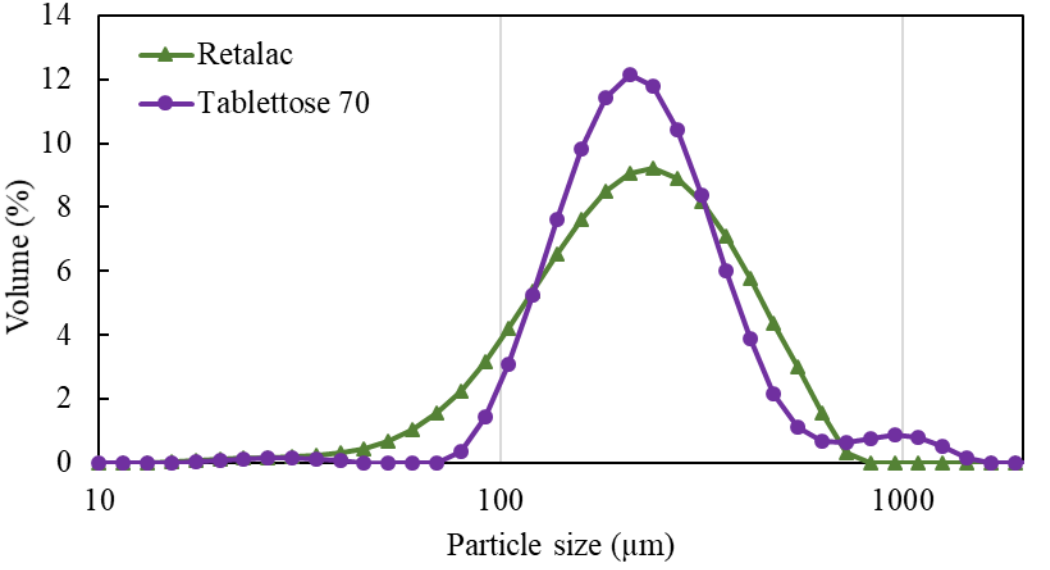


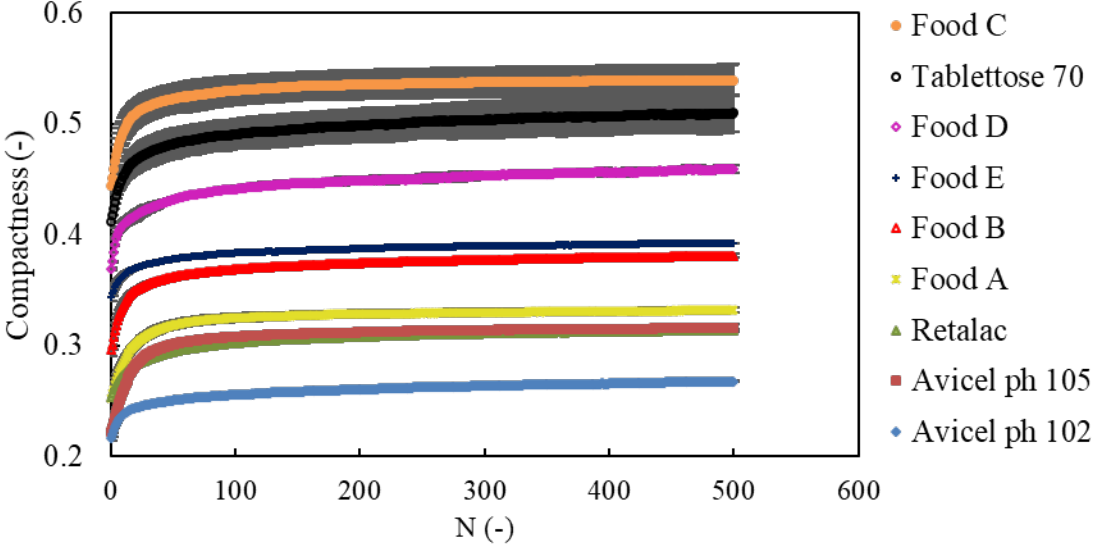


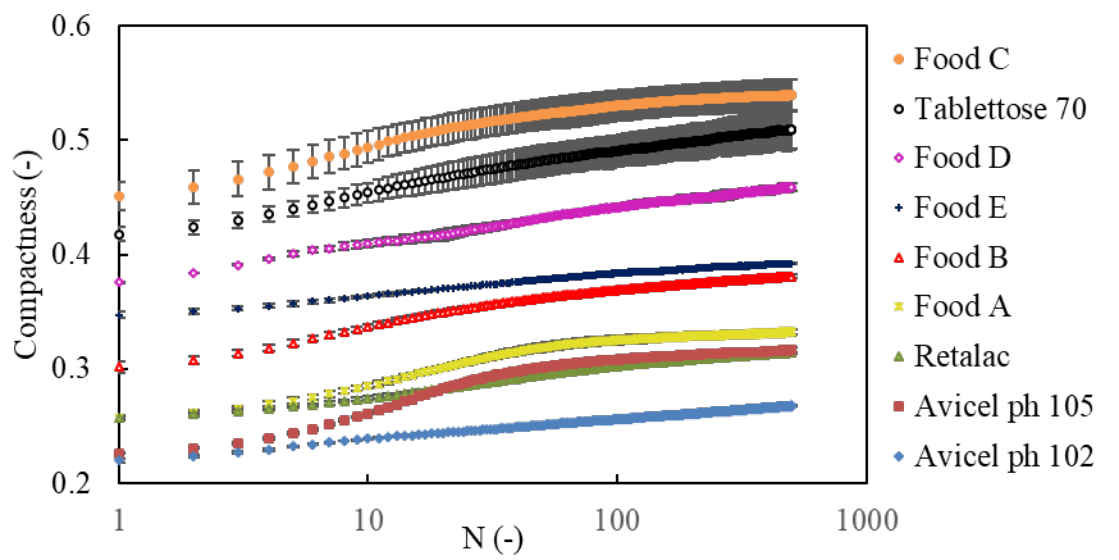


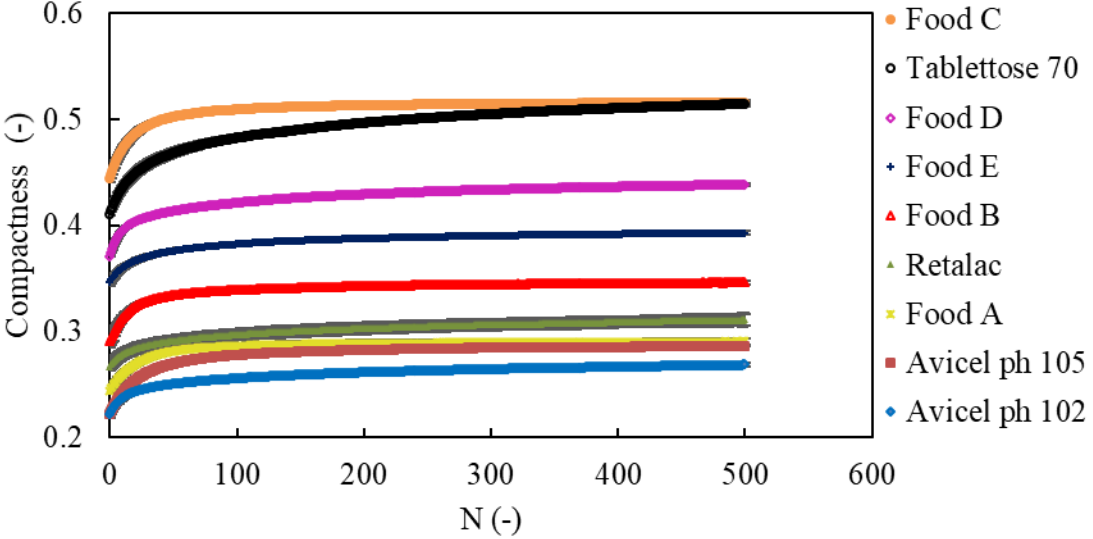

Food A
Food B
Food C
Food D
Food E
Volume (%)
0
2
4
6
8
10
12
0.1
1
10
100
1000
Particle size (μm)

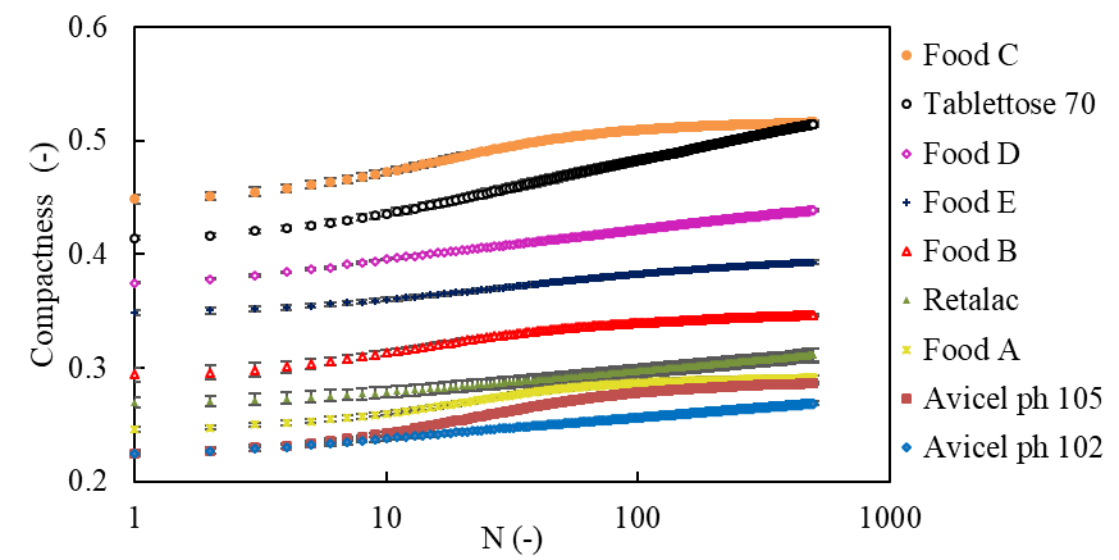

Avicel ph 102
Avicel ph 105
Volume(%)
10
8
6
4
2
0
0.1
1
10
100
1000
Particle size (μm)

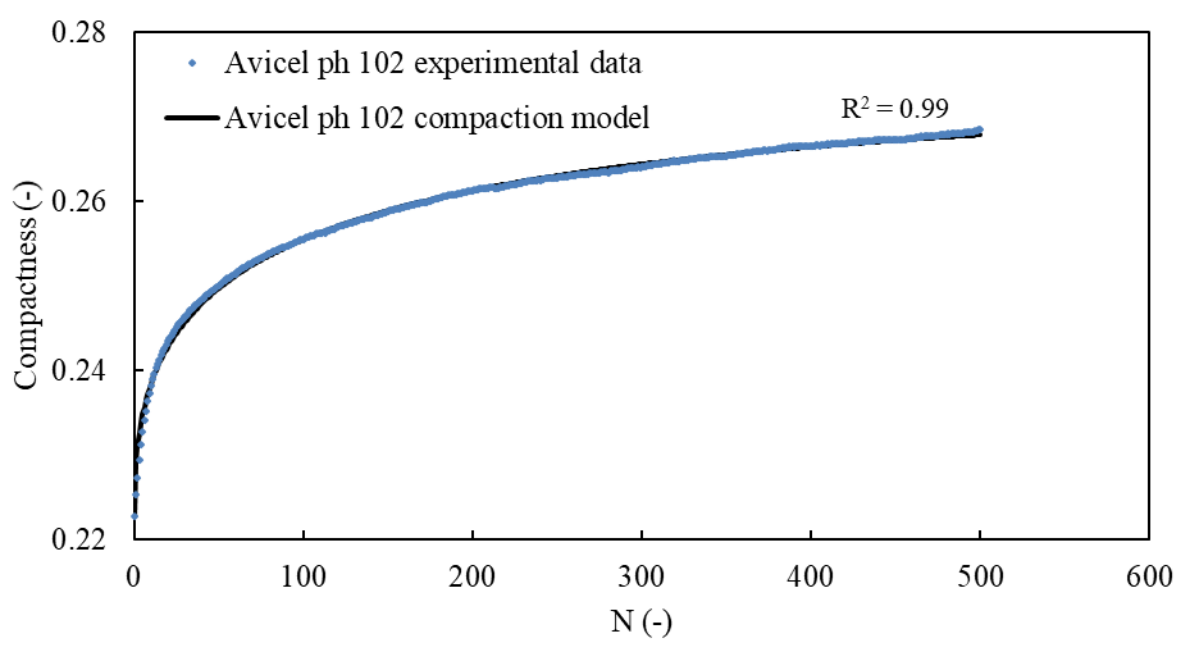

Retalac
Tablettose 70
Volume (%)
14
12
10
8
6
4
2
0
10
100
1000
Particle size (µm)

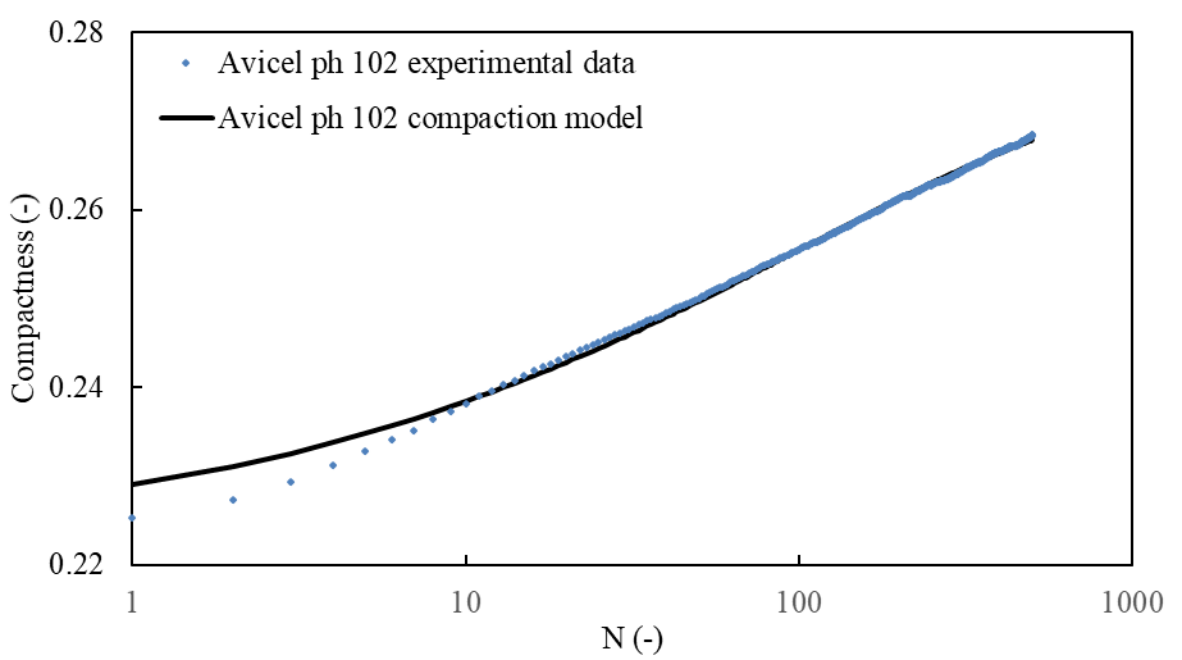

0.6
0.5
0.4
0.3
0.2
Compactness (-)
0
100
200
300
400
500
600
N (-)
Food C
Tablettose 70
Food D
Food E
Food B
Food A
Retalac
Avicel ph 105
Avicel ph 102

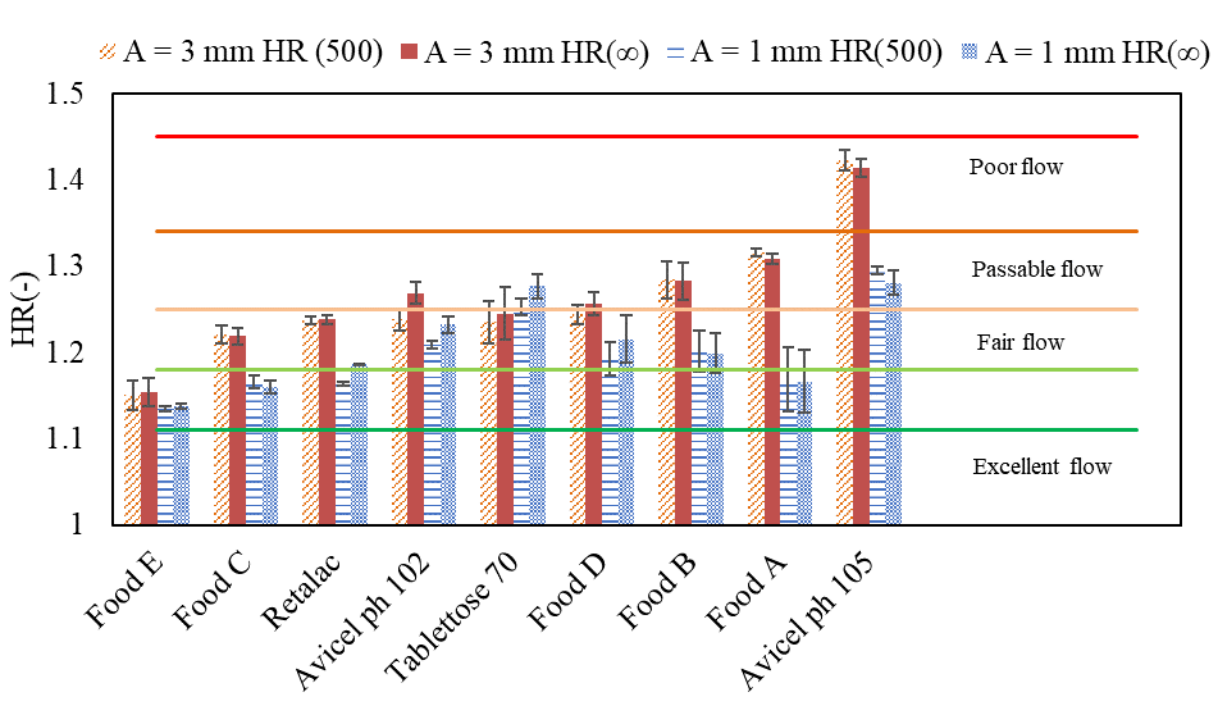

0.6
0.5
0.4
0.3
0.2
Compactness (-)
1
10
100
1000
N (-)
Food C
Tablettose 70
Food D
Food E
Food B
Food A
Retalac
Avicel ph 105
Avicel ph 102

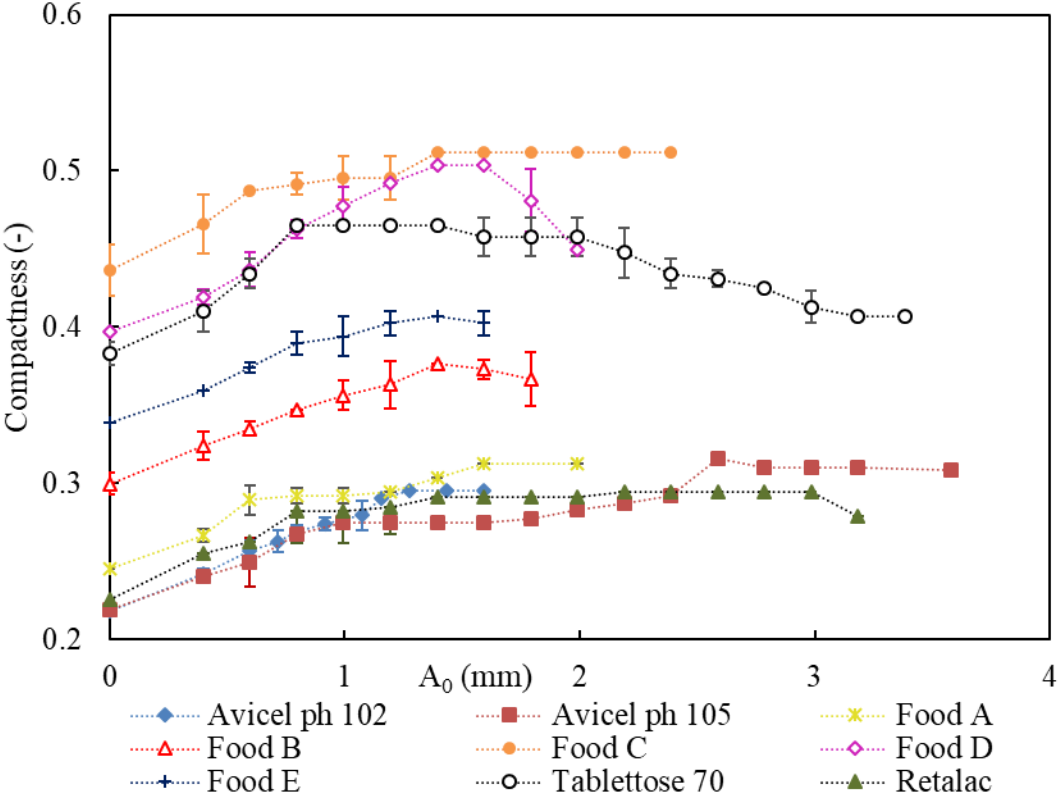

0.6
0.5
0.4
0.3
0.2
Compactness (-)
0
100
200
300
400
500
600
N (-)
Food C
Tablettose 70
Food D
Food E
Food B
Retalac
Food A
Avicel ph 105
Avicel ph 102

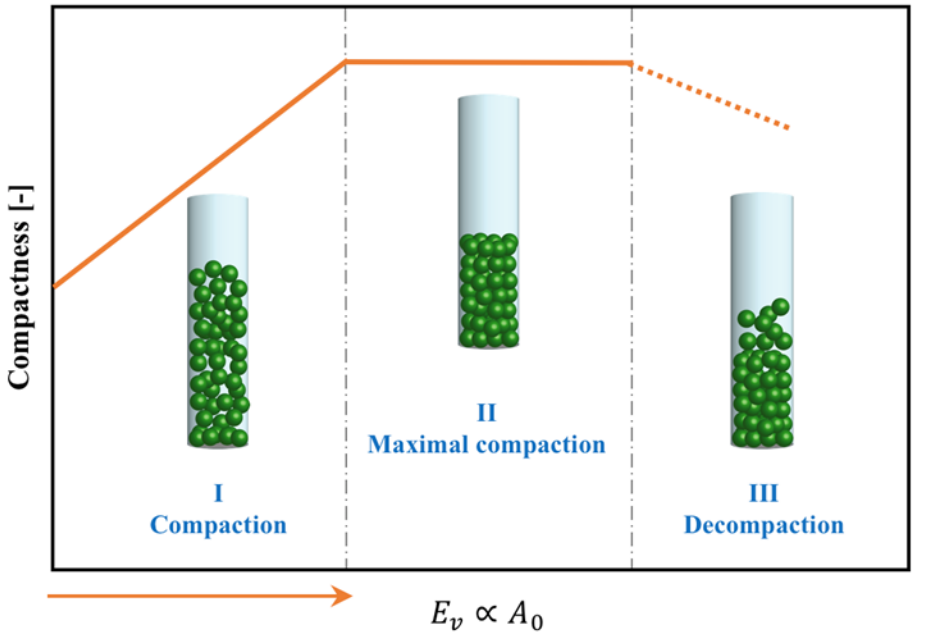

0.6
0.5
0.4
0.3
0.2
Compactness (-)
1
10
100
1000
N (-)
Food C
Tablettose 70
Food D
Food E
Food B
Retalac
Food A
Avicel ph 105
Avicel ph 102

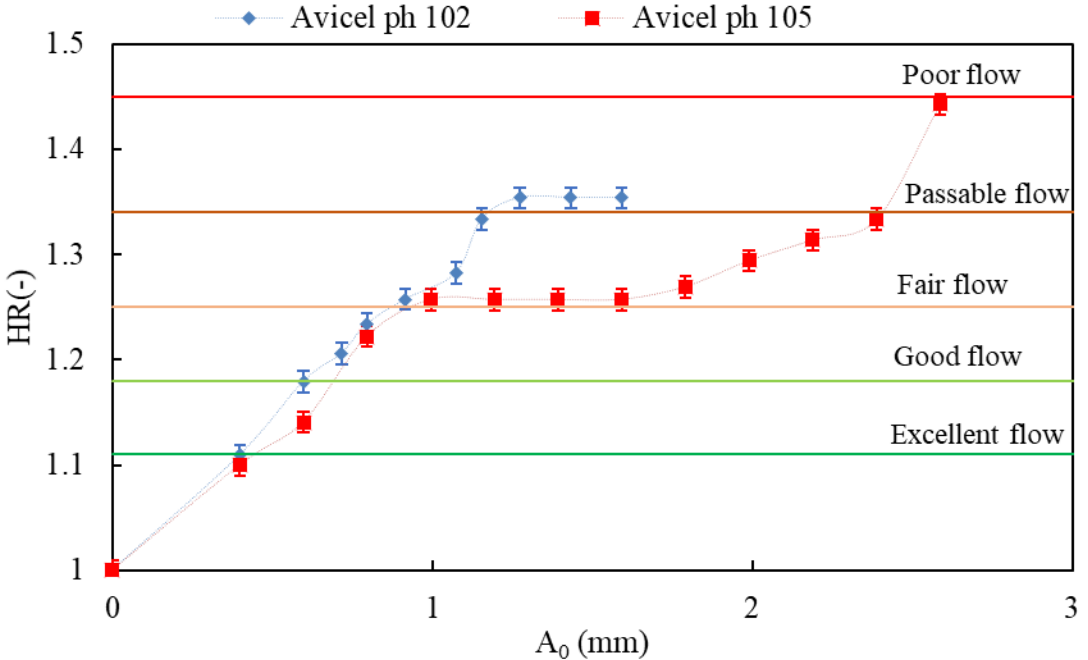

Avicel ph 102 experimental data
Avicel ph 102 compaction model
$R^2 = 0.99$
Compactness (-)
N (-)
0.28
0.26
0.24
0.22
0
100
200
300
400
500
600

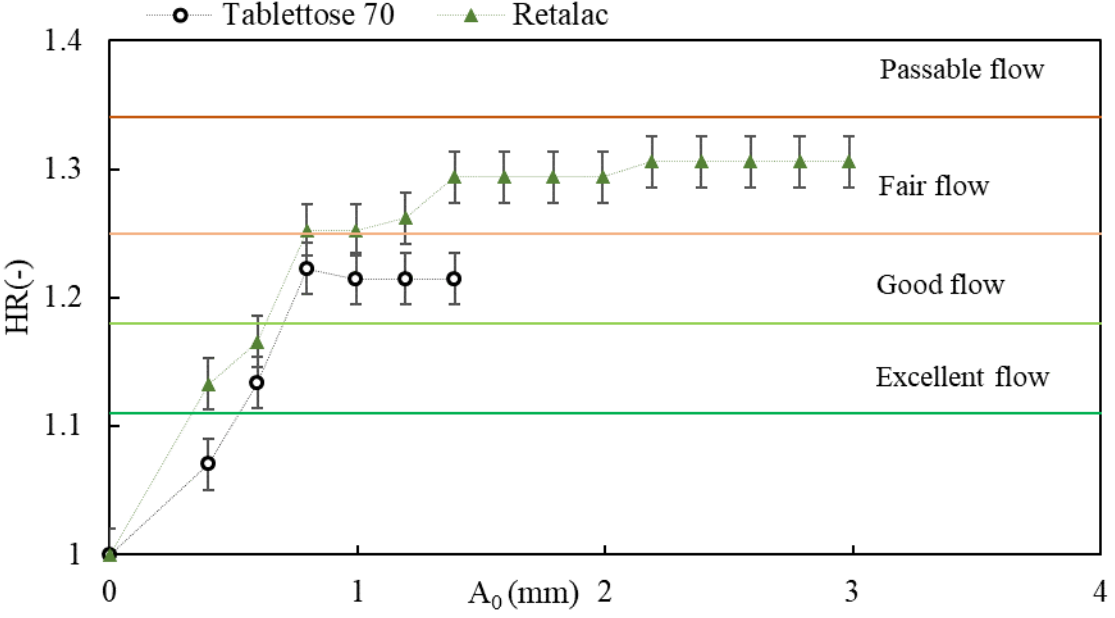

Avicel ph 102 experimental data
Avicel ph 102 compaction model
Compactness (-)
0.28
0.26
0.24
0.22
1
10
100
1000
N (-)

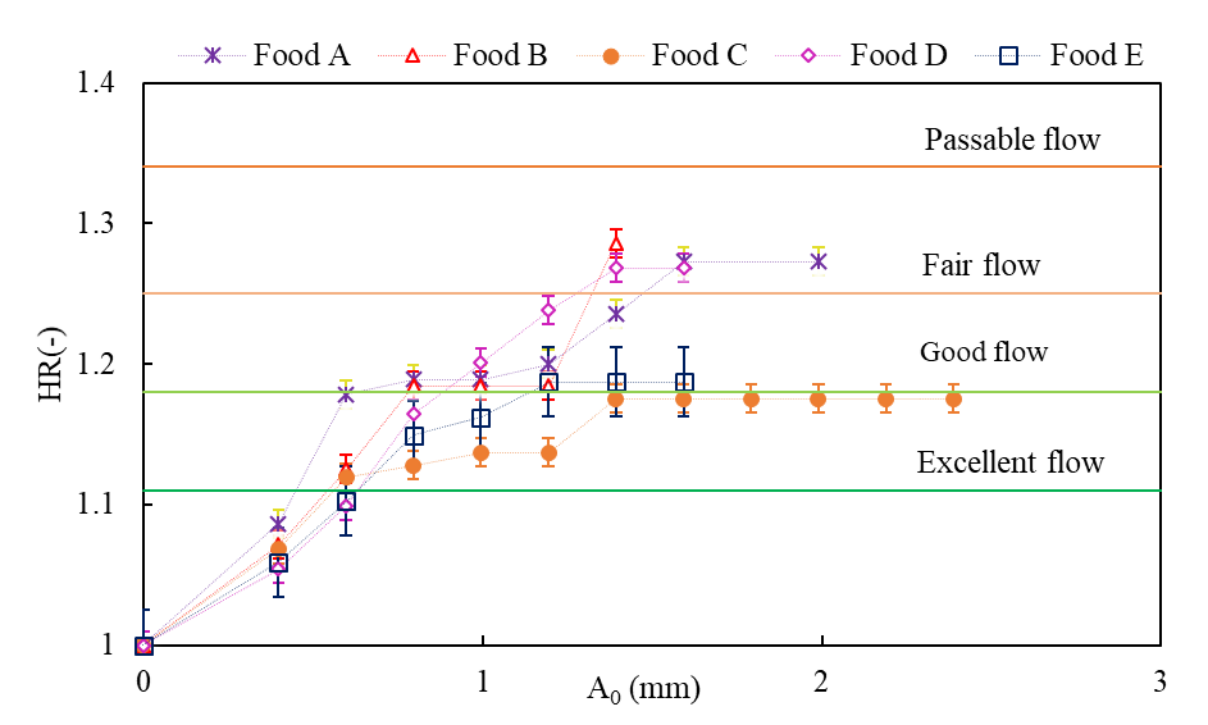

A = 3 mm HR (500)
A = 3 mm HR(∞)
A = 1 mm HR(500)
A = 1 mm HR(∞)
HR(-)
1.5
1.4
1.3
1.2
1.1
1
Poor flow
Passable flow
Fair flow
Excellent flow
Food E
Food C
Retalac
Avicel ph 102
Tablettose 70
Food D
Food B
Food A
Avicel ph 105

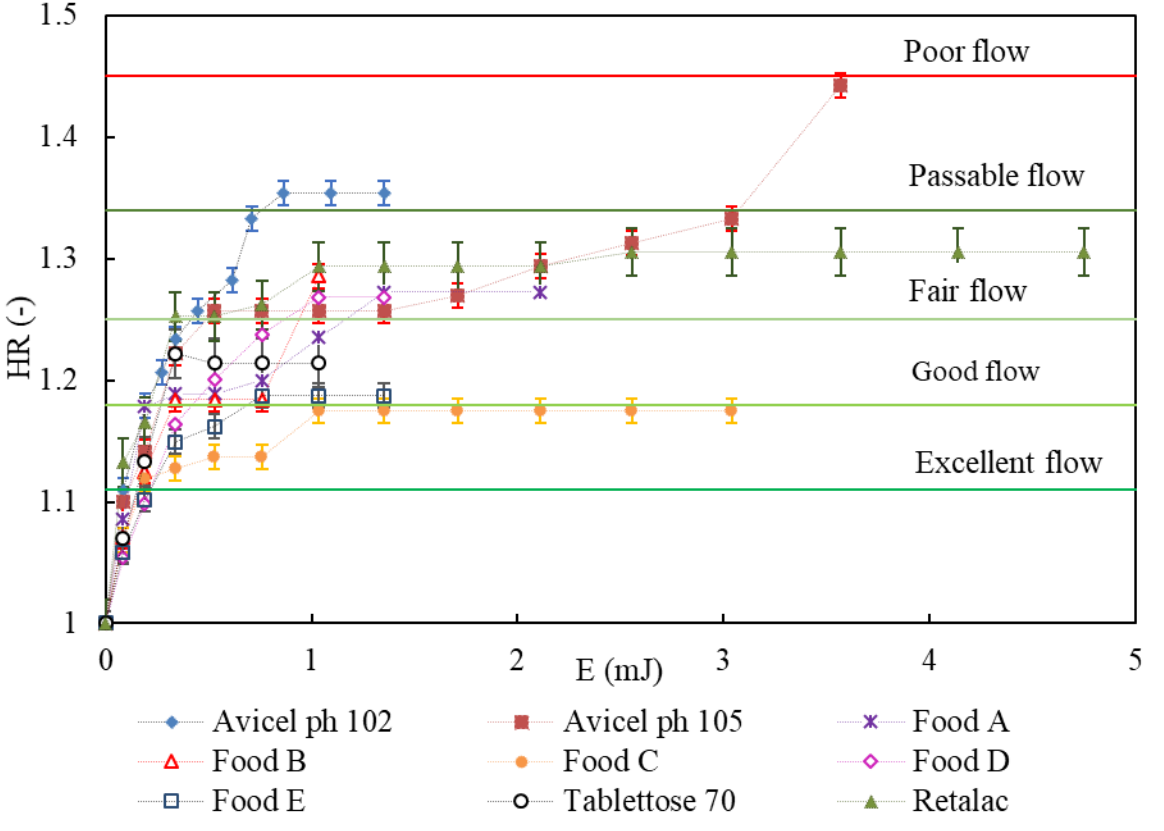

0.6
0.5
0.4
0.3
0.2
Compactness (-)
0
1
2
3
4
A_0 (mm)
Avicel ph 102
Avicel ph 105
Food A
Food B
Food C
Food D
Food E
Tablettose 70
Retalac

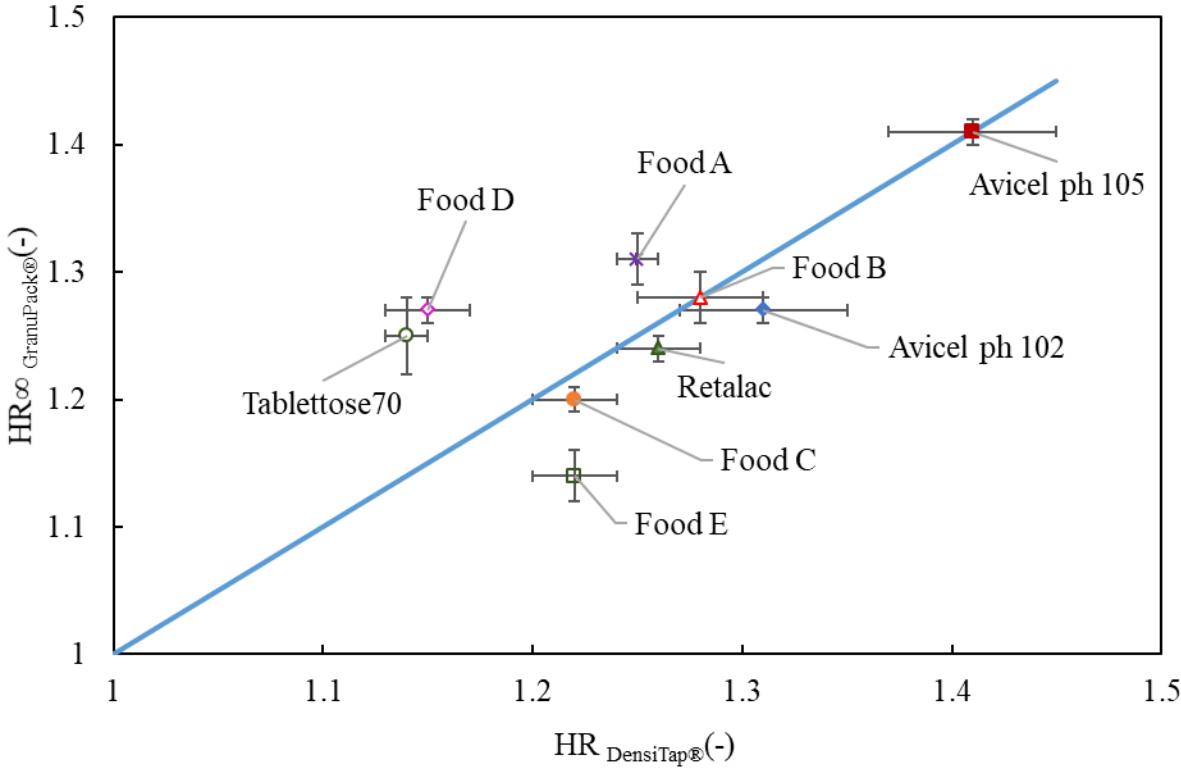

Compactness [-]
I
Compaction
II
Maximal compaction
III
Decompaction
$E_v \propto A_0$

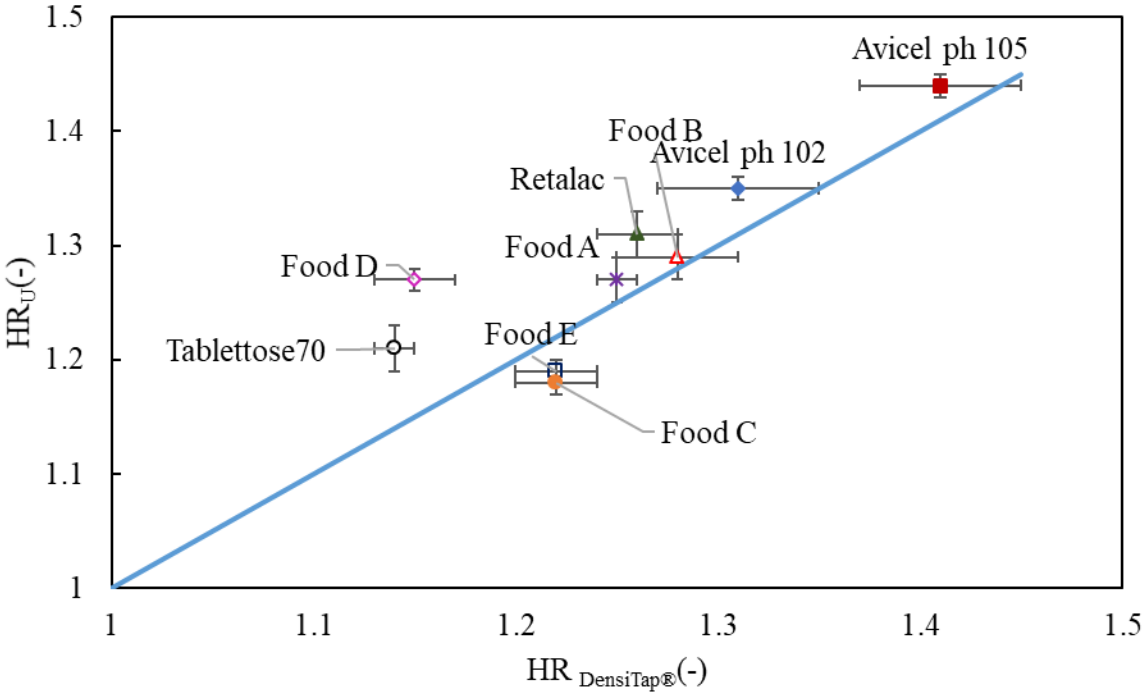

Avicel ph 102
Avicel ph 105
Poor flow
Passable flow
Fair flow
Good flow
Excellent flow
HR(-)
1.5
1.4
1.3
1.2
1.1
1
0
1
2
3
A0 (mm)


Tablettose 70
Retalac
Passable flow
Fair flow
Good flow
Excellent flow
HR(-)
1.4
1.3
1.2
1.1
1
0
1
2
3
4
A_0 (mm)

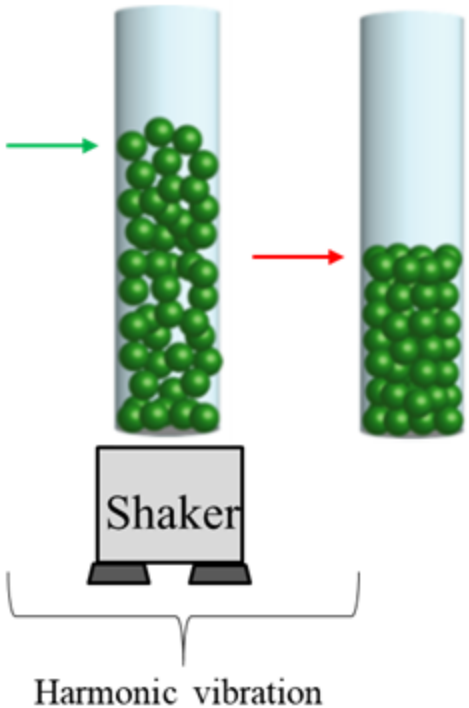

Food A
Food B
Food C
Food D
Food E
Passable flow
Fair flow
Good flow
Excellent flow
HR(-)
1.4
1.3
1.2
1.1
1
0
1
2
3
A_0 (mm)


1.5
1.4
1.3
1.2
1.1
1
HR (-)
0
1
2
3
4
5
E (mJ)
Poor flow
Passable flow
Fair flow
Good flow
Excellent flow
Avicel ph 102
Avicel ph 105
Food A
Food B
Food C
Food D
Food E
Tablettose 70
Retalac


1.5
1.4
1.3
1.2
1.1
1
1
1.1
1.2
1.3
1.4
1.5
HR∞ GranuPack®(-)
HR DensiTap®(-)
Food A
Food D
Food B
Avicel ph 105
Avicel ph 102
Retalac
Tablettose70
Food C
Food E


1.5
1.4
1.3
1.2
1.1
1
1
1.1
1.2
1.3
1.4
1.5
HR_UT(-)
HR DensiTap®(-)
Avicel ph 105
Food B
Avicel ph 102
Retalac
Food A
Food D
Food E
Tablettose70
Food C

$$HR = \frac{initial\ volume}{final\ volume}$$



$$E_v = \frac{1}{2} m (2\pi f)^2 {A_0}^2$$