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Development of a Dual Approach to Assess Powder Flow from Avalanching Behavior

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ABSTRACT The purposes of this investigation were to develop a method to evaluate flow properties of powders from avalanching tests and to detect similarities and relationships between these data and conventional powder flow properties. The API AeroFlow® automated flowability analyzer was tested using 6 pharmaceutical excipients. Data were presented as mean time to avalanche (MTA), scatter, and a classification based on the type of motion of the powder bed. Powders were also characterized in terms of particle size, particle shape, loss of weight on drying, Carr's compressibility index, and critical orifice diameter to prevent ratholing. A dual approach, which combines visual observation of the type of motion of the powder bed in the rotating drum with numerical descriptors such as MTA and scatter, was found to be more accurate in the assessment of powder flow than the current practice of using only MTA and scatter values. Statistical analysis established that there are relationships and similarities between the ranking of powder flow properties obtained from the avalanching test and Carr's compressibility index and the critical orifice diameter. An interaction between particle size and shape, both influencing powder flow, when evaluated with these methods was found. The assessment of the flowability of powders on the basis of avalanching tests should include both the determination of numerical descriptors of flow such as MTA and scatter, and a determination of the type of motion of the powder bed in order to increase the sensitivity of the method to small changes in powder flow properties.

KEYWORDS: Avalanching of Powders, Carr's Compressibility Index, Critical Orifice Diameter, Perceptual Mapping, Powder Flow

INTRODUCTION

There are 2 differences between the behavior of a powder and the behavior of matter in any other state. First, a powder can expand or contract over a limited range by voidage change and remain in the expanded or contracted state afterward. Second, although shear forces can deform a powder so that it can flow like a viscous liquid, the shear force required to produce deformation is a function of the load applied in the direction at right angles to the shear plane. Powder behavior is therefore difficult to quantify and to predict [**1**].

The treatment of powders as a continuum provides a method to describe their macroscopic properties. However, force or energy transmission inside a powder bed is possible only via the solid-solid interface between single particles. As a consequence, different test methods, which vary in the nature and degree of force and energy transmission, will quantify the macroscopic properties of a powder in a different way. Therefore, different test procedures, for example, for powder flow will provide different answers and classifications of powders.

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The underlying physical phenomenon of interparticulate interactions has been exploited in the assessment of powder flow by evaluation of packing properties through bulk density determination such as Carr's compressibility index [2], Hausner's ratio [3], and Kawakita's constant [4]. Other methods of predicting powder flow include shear cell measurement [5] and determination of the critical orifice diameter [6]. With a shear cell such as the Jenike shear cell [7,8] or the annular shear cell [5], the shear strength of the powder is evaluated under the influence of different normal loads. For Carr's compressibility index, the values are reliable only if certain equipment specifications and working protocols are adopted [9–11]. While Carr's compressibility index was somewhat useful in predicting capsule filling performance [12,13], Trowbridge et al. [14] could not identify a relationship to tabletting performance. The critical orifice diameter determination has been used successfully to predict arching and plug formation in capsule filling [15,16] and in the manufacture of mini-tablets [17].

This work is concerned with the assessment of powder flow using an avalanching powder system and comparing it to the methods of determining Carr's compressibility index and the critical orifice diameter approach. A commercial system to study avalanching in powders was first described by Kaye et al [18].

Powder avalanching in a rotating drum evaluates dynamic powder flow characteristics based on the theory of deterministic chaos [19]. Powders are sensitive to their initial conditioning, and a small perturbation of the system changes their subsequent behavior [20]. Although any chaos has its order [21], the underlying causes of the apparently arbitrary behavior and structural organization have not yet been determined [22]. Results from avalanching measurements are described as "strange attractor plots," or numerical values are sought to quantify the avalanching process. A strange attractor plot is constructed by joining points defined by the time between a set of avalanches (t_n, t_{n+1}) and similar subsequent events (t_{n+2}, t_{n+3}); thus 2 events are needed to define a single point in the 2-dimensional strange attractor plot. The centroid of this plot is called the mean time to avalanche (MTA). The expansion of the strange attractor plot in X- and Y-direction reflects the time scatter of the avalanching process. MTA is a measure of the flowability of the powder, and the scatter value defines the regularity of the flow behavior [14]. A powder with good flow properties will have an MTA close to zero and a low scatter value.

Henein et al [23] described 6 different types of powder movement in a rotating cylinder, all of which can also be seen when monitoring powder movement during an avalanching experiment. The rolling bed motion appears desirable for powder mixing [24], whereas cascading and cataracting are signs of poor powder flow. It is assumed that powders that show slipping behavior give a lower MTA value, because these powders climb the drum wall only slightly before the powder bed slips and an avalanching event is detected [14]. Yet, these powders do not necessarily flow. Without visual observation, a low MTA value due to powder bed slippage could be mistaken for an indication of good powder flow properties.

MATERIALS AND METHODS

Materials

Six commonly used excipients were chosen for this study: microcrystalline cellulose (Avicel PH 101®, "MCC PH101"; Avicel PH 102®, "MCC PH102"; FMC Corporation, Cork, Ireland); lactose monohydrate crystals ("LR"; Borculo Whey Products, Saltney, UK); lactose monohydrate "Fast-Flo" ("LFF," Foremost Ingredients Group, Baraboo, WI); pregelatinized maize starch (Starch 1500®, Colorcon, Dartford, UK); and calcium phosphate, dibasic anhydrous ("DCP," Monsanto Co., St. Louis, MO). Isopropyl alcohol (BDH, Poole, UK) was used as a cleaning agent.

Sample Conditioning Prior To Testing

In ambient room conditions (35–50% relative humidity, 24° C ± 1° C), a bag containing a thin layer of a sample was left open for 2 hours, and the sample was periodically redistributed. The environmental conditions were monitored using RS Tinytalk® II probes (RS Components, Corby, UK) and a Rotronic Humidity Probe Model Hygromer®, MI Rotronic Instruments Corp., Huntington, NY).

Loss of weight on drying

Powders were dried to mass constancy in a drying oven (Pickstone Equipment Ltd., Rutherford, UK) at 105° C. Results are the mean of 3 determinations.

Carr's compressibility index

The tap density apparatus (Model SVM10, Copley/Erweka, Nottingham, UK; lift height 3 mm, tapping frequency 150 taps/min) was used to determine Carr's compressibility index. The tared 250-mL measuring cylinder was filled with test material to 100 mL, and the volume and weight of the material were noted. With minimal disturbance to the measuring cylinder, it was transferred to the tap density apparatus. The method of determination recommended in USP-NF 7th Supplement (p. 3936/7) was used.

Critical orifice diameter determination

Conditioned excipients were tested using the modified Flodex® apparatus (Model 21-101-050, Hanson Research Corporation, Chatsworth, CA). A suitable disk diameter was selected and mounted onto a cylindrical stainless steel hopper that was 7.2 cm in height and 5.7 cm in inner diameter. After the plunger was secured, the hopper was completely filled. If the powder passed the orifice of a disk under the influence of gravity, the test was repeated using a disk of smaller diameter. The reduction in disk diameter was continued until no flow was observed. The test was repeated 3 times using the diameter above that, which gave no flow.

Determination of avalanching behavior

The API AeroFlow® (Model 0-8030, Amherst Process Instruments, Hadley, MA) was used in a Bigneat cabinet (Bigneat Ltd., Hants, UK) to guarantee complete protection of the equipment from external light.

The equipment consisted of a clear cast acrylic drum with an inner diameter of 15 cm. An etched stainless steel collar can be placed on the drum's circumferential wall to prevent the sample from sliding on the drum wall instead of avalanching. A light source was positioned vertically in front of the drum so that the bottom of the powder sample cast a shadow onto an array of photovoltaic cells. This array detected the position of the powder in the drum in real time, and the raw data collected by a computer at a sampling rate of 5 measurements per second consisted of the detector array output and the time. Schematic drawings of the equipment layout have been presented in detail by Kaye et al [18,19].

The drum, drum cover, and etched metal collar were flushed out with pressurized air between tests of the same sample and thoroughly cleaned with isopropyl alcohol or other suitable solvents between investigations of different materials. When cleaning the drum with solvent, the drum was flushed with pressurized air and dried. Between runs of the same material samples, all equipment used was thoroughly cleaned using the same procedure as for the cleaning of the drum between different material samples. The rim of the drum cover and the port were greased using food grade grease between tests of different sample materials. The drum and drum cover were sprayed with antistatic spray and wiped dry. The etched metal collar was then fitted into the inner circumference of the drum wall, 50 mL of test sample was loaded into the drum, and the drum was sealed. The filled drum was mounted onto the port and the light source was aligned vertically. The velocity of the drum's rotation is limited to a maximum of 30 and a minimum of 200 seconds per revolution. The program was run for 5 minutes at a defined rotational velocity with the doors of the Bigneat cabinet open to observe the flow behavior. This was followed by 5 runs with the program set for 20 minutes at the same rotational velocity, conducted with the doors closed. A set of preruns was performed to determine the apparent optimum in the rotational velocity of the drum, using LR and DCP as test materials. At a drum velocity of 145 seconds per revolution the flow pattern appeared to be more consistent than at all other rotational velocities. Therefore, all main experiments were performed at this drum velocity.

Equipment-dependent software was used to determine the mean time to avalanche (MTA) and the scatter.

Particle size determination

The Malvern Mastersizer (Model S, Malvern Instruments Ltd., Worcestershire, UK) was used to determine the equivalent volume of sphere diameter (d_v). All powders showed standard left-shifted distribution functions, and because laser diffraction data are always fitted to a suitable distribution function, the measures of spread of the particle size distribution were found to correlate with the mean size obtained. Hence, the latter appeared not to add further information to the particle size characteristics, and only the mean size will be reported here.

Particle shape analysis

A small amount of powder on a glass slide was suspended in 3 drops of an appropriate immersion liquid (Refractive index liquids, series RF, McCrone Scientific Ltd., London, UK). A cover slip was placed to allow the suspension to settle evenly between the 2 glass surfaces. The shape factor [25] was measured by a combination of microscopy and image analysis (Seescan Solitaire 512, Seescan, Cambridge, UK; BH-2, Olympus, Hamburg, Germany), and 586 particles were examined for each excipient.

RESULTS AND DISCUSSION

Avalanching behavior

The humidity range (35–50%) under which experiments were carried out was below the critical level of 55%, the level at which larger capillary condensation of moisture and capillary forces develop [26]. Hence, it can be assumed that the equilibrium moisture contents found (MCC PH101: 5.35%, MCC PH102: 4.56%, LR: 5.38%, LFF: 5.67%, Starch 1500: 9.60%, DCP: 1.04%) are typical for the powders tested, and the powders should exhibit genuine flow behavior as a result of their particulate properties only.

The 4 types of powder bed motion observed were "rolling" (flow category 1), "slumping" (flow category 2), "slipping" (flow category 3), and "cataracting" (flow category 4). These are illustrated schematically in **Figure 1**.

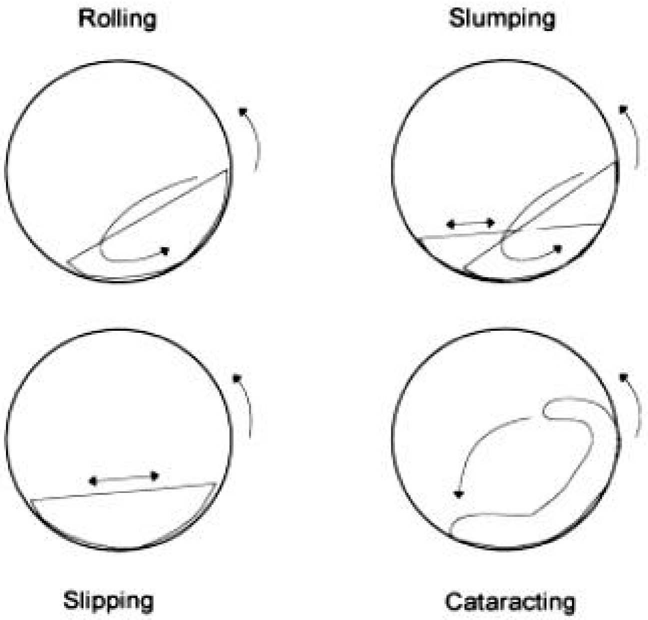

Figure 1. Types of powder bed motion (flow category) observed in this study.

As the value of the flow category increases, powder flow becomes worse. The MCC powders presented a mixed behavior. The outer layer of the powder adhered strongly to the drum walls cataracting at times, while the majority of the powder bulk showed slumping (flow category 2). This could be attributed to a larger difference between the adhesion forces acting between powder particles and drum wall, and the autoadhesion forces acting between individual powder particles, which are obviously much smaller.

The powder bed movement of LR also followed the slumping pattern (flow category 2), but autoadhesion and adhesion forces appeared to be more balanced, and more powder fell off the edged metal collar of the drum. A mixture of slumping and predominantly slipping (flow category 3) was observed for Starch 1500, indicating a reduced adhesion force between powder particles and drum wall. The typical powder bed movement for DCP was cataracting (flow category 4), hence representing the poorest powder flow. The best powder flow properties would be expected for

LFF, because here mainly rolling (flow category 1) was observed, occasionally interrupted by a slumping event.

The predominant types of powder bed motion or flow category obtained from visual inspection of the flow pattern, the MTA, and the scatter values are summarized in **Table 1**, together with a ranking based on both observation and numerical values — "the dual approach".

Table 1. Ranking, MTA, and Scatter Values for the 6 Powders Obtained From Avalanching Experiments

Powder	Flow category*	MTA	Scatter	Ranking
				(dual approach)
DCP	4	4.45 ± 0.21	1.46 ± 0.21	6
LFF	1	2.80 ± 0.06	0.99 ± 0.04	1
LR	2	3.02 ± 0.08	1.06 ± 0.11	2
MCC PH101	2	7.26 ± 0.10	2.40 ± 0.18	4
MCC PH102	2	5.11 ± 0.46	2.21 ± 0.06	3
Starch 1500	3	9.55 ± 0.32	4.11 ± 0.18	5

*Predominant type of powder bed motion observed for the powder: 1=rolling; 2=slumping; 3=slipping; 4=cataracting; MTA, mean time to avalanche

In the dual approach, priority is given to the visually observed mode of powder motion, and the lower the flow category, the lower the rank number. In similar categories of motion, the powder is ranked according to the MTA value, and the lower the MTA value, the lower the rank number. The scatter values were directly proportional to the MTA and therefore did not provide further information. Hence, they were not considered here, but in principle these values could provide a third ranking criterion, if in 1 flow category 2 powders are characterized by the same MTA value.

Carr's compressibility index

The results for Carr's compressibility index are listed in **Table 2**.

Table 2. Carr's Compressibility Index and Critical Orifice Diameter for Different Powders Tested*

Powder	CI (%)	COD (mm)	d_v (μm)	NS
DCP	32	24	14.72	7.32
LFF	15.3	14	99.29	10.4
LR	13.6	20	144.21	6.91
MCC PH101	27.5	26	70.51	7.2
MCC PH102	22.6	24	115.03	7.18
Starch 1500	21.3	24	77.84	7.4

*CI, Carr's compressibility index; COD, Critical orifice diameter; d_v, volume diameter; NS, shape factor as defined by Podczeck [25].

The 2 brands of lactose monohydrate should be classified as free-flowing powders, Starch 1500 and MCC PH102 provide fair flow properties, whereas the other materials appear to have poor (MCC PH101) or very poor (DCP) flow properties.

To identify interrelationships between Carr's compressibility index and the avalanching behavior — here represented by the dual approach — a 3-dimensional graph was constructed showing Carr's index as a function of the MTA and the flow category (**Figure 2**).

Due to the limited number of observations, statistical modeling appeared inappropriate. However, the graph shows some trends. First, Carr's index increases with increasing MTA. Second, there is some tendency for an increase in Carr's index with flow category. These results emphasize the need for using the dual approach rather than characterizing the powders solely on the basis of strange attractor plots and their derived numerical values such as MTA and scatter.

Critical orifice diameter

The results for the critical orifice diameter are summarized in **Table 2**. The values obtained are significantly larger than those reported by authors such

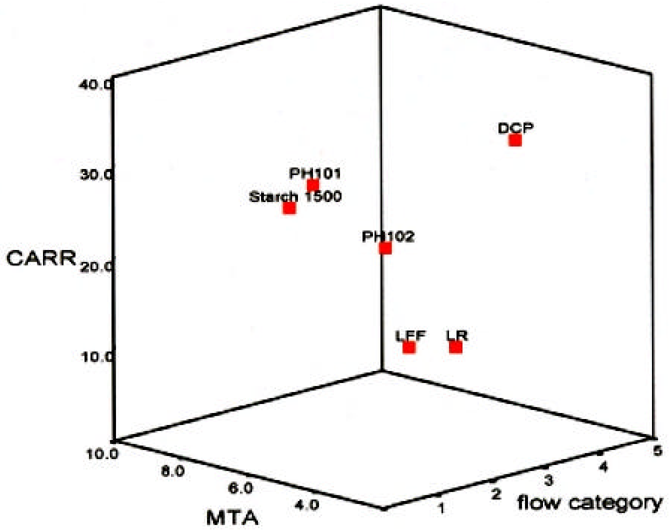

Figure 2. Carr's compressibility index (CARR) as a function of the mean time to avalanche (MTA) and the type of powder bed motion (flow category) observed.

as Flemming and Mielck [17]. Flow through an orifice is dependent on the hopper size, as well as the standing time of material, the filling procedure, and other operator-related variations of protocol. Prolonged standing time, for example, could cause consolidation of the powder under the influence of its own weight, which in turn causes larger critical orifice diameters to be determined. In addition, it has to be considered that Flemming and Mielck [17] used a conical funnel as opposed to a cylinder used here. The correct determination of the critical orifice diameter, however, is based on the physical principles of arch formation in cylinders [27], allowing the use of such values, for example, for stress analysis in powders [28] and plug strength calculations [16]. The physically correct critical orifice diameter is equal to the smallest circular opening through which "ratholing" can occur, that is, the diameter at which only the powder above the orifice will flow out, while the rest of the powder remains in the cylindrical hopper forming a tube-like assembly. The conical funnel used by Flemming and Mielck [17], however, leads to a complete powder discharge, most certainly dominated by powder–hopper wall friction instead of only interparticulate forces. The values reported here are, therefore, the physically correct ones and hence represent the true powder properties.

The results for the critical orifice diameter are shown in **Figure 3** employing a similar technique used for Carr's compressibility index. Here, 2 clear trends can be observed. The critical orifice diameter increases with an increase in MTA, and there is also a strong increase depending on the type of motion of the powder bed (i.e. flow category). The relationship between the results obtained from the 2 flow assessment methods is much more obvious than for Carr's compressibility index.

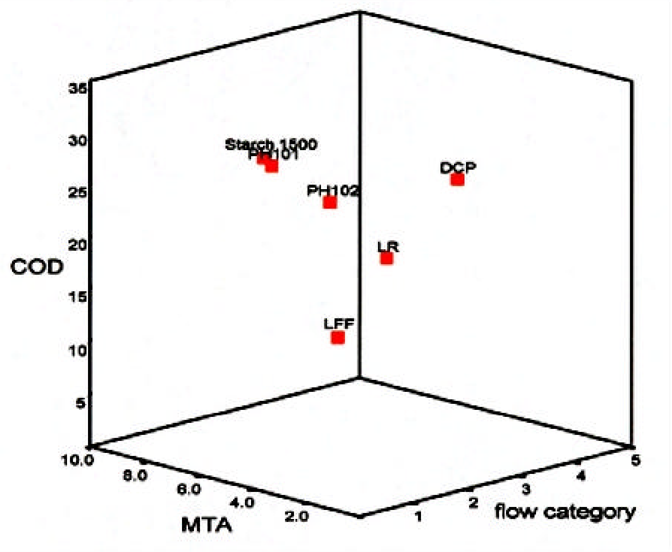

Figure 3. Critical orifice diameter (COD) as a function of the mean time to avalanche (MTA) and the type of powder bed motion (flow category) observed.

Particle size and shape

Particle size and shape factor values are listed in **Table 2**. A common observation in powder technology is that an increase in particle size increases powder flow and makes the powder bulk behave less cohesively. However, the influence of particle size and shape on the autoadhesion forces between individual particles is not easy to quantify [26], because additional factors such as surface roughness, surface free energy, and electrostatic charges must be considered [11]. Hence, it cannot be expected that powder flow properties are predictable from particle size and shape values. Additionally, the definition of particle shape is not a simple procedure [29]. The shape factor used in this work [25] indicates larger differences in particle shape between the materials, especially LFF. This is not surprising, because the material is a spray-dried product comprising agglomerates rather than single particles. Although these agglomerates are irregular with respect to a larger number of edges in the 2-dimensional particle outline, they are generally round and hence free-flowing aggregates. In **Figure 4**, the complex relationship between particle size, shape, and rank number obtained from avalanching experiments using the dual approach (**Table 1**, column 5) is illustrated in a 3-dimensional graph.

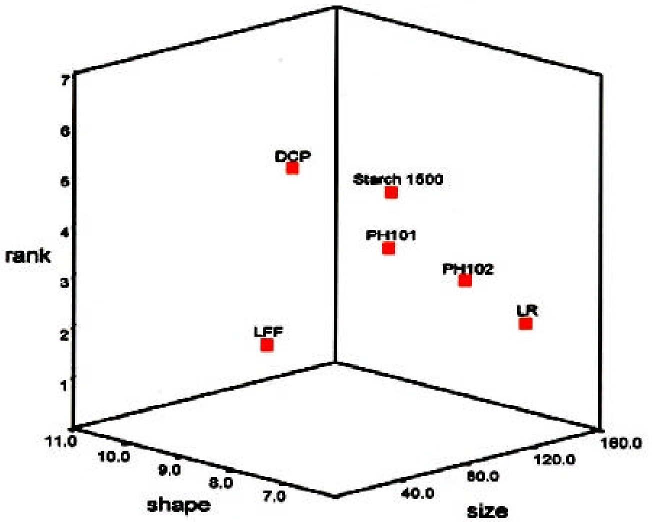

Figure 4. Relationship between the powder flow behavior estimated from avalanching experiments using the dual approach (rank) and particle size and shape.

With the exception of LFF, the rank number increases with decreasing particle size and increasing value of the shape factor. Except for LFF, the shape factors generally represent angular particles, but the regularity of the geometric figures increases with decreasing shape factor. Hence, rolling in the rotating drum appears to become hindered if the particle shape becomes more irregular. For a similar particle shape the increase in particle size improves powder flow.

Multivariate data comparison

To identify global relationships and similarities between the different flow properties, a perceptual map was constructed on the basis of nonlinear principal components [30] using the SPSS ("Statistical Package for Social Sciences", Version 8, SPSS Inc., Woking, UK). The rankings from the dual approach, the type of bed motion, MTA, the scatter value, the critical orifice diameter, and Carr's compressibility index were used as raw data. **Figure 5** shows the perceptual map obtained.

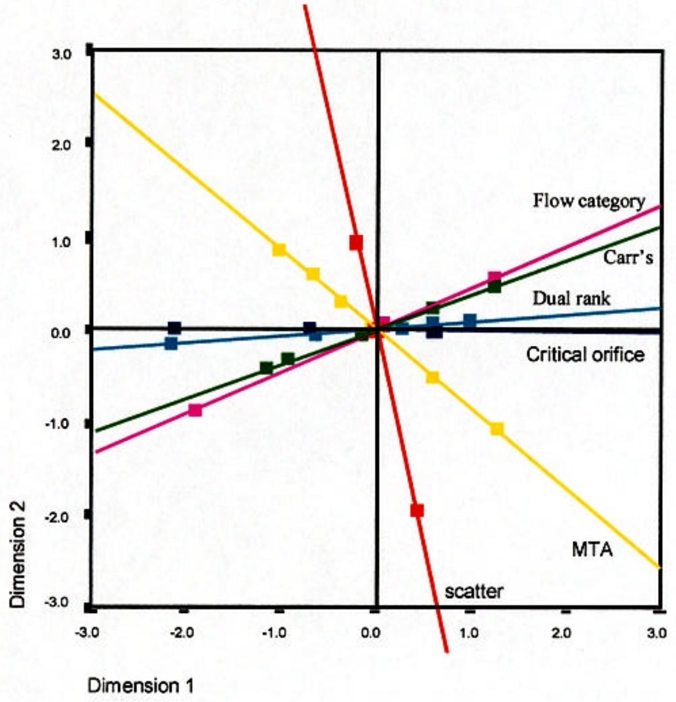

Figure 5. Similarities and differences between the different methods of powder flow assessment investigated.

Two main nonlinear principal components (Dimensions 1 and 2 in **Figure 5**) represent the information contained in the data. The information can be condensed to a single point in the coordinate system for each property measured and each powder. It is then possible to construct correlation lines linking the points for each powder, with each line representing 1 measured property. Properties that have a similar origin for their behavior or represent similar information (ie, are closely linked) have their correlation lines close to each other. Here, this is the case for (a) the type of motion of the powder bed (flow category) and Carr's compressibility index, and (b) the dual ranking result and the critical orifice diameter. On the other hand, the MTA or the scatter value alone is not linked to Carr's compressibility index or the critical orifice diameter. Comparing the similarities indicated in the graph with the raw data it can be seen that, for example, a rolling bed motion and a low value for Carr's compressibility index, and cataracting and a high value for Carr's compressibility index are connected. The smaller the critical orifice diameter, the lower the rank number of the powder in the dual approach. Only MCC PH101 breaks this relationship for unknown reasons. The isolation of the MTA and scatter values from the other measured properties is demonstrated in **Figure 5**. The close connection of the dual ranking results emphasizes again that the dual approach proposed is more useful in describing powder avalanching than single numbers such as MTA and scatter values. Hence, the dual approach might be useful as a predictor of powder flow.

CONCLUSIONS

All current techniques for determining powder flow are based on different principles and have advantages and disadvantages. The choice of an appropriate technique to assess powder flow should be made in relation to the technological process to be studied. Yet, there appears to be some similarities between the various methods available.

When using an avalanching approach to determine flow properties, the results cannot be based solely on strange attractor plots or derived numerical values such as mean time to avalanche or scatter. A dual approach, which combines a visual observation of the type of motion of the powder bed with the numerical values, appears more accurate in the assessment of powder flow.

Statistical analysis established that there are relationships and similarities between the ranking of powder flow properties obtained from the avalanching test and Carr's compressibility index and the critical orifice diameter. Also, there is an interaction between particle size and shape, both influencing powder flow, when evaluated with these 3 methods.

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