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Original Article

Impact of fine particles on the rheological properties of uranium dioxide powders

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ABSTRACT

This study aims at characterizing the rheological properties of uranium oxide powders for nuclear fuel pellets manufacturing. The flowability of these powders must be compatible with a reproducible filling of press molds. The particle size distribution is known to have an impact on the rheological properties and fine particles (<100 μm) are suspected to have a detrimental effect. In this study, the impact of the particle size distribution on the rheological properties of UO_2 powders was quantified, focusing on the influence of fine particles. Two complementary approaches were used. The first approach involved characterizing the powder in a static state: density, compressibility and shear test measurements were used to understand the behavior of the powder when it is transitioned from a static to a dynamic state (i.e., incipient flow conditions). The second approach involved characterizing the behavior of the powder in a dynamic state. Two zones, corresponding to two characteristic behaviors, were demonstrated for both types of measurements. The obtained results showed the amount of fines should be kept below 10 % wt to ensure a robust mold filling operation (i.e., constant mass and production rate).

1. Introduction

The integrated technology demonstrator called ASTRID (Advanced Sodium Technological Reactor for Industrial Demonstration) is a sodium-cooled fast reactor (SFR) currently in design phase [1]. This reactor will use a different fuel from that of pressurized water reactors (PWR). SFR fuel has specific characteristics that requires a suitable manufacturing process. The fuel pellets are obtained in two main steps: first, the powder is shaped by uniaxial compression (pressing step), and it is then sintered in a second step (sintering step). The pressing step should be optimized to minimize density gradients in the pellets for a given target weight. Indeed, density gradients result in a deformation of pellets during sintering (imperfect cylindrical shape), which can lead to a need for rectification because tolerances are often low on diameter variations (typically given in hundredths of millimeter). The need and amount of rectification are then directly linked to the variation of the weight of pellets around the target. Consequently, before the actual pressing step, a reproducible, homogenous and fast filling of press molds is needed, with the adequate powder weight. For this purpose, optimal flow properties are required, to avoid detrimental defects (chips, cracks, end-capping effect, off-specification dimensions and weight, etc.) as well as rectification of the pellets.

The current reference powder does not have the properties required to ensure a robust mold filling operation (i.e., constant mass and production rate), which would allow a direct pressing phase and minimal rectification. Indeed, at the current stage of research, the pellets may require rectification after sintering. It is not only costly to rectify the sintered pellets in this manner, but also radiologically penalizing because it generates radioactive dust. The aim of this study is therefore to modify the characteristics of the powder in a suitable manner to achieve a reproducible, homogenous and fast filling of press molds and thus avoid detrimental defects and rectification.

Fu et al. [2] investigated the effect of particles shape and size on the flow and consolidation of lactose powders. Their results indicated that the shape and size of particles both significantly affect the flowability of the powder. The influence of particle size on flowability is also described in several publications [3–5]. According to Li et al. [3], granular materials can be classified into two categories: powders smaller than 100 μm (called fines) and those

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larger than 100 μm. Fine particles are suspected to have a strong impact on powder behavior. Firstly, cohesion forces (Van der Waals forces) are considered predominant in these fines. Secondly, fines have a strong impact on the reorganization of the powder bed and can cause stacking defects [6]. The fines can adsorb to the surface of larger particles, increasing frictional forces and thus resulting in a poor flow. In other cases, the fines can coat the surface of the coarse particles and insert into their contact points, increasing the inter-particle space and reducing the attractive forces, which improves the flowability [7].

There are different methods for characterizing the flow properties of a powder for good filling conditions [2], making it possible to characterize the powders in their industrial context [8]. Leturia et al. [9] compared the traditional powder flow characterization methods with powder rheometer and shear cell measurements. They concluded that the choice of a powder characterization methods depended on the process conditions to be replicated. The flow properties of a powder could not be characterized by a single index but by a combination of different tests.

Generally speaking, a powder can be characterized under incipient or dynamic state. To understand its rheological properties, it is necessary to observe how the powder moves from a static to a dynamic state (i.e., incipient flow conditions), and thereafter how it behaves in dynamic mode [10]. Methods of observation include density measurements, compressibility and shear tests. Other methods are used to characterize the powder flow and the filling behavior, i.e., dynamic rheometer measurements, permeability and aeration measurements.

The aim of the present study is to characterize the influence of the quantity of fine particles on UO_2 powder mixtures flowability. It should be kept in mind that from the industrial point of view, the main goal is to produce fuel pellets having a constant weight and a homogenous microstructure, maintaining the desired production rate and avoiding detrimental defects and rectification. For this purpose, the powder must have adequate flow properties and its bulk density should not be sensitive to the fines content (in order to obtain a constant filling weight). Moreover, in the fabrication chain, from the storage silo to the filling of press molds, the powder is transferred between several unit operations and undergoes different stress levels. The flowability of the powder through the entire chain should be adequate in order to produce fuel pellets having the expected properties (constant weight and homogenous microstructure). It is thus required to characterize the powder flow properties under both high and low stress levels, as well as incipient and dynamic states.

In this study, a powder with a broad particle size distribution is used, in order to identify the maximum permissible fines content, to ensure a reproducible and homogenous press mold filling operation at the industrial level. Preliminary experiments were carried out on three granulometric classes of UO_2 granules [11,12]: 0–100 μm, 100–200 μm and 200–300 μm. The population of fine particles (<100 μm) was shown to correspond to a very cohesive powder with a poor flowability [11,12]. In this work, five powder batches are first prepared, with different amounts of fines. Several flow characterization indices are then compared directly as a function of the fines content in the powder mixtures.

To our knowledge, this is one of the first papers dealing with UO_2 powders flowability. The characterization of this kind of materials constitutes a challenging task because the maximum information has to be obtained from the smallest possible sample size. Indeed, due to its radioactivity, a minimum amount of UO_2 powder has to be used in order to minimize the wastes generated by the flowability test methods. Consequently, the same sample may be used for successive characterization tests. The order of execution of the tests has to be chosen so that the sample properties are not modified (agglomerates rupture, attrition, etc.). The optimal tests sequence followed in this work was established and validated by preliminary studies [11,12].

2. Materials and methods

In this work, all the UO_2 powder samples were stored and handled in a glove box (Fig. 1b), where the Relative Humidity and Temperature were maintained constant (T = 20 °C and RH = 40%). The different tests were also performed in similar glove boxes under the same Relative Humidity and Temperature conditions.

2.1. Powder mixtures preparation

The raw powder was first compressed at 150 MPa to obtain granules (Fig. 1b). At this pressure, it was possible to obtain granules that were strong enough to be handled and studied, but that would break easily during pressing without having any effect on the final pellet microstructure. A sieve grinder (Freewitt-type OxcilloWitt-Lab) was used to perform grinding and sieving in a reproducible manner. It was thus possible to obtain reproducible granules with little operator effect. The obtained granules were considered as representative of the dense agglomerates obtained after a preliminary milling step as can be observed in the current reference industrial process. Reproducibility and repeatability were demonstrated via measurements of the particle size distributions with a dry granulometer (Malvern Mastersizer 3000).

The maximum size of the granules was about 550 μm. The fine particles (<100 μm) were separated by sieving and then added once again to the population of large particles, in order to obtain several "calibrated mixtures" (i.e., with a controlled fines content). In this way, five powder batches (A, B, C, D and E) were prepared with different fines contents: 0, 10, 30, 50 and 100 wt% of fines, respectively. The five "calibrated mixtures" (= 5 powder batches) were obtained using a Turbula mixer for 10 min at a speed of 23 min^{-1}. The particle size distributions of these five batches were measured using a laser granulometer and are presented in Fig. 2.

The median volume diameter Dv_{50} can be used to describe the particle size distribution, supplemented by the width of the particle size distribution, also called Span. The span is defined according to the expression:

$$Span = \frac{Dv_{90} - Dv_{10}}{Dv_{50}} \quad (1)$$

Dv_{10}, Dv_{50} and Dv_{90} represent the particle diameters for which respectively 10%, 50% and 90% of the volume of the population is below these diameter values (indicated in Table 1).

For each type of measurement (cohesion measurement, flow energy, etc.), the standard deviation is determined on the basis of three measurements for each powder batch:

$$Standard\ deviation = \sqrt{\frac{\sum(x - \bar{x})^2}{N - 1}} \quad (2)$$

where x is the value of the measurement, $\bar{x}$ the average value and N the number of measurements for the same powder.

2.2. Density measurements (tapping test)

The bulk density of a powder is defined as its mass divided by the volume it occupies. This property depends on the consolidation state of the granular material, and hence aerated and tapped densities of a powder have very different values. If a powder is gently poured into a container, it will have a loose arrangement. A

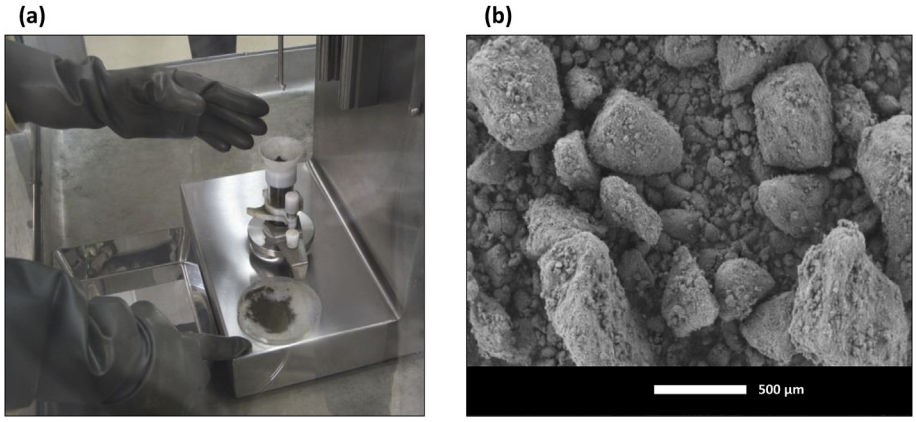


Fig. 1. Powder handling in a glove box (a) and SEM image (SE Mode) of the uranium dioxide powder (b).

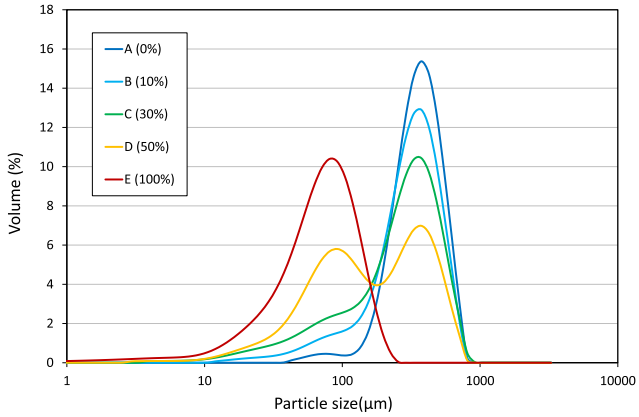


Fig. 2. Particle size distributions (volume fraction).

powder will settle under the effect of numerous vertical shocks, thereby changing the organization of the initial powder until a compact reorganization is obtained. A powder undergoes rearrangement when the interstitial air is removed. A volumenometer (STAV 2003 Stampf Volumeter) allows this transition from an aerated density ρ_0 to a packed density ρ_t following the AFNOR NF ISO 9161 protocol. For the tapping test, a 50 mL tube containing the powder was used and the device frequency was set to 245 strokes/min.

The Carr index [13–15] can be calculated via measurements of the aerated and tapped densities, as follows:

$$Carr\ Index = 100 \times \frac{\rho_t - \rho_0}{\rho_t} \quad (3)$$

Depending on the Carr index, the powder flowability can be predicted qualitatively (Table 2) [13].

Table 2
Flowability indicated by the Carr index.

Flowability	Carr index (%)
Excellent	<15
Correct	15–25
Poor	>25

2.3. FT4 rheometer

Freeman Technology's FT4 powder rheometer uses several characterization tests to analyze the flowability of powders [16].

2.3.1. Dynamic test (Variable Flow Rate, VFR test)

The principle of the test is to introduce a powder sample into a cylindrical cell in which a specific blade rotates while moving vertically along a helical path. The blade can move at different speeds. In this study, we used a 10 mL cell and a blade with a diameter of 25 mm.

Different indices were determined to evaluate the flow properties of the powder [9]. For each test, 11 measurements were made:

- A series of 7 tests was repeated at the same blade rotational speed (−100 mm/s). The value of the energy obtained during the last test was used to determine the energy necessary to move a given volume of powder, called the Basic Flow Energy (BFE). The Normalized Basic Flow Energy (NBFE) was also used to compare powders [9], to take into account the different sample masses:

$$NBFE = \frac{BFE}{Sample\ mass} \quad (4)$$

- The results obtained also made it possible to define a Stability Index, noted SI. This index represents the ratio of the total energy during the last test to the total energy during the first test.

Table 1
Fines contents and characteristic diameters.

Powder batch	Fines content (%wt)	Dv (10) (µm)	Dv (50) (µm)	Dv (90) (µm)	Span
A	0	210	360	570	1
B	10	120	310	540	1.5
C	30	60	280	530	1.5
D	50	40	160	480	3
E	100	20	70	140	1.5

- Another series of 4 tests at different blade rotational speeds (−100, −70, −40 and −10 mm/s) was performed. The blade rotational speed was reduced by a factor of 10 to determine the impact of the flow speed on the total energy. A Flow Rate Index, noted FRI, quantifies this effect by comparing the values obtained for the maximum and minimum speeds (ratio of the total energy at −10 mm/s to the total energy at −100 mm/s).
- Finally, the Specific Energy (SE) corresponds to the energy required to displace the conditioned powder during upwards testing (rise of the blade through the powder bed). This parameter characterizes the powder flow properties in an unconfined state.

2.3.2. FT4 compressibility test

With respect to the FT4 device [9], compressibility refers to the ability to reduce the volume of a bulk powder when it is slowly compressed by a piston under a given normal force:

$$\text{Compressibility} = \frac{\text{Volume variation}}{\text{Initial volume}} = \frac{\Delta V}{V_0} \quad (5)$$

In this work, the powder was poured into a cylindrical cell with a diameter of 25 mm and a volume of 10 mL. A porous compression piston of 23.5 mm diameter was applied onto the powder. The tests were carried out at normal stresses ranging from 1 kPa to 15 kPa.

2.3.3. Permeability

By measuring the permeability, it is possible to evaluate the capacity of a powder bed to let air flow through it, at different consolidation stresses and for a constant flow rate. For this test, we used a 25 mm diameter container with a volume of 10 mL, coupled to a porous aeration base connected to an aeration module. The powder was first conditioned, then a normal stress varying between 1 and 15 kPa was applied on the top of the powder bed. A constant air flow rate of 2 mm/s was introduced through the powder bed continuously. The pressure drop in the powder bed was then measured as a function of the applied normal stress.

The permeability of the bed was calculated as:

$$k = \frac{q\,\mu\,L}{\Delta P} \quad (6)$$

where k is the permeability (m^2), q the superficial velocity (m/s), μ the air viscosity (Pa.s), L the length of the powder bed (m), and ΔP the pressure drop across the powder bed (Pa).

2.4. Shear cell

Jenike [17] developed a method based on measuring the shear strength of the material. The shear cell measures the variation in shear stress required to set the consolidated powder bed into motion at different normal stresses. Bell [18] confirmed the validity and reproducibility of this method for different powders.

During a shear test, two stages can be distinguished: the pre-shearing step followed by the shearing step. The pre-shearing step is necessary before each shear measurement. This step makes it possible to repeat the measurements under the same conditions and to erase powder history. During the pre-shearing step, the powder is first compressed (normal stress σ_p) at a given value (σ_p = 3, 6, 9, or 15 kPa in the present work), and it is pre-sheared under this same normal stress until reaching steady-state flow (constant shear stress and bulk density).

During the shearing step, a normal stress σ lower than the normal pre-shear stress (σ_p) is applied, then the shear stress τ is increased until the powder bed breaks. Several couples (τ, σ) are obtained, in order to plot the yield locus (Fig. 3a).

The yield locus was assumed to be linear, according to the Mohr-Coulomb failure criterion [19]:

$$\tau = \mu\sigma + c \quad (7)$$

Thus, the following parameters could be determined [8,20]:

- The major principal stress (MPS) σ_1 obtained by drawing the large Mohr circle: it characterizes the maximum stress applied to the powder;
- The unconfined yield strength (UYS) σ_c obtained by drawing the small Mohr circle: it characterizes the resistance of the powder subjected to a simple compression;
- The slope μ is the internal coefficient of friction: $\mu = \tan(\phi)$. ϕ is called the internal friction angle;
- At $\sigma = 0$, the cohesion C of the powder was obtained. This point corresponds to the initial cohesion of the powder without any constraint being applied on it.

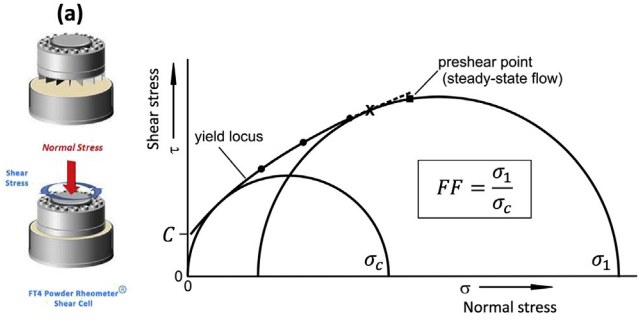
The flow factor FF can be defined as:

$$FF = \frac{\sigma_1}{\sigma_c} \quad (8)$$

This allows the flow behavior of the powder to be assessed (Fig. 3b).

After having measured several yield loci (i.e., at different pre-shear stresses σ_p), several couples (σ_c, σ_1) were obtained. The curve passing through the different couples represents the flow function. According to Jenike [17], the flow properties of a powder can be determined by its flow function.

The FT4 rheometer can be equipped with a shear cell with different volumes [16]. A 10 mL shear cell was used for our study. This cell size was found to be a good compromise between the



(b)

Flow Factor FF	Flow behaviour
FF < 1	Not flowing
1 < FF < 2	Very cohesive
2 < FF < 4	Cohesive
4 < FF < 10	Easy-flowing
10 < FF	Free-flowing

Fig. 3. Shear test and yield locus (a); classification with the flow factor FF (b) [20].

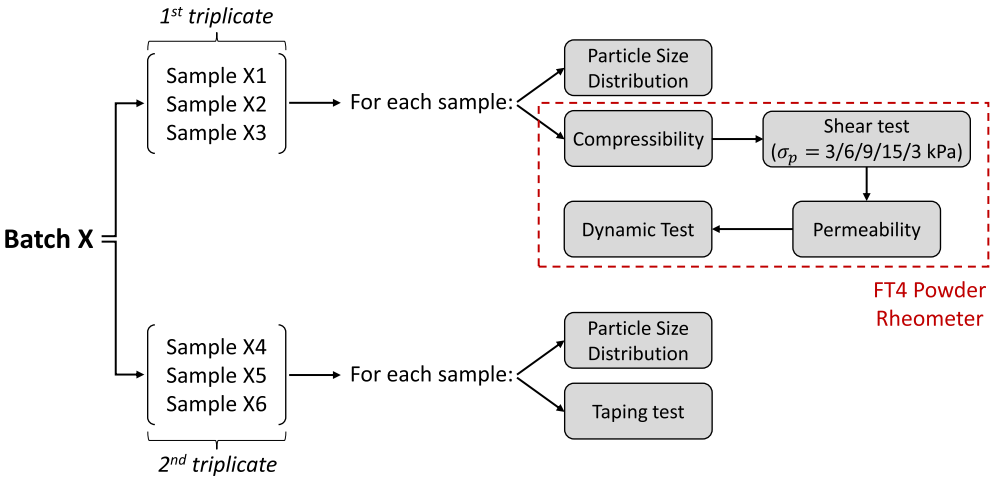


Fig. 4. Tests sequence followed in this work for the characterization of each powder batch ("Batch X" refers to Batch A, B, C, D or E).

quality of the results and the quantity of powder involved [11].

2.5. Tests sequence (order of execution)

The characterization of radioactive powders is a challenging task because the maximum information has to be obtained from the smallest possible sample size. In order to minimize the generated waste amount, the same sample may be used for successive characterization tests. The tests sequence followed in this work for the characterization of each powder batch is presented in Fig. 4. If the powder "Batch X" is considered (where "Batch X" refers to Batch A, B, C, D or E, described in Table 1):

- First, the powder Batch X was divided into six samples (noted X1, X2, X3, X4, X5 and X6);
- A small part of each sample was used to measure its particle size distribution, using a laser granulometer. The average particle size distribution obtained for the powder Batch X is presented in Fig. 2;
- Samples X1, X2 and X3 were submitted to four successive characterization tests, all executed with the FT4 powder rheometer: compressibility test, shear test (with σ_p = 3, 6, 9 15 and 3 kPa), permeability test and dynamic test;
- Samples X4, X5 and X6 were submitted to the tapping test.

3. Results

3.1. Evaluation of the powder transition from static to dynamic state (incipient flow conditions)

3.1.1. Tapping test and compressibility test

The tapping test is used to understand the rearrangement of the particles by mechanically tapping a graduated cylinder containing the sample (without applying any compressive stress, via a piston, on the sample). For the compressibility test, however, the rearrangement of grains is achieved by applying a normal stress. These two tests do not provide a direct measurement of the flow properties, but allow a qualitative prediction of the powder flowability. The results are given in Table 3 (for reference, the true density of UO_2 is 10.96 g/cm^3).

The FT4 can also be used to measure the aerated density (called Conditioned Bulk Density, CBD). In general, aerated density measurements are strongly affected by the initial powder filling operation carried out by the operator. However, as shown in Table 3, similar aerated density values were obtained with both methods. This confirms that the operator effect may be low.

In the context of this paper, these compressibility measurements are also used to evaluate if the powder density is sensitive to the fines content (this should be avoided to ensure a robust mold filling operation, i.e., a constant filling weight).

Fig. 5a shows that the powder with 100% fines has the highest compressibility (FT4) at all the applied stresses compared with other powders. The higher the amount of fines, the greater the compressibility. It can be assumed that more air is trapped in the powder bed at higher fines contents and therefore that the powder has a higher compressibility.

The Carr Index (tapping test) follows the same tendency as the FT4 compressibility. The flowability qualitatively estimated from the Carr index seems to decrease when the fines content increases. However, whereas a linear correlation can be observed between the compressibility and the quantity of fines (Fig. 5b), we observe a fines threshold above which the powder behavior changes very little for the Carr index.

For each powder, the compressibility test was carried out three

Table 3
Density measurements and compressibility (FT4).

Volumenometer test					FT4		
Powder batch	Ratio of fine particles (wt%)	Aerated density (g/cm^3) *[+/−0.3]*	Tapped density (g/cm^3) *[±0.3]*	Carr's Compressibility Index (%)	CBD (g/cm^3) *[+/−0.2]*	Density at 15 kPa (g/cm^3) *[+/−0.2]*	Compressibility at 15 kPa (%) *[+/−0.8]*
A	0	2.8	3.3	15 *[+/−3]*	2.7	2.9	5.6
B	10	2.7	3.3	18 *[+/−3]*	2.8	2.9	7.0
C	30	2.5	3.3	24 *[+/−5]*	2.4	2.6	9.8
D	50	2.5	3.4	26 *[+/−6]*	2.3	2.6	15.0
E	100	2.1	2.9	28 *[+/−7]*	2.1	2.5	21.3

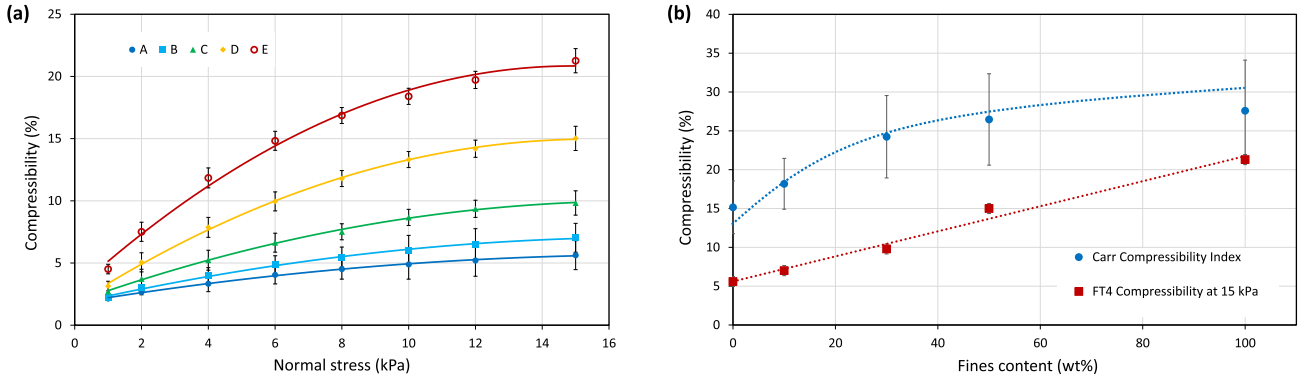


Fig. 5. FT4 compressibility as a function of the normal stress (a); Carr compressibility index and FT4 compressibility at 15 kPa as a function of the fines content (b).

times in succession on the same sample, in order to evaluate if the physical characteristics of the sample were modified during the test (agglomerates rupture, attrition, etc.). The results were similar for each test, which shows that the compressibility test can be considered as a non-destructive test. Moreover, the particle size distribution was measurement before and after each compressibility test. Again, the obtained results were similar, which showed that the granules had a sufficient mechanical strength and are not friable under a 15 kPa normal load. Consequently, the evolution of the compressibility is mainly caused by a forced rearrangement of the granules by occupying the intergranular spaces.

Finally, these results show that for fines contents below 10 %wt, the powder density is almost insensitive to the amount of fines, whereas for higher contents the density slightly decreases.

3.1.2. Shear test

Shear tests are used to characterize the complex behavior of the powder during its transition from a static state to a dynamic state (i.e., incipient flow conditions), e.g., when opening the bottom of a hopper [21].

3.1.2.1. Flow function. For each sample (Fig. 4), the shear test was performed with four different pre-shear stress levels: 3, 6, 9 and 15 kPa [22]. The same sample was used for all the shear tests, in order to use the smallest possible amount of UO_2 powder and to minimize the wastes generated by the flowability test methods. The shear tests were therefore performed in order of increasing pre-shear stresses. To verify that the powder was not modified by the test, a last 3 kPa pre-shear test was performed and its results were compared with those of the first 3 kPa pre-shear test. Preliminary experiments showed that the powder sample remained unchanged by the shearing tests.

The shear test results were used to plot the flow function for each powder (Fig. 6) [8]. Powder batches A and B, consisting of 0 and 10% fines respectively, are seen to have better flow properties than the other batches. The flow factor (FF) cannot be differentiated between powders with fines content greater than 30% (Fig. 7a). Because of their size, fine particles have a larger specific surface area than larger particles. This may explain why they dominate the behavior of the mixture from a certain threshold of fine particles. This could also explain the similarities between powder batches C (30%), D (50%) and E (100%).

3.1.2.2. Cohesion. Cohesion was also measured during the shear test. The cohesion of the powder is plotted as a function of the quantity of fines below (Fig. 7b). The cohesion clearly increases

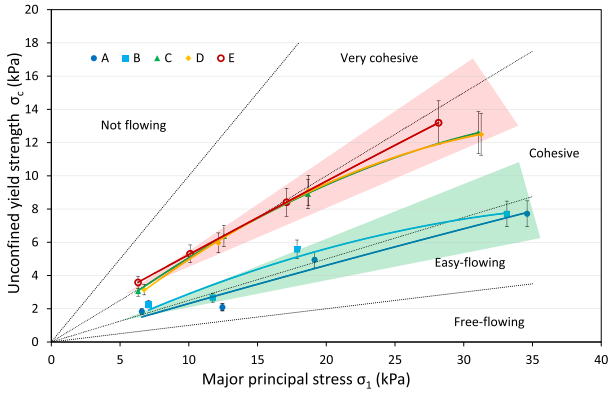


Fig. 6. Flow functions for different fines contents (A: 0 wt% of fines, B: 10 wt%, C: 30 wt %, D: 50 wt%, E: 100 wt%).

with the fines content: the powder with 100% fines (batch E) is cohesive compared with the powder with no fines (batch A).

The cohesion values are almost equivalent for the batches with 0 and 10% of fines. It is higher for 30% of fines and remains around the same value above 30% and up to 100% of fines. This means that the cohesion criterion cannot differentiate between powders with a ratio of fines higher than 30%.

3.2. Evaluation of the powder flowability in a dynamic state

3.2.1. Permeability test

In the industrial process, the air initially trapped in the molds must be removed when filling them with the UO_2 powder. The permeability test is used to characterize the filling ability because it indicates how easily the air trapped in the mold can pass through the powder bed to ensure the regular flow of the powder and therefore a reproducible filling ratio. The permeability k (equation (6)) is plotted as a function of the consolidation stress applied on the powder bed in Fig. 8a.

The pressure drop is close to a linear function of the consolidation stress for each powder. It is higher for powders with a high amount of fines. The fines cause an increase in the pressure drop because they reduce the interstitial spaces between particles. As a result, the pressure drop in powder bed E (100%wt fines) was the highest.

The permeability of the powder can then be determined as a function of the fines content. As seen previously, the permeability is

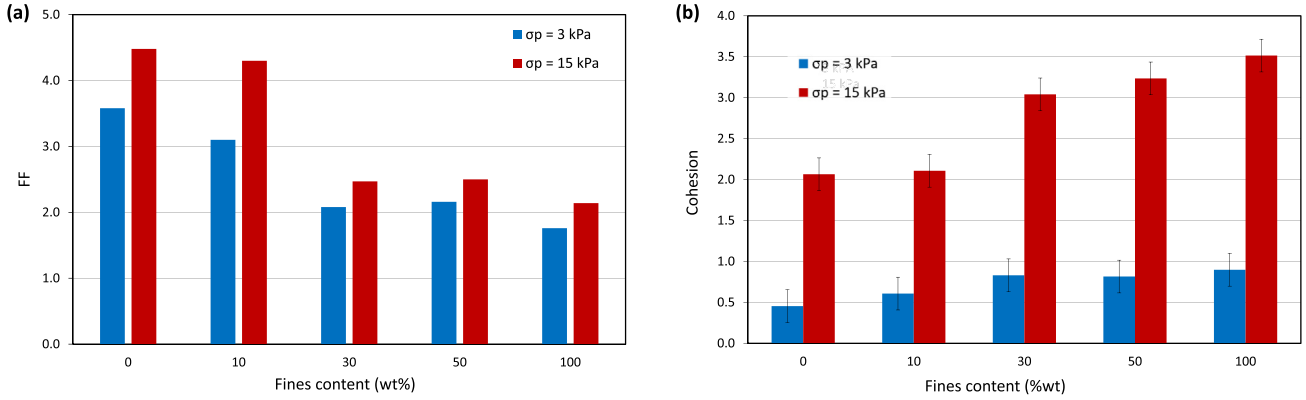


Fig. 7. Flow factor (a) and cohesion (b) as a function of the fines content (at σ_p = 3 kPa and 15 kPa).

inversely proportional to the pressure drop. As the quantity of fines increases, the pressure drop increases and the permeability *k* decreases (Fig. 8b).

The permeability is linked to the volume and structure of the inter-particulate porosity. When the quantity of fines is zero (batch A), a high level of permeability is observed. As soon as the fines are added to the mixture, they occupy the inter-particle spaces: the air passes through the powder bed with difficulty, which explains a sharp drop in permeability [16]. For this reason, the fines have a high impact on the powder flow during mold filling and therefore on the ability to obtain a reproducible and stable filling process. Unlike the shearing test, the permeability test was able to differentiate between all our powders, even those with a ratio of fines greater than 30%. The fines may play an essential role in the filling process according to the criterion of air removal.

3.2.2. FT4 dynamic test

FT4 dynamic tests were carried out on the five powders to give a better understanding of their behavior during transfer and filling of press molds. The results of the FT4 dynamic test (Variable Flow Rate, VFR test) and the characteristic indices of the different mixtures are shown in Fig. 9.

From the results of the permeability and shear tests, it can already be supposed that increasing the fines content causes an increase of the powder cohesion. The Basic Flow Energy, BFE, corresponds to the flow energy of test n°7 (Figs. 9 and 10). The measured values for powders A and B (0 and 10 wt% of fines) are higher than for powders C, D and E100 (30, 50 and 100 wt% fines).

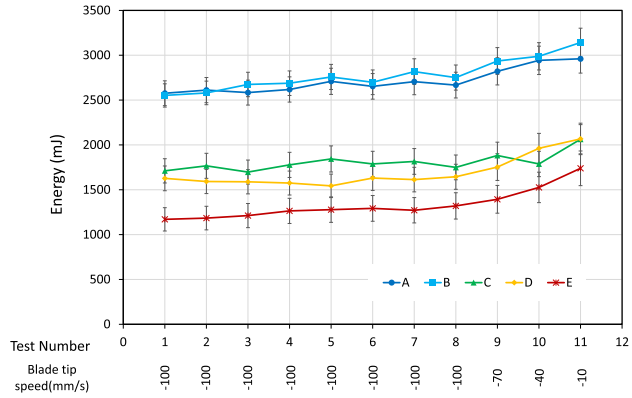


Fig. 9. FT4 dynamic tests as a function of the fines content.

Two domains can be distinguished: below 10 wt% fines and aver 30 wt% fines.

The results presented in Fig. 9 seem to indicate that the higher the amount of fines, the lower the energy measured by the blade. An interpretation of this energy reduction is due to a compressibility effect. In fact, when the amount of fines increases, the compressibility increases, as shown in Fig. 5a and b. Therefore, when the blade passes through the powder bed, the powder is compacted and the flowing zone around the blade is relatively localized, resulting in a low flow energy [23].

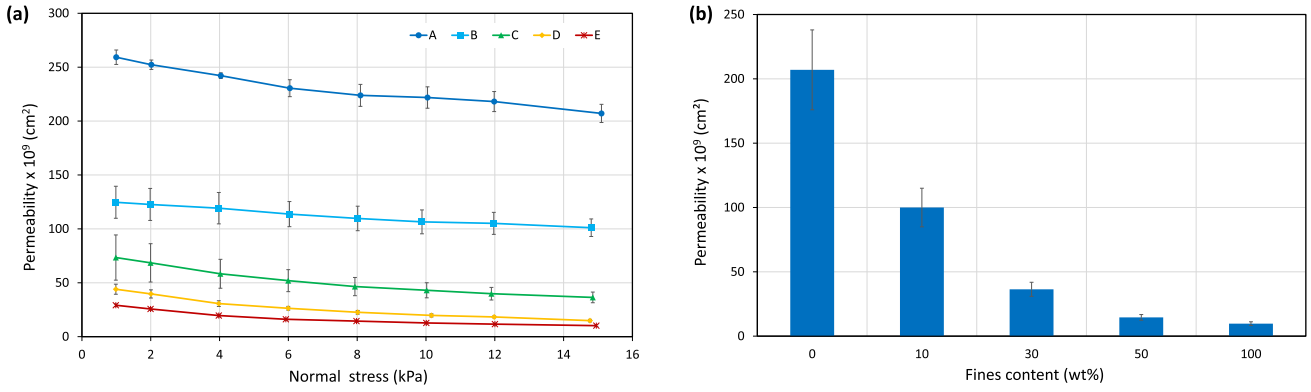


Fig. 8. Permeability as a function of the normal stress (a) and permeability at normal stress = 15 kPa (b) for different fines contents.

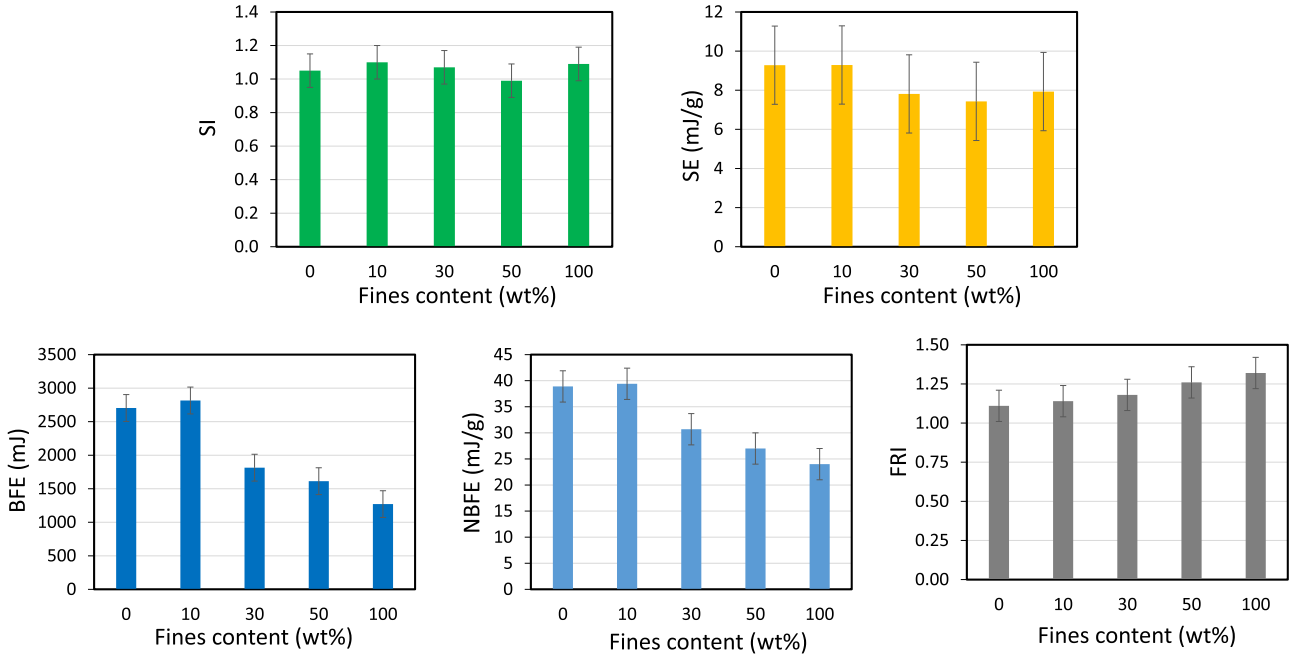


Fig. 10. Different indices (FT4 dynamic test) as a function of the fines content (BFE, NBFE, FRI, SI and SE).

The specific energy (SE) characterizes the cohesion of the powder during the rise of the blade, which is characteristic of an unconfined flow. The results presented in Fig. 10 show that the SE is not very sensitive to the amount of fines.

The FRI represents the ratio between flow energies at different rotational speeds of the blade. The FRI increases slightly with the amount of fines (Fig. 10), this phenomenon is due to the cohesion of the powder. When the amount of fines increases, the higher cohesion of the powder induces an increase of flow energy of test n°11 (with a low rotational speed of −10 mm s^{-1}) compared to the flow energy of test n°8 (at −100 mm. s^{-1}). These results are in agreement with those found in the literature: when the amount of fines increases, the powder cohesiveness increases and the flow becomes difficult.

Regarding the Stability Index (SI) (Fig. 10), this parameter is identical for the different powder mixtures, with a value close to 1. The SI value does not depend on the quantity of fines introduced into the mixture, but reflects a good stability under the dynamic test conditions.

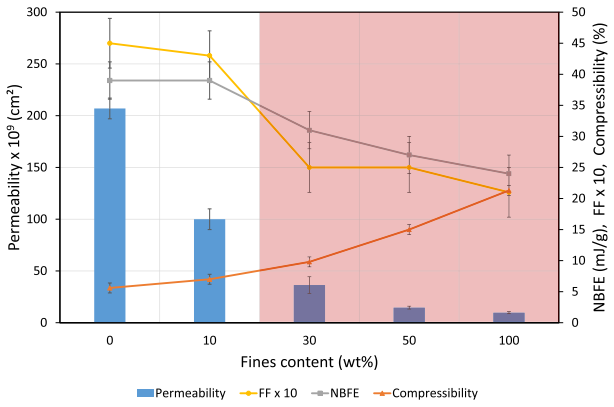


Fig. 11. Permeability (air flow = 2 mm/s, σ_p 15 kPa), compressibility, NBFE and flow factor as a function of the fines content.

4. Discussion

From the industrial point of view, the main goal of the tableting operation is to produce fuel pellets having a constant weight and a homogenous microstructure, maintaining the desired production rate. At first, this requires a reproducible, homogenous and fast filling of press molds. The powder must then satisfy the following criteria:

- Its bulk density should not be sensitive to the fines content in order to obtain a constant filling weight;
- Compatible flow properties to ensure a reproducible, homogenous and fast filling.

Table 3 (Aerated density, Tapped density, CBD) shows that for fines contents below 10 %wt, the powder density is almost insensitive to the amount of fines, whereas for higher contents the density slightly decreases. These results indicate that the amount of fines should be kept below 10 %wt to ensure a robust mold filling operation (i.e., a constant filling weight).

The flow factor (FF) and compressibility tests are representative of the powder transition from a static to a dynamic state (i.e., incipient flow conditions), while the permeability and FT4 dynamic tests reflect the powder flowability during the mold filling operation (dynamic conditions). We set out to identify different zones according to these two states: the flow initialization (transition from a static to a dynamic state) and the actual flow (dynamic state).

The FF, the permeability, the compressibility and the NBFE (Normalized Basic Flow Energy) are plotted as a function of the ratio of fines in Fig. 11. The NBFE measurement can be interpreted as a compression and shearing motion of the powder (while the blade is descending through the powder bed). According to the compressibility test, the increase of the fines content gives a very compressible and cohesive powder. Powders with large particles

are less compressible than fines because less air is trapped in the powder bed. During the FT4 dynamic test, this results in a high transmissibility of forces from particle to particle, which leads to a large flowing zone around the blade. As a consequence, a high proportion of the sample volume is required to move, and a high Basic Flow Energy (BFE) is observed for powders consisting of large particles. When the amount of fines is high, the blade passes through a more cohesive and compressible powder. Therefore, when the blade passes through the powder bed, the powder is compacted and the flowing zone around the blade is relatively localized, resulting in a low flow energy.

The permeability decreases significantly after the introduction of an amount of fines equivalent to 10 wt%. Permeability is related to the volume and structure of the interparticle porosity. The fines fill the interparticle spaces, which leads to the formation of a compact medium preventing air from passing easily through the powder bed. For this reason, the fines have a significant impact on the flow of the powder during mold filling, and thus on the possibility to obtain a reproducible and stable filling operation.

Fig. 11 provides a comparison of the different static and dynamic tests. Two behaviors can be identified:

- Cohesive behavior, with FF < 4 and $k < 100 \times 10^{-9}$ cm^2 (with air flow of 2 mm/s): for batches C, D and E (the mass fraction of fines is greater than 30 wt%). This corresponds to the very cohesive powder region. The powder does not allow air to pass through it and is therefore trapped. The powder is very cohesive and flows with difficulty. As the air is easily trapped in the powder, it therefore has a high compressibility (inducing a high FRI value and a low NBFE value).
- Non-cohesive behavior, with FF > 4 and $k > 100 \times 10^{-9}$ cm^2 (with air flow of 2 mm/s): for batches A and B (0 and 10 wt% of fines), the compressibility varies little, there is a small increase in cohesion (between 0.5 and 2 kPa) and the powder bed is highly porous. The powder demonstrates a better flow behavior than powders in the cohesive zones. This zone is in favor of a reproducible and homogeneous filling of press molds [24].

Contrary to the shear test, the permeability test made it possible to differentiate all the powders, even those having a fines content greater than 30 wt%. The behavior of the mixture appears conditioned by the amount of fine particles. Below 10 wt% fines, the behavior of the powder is governed by the large particles. Beyond 30 wt% fines, the fine particles fill the voids between large particles, which reduces the number of direct contacts between large particles and causes the formation of a cohesive powder.

Again, these results indicate that the amount of fines should be kept below 10 %wt to ensure a proper flowability, which leads to a reproducible, homogenous and fast filling of press molds.

Finally, it should also be observed, when using different methods of rheological characterization, that some tests are more sensitive to the increase of the fines content than others. It is therefore necessary when studying the rheological properties of a powder to take into consideration a type of test representative of the process conditions.

5. Conclusion and perspectives

In this work, the influence of the fines content on the powder bulk density and flow properties was studied. The objective was to identify the acceptable range of fines content to obtain a reproducible, homogenous and fast filling of press molds.

Various methods were used to characterize the flowability of different powder mixtures (i.e., different fines contents). The characterization was performed at two levels:

- During the powder transition from a static to a dynamic state: density, compressibility and shear measurements gave a better understanding of the behavior of the powders at the initialization of the flow. Shear tests demonstrated an increase in cohesion with an increase of the fines content in the mixture. This is not surprising because cohesion forces are known to overcome gravitational forces for particles smaller than 100 μm.
- Behavior of the powder in a dynamic state: this characterization was required to assess the mold filling capacity of the powder. The permeability and dynamic tests enabled us to observe the influence of fines on the powder capacity to let air flow through it, as well as to determine the energy required to displace the powder. A powder with a high fines content tends to trap air easily, which results in a poor filling behavior. When the amount of fines is increased, the flow deteriorates and the press mold filling operation can become heterogeneous and non-reproducible.

The results showed the amount of fines should be kept below 10 %wt to ensure a robust mold filling operation (i.e., constant mass and production rate).

Also, it proved complex to establish direct links between the two powder states (static to dynamic vs. dynamic). The combination of these two types of tests often produced contradictory results. To predict the powder filling behavior, the powders needed to be characterized in both states.

Finally, a series of filling tests will be performed to evaluate the “filling ability” of these powders, and thus to identify the maximum permissible fines content to ensure a reproducible press mold filling operation at the industrial level. Also, pressing and sintering tests in representative conditions are planned in order to link the flow properties of the powder, the measured weight variations and the need for rectification.

Declaration of competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

References

[1] Commissariat à lénergie atomique et aux énergies alternatives (CEA), Sodium-cooled nuclear reactors, Editions Le Moniteur, France, 2016.
[2] X. Fu, D. Huck, L. Makein, B. Armstrong, U. Willen, T. Freeman, Effect of particle shape and size on flow properties of lactose powders, Particuology 10 (2012) 203–208.
[3] Q. Li, V. Rudolph, B. Weigl, A. Earl, Interparticle van der Waals force in powder flowability and compactibility, Int. J. Pharm. 280 (2004) 77–93.
[4] E.C. Abdullah, D. Geldart, The use of bulk density measurements as flowability indicators, Powder Technol. 102 (1999) 151–165.
[5] D. Geldart, E. Abdullah, A. Verlinden, Characterisation of dry powders, Powder Technol. 190 (2009) 70–74.
[6] K. Traina, R. Cloots, S. Bontempi, G. Lumay, N. Vandewalle, F. Boschini, Flow abilities of powders and granular materials evidenced from dynamical tap density measurement, Powder Technol. 235 (2013) 842–852.
[7] Z. Chik, L.E. Vallejo, Characterization of the angle of repose of binary granular materials, Can. Geotech. J. 42 (2005) 683–692.
[8] D. Schulze, Powders and Bulk Solids: Behavior, Characterization, Storage and Flow, Springer, 2008.
[9] M. Leturia, M. Benali, S. Lagarde, I. Ronga, K. Saleh, Characterization of flow properties of cohesive powders: a comparative study of traditional and new testing methods, Powder Technol. 253 (2014) 406–423.
[10] M. Krantz, H. Zhang, J. Zhu, Characterization of powder flow: static and dynamic testing, Powder Technol. 194 (2009) 239–245.
[11] A. Madian, C. Ablitzer, M. Leturia, P. Matheron, K. Saleh, G. Bernard-Granger, Etude des propriétés rhéologiques de poudres d'oxyde d'uranium, in: 16ème Congrès de la Société Française de Génie des Procédés, Nancy, 2017.
[12] A. Madian, Étude de l'in<abstractstart>fluence des caractéristiques physiques des poudres UO2 sur leurs propriétés rhéologiques (Ph.D thesis), Université de Technologie de Compiègne, 2019.
[13] R.L. Carr Jr., Evaluating flow properties of solids, Chem. Eng. 72 (1965)

163–168.
[14] H.H. Hausner, Friction Conditions in a Mass of Metal Powder, Polytechnic Inst. of Brooklyn. Univ. of California, Los Angeles, 1967.
[15] A. Saker, M.-G. Cares-Pacheco, P. Marchal, V. Falk, Powders flowability assessment in granular compaction: what about the consistency of Hausner ratio? Powder Technol. 354 (2019) 52–63.
[16] R. Freeman, Measuring the flow properties of consolidated, conditioned and aerated powders—a comparative study using a powder rheometer and a rotational shear cell, Powder Technol. 174 (2007) 25–33.
[17] A.W. Jenike, Storage and Flow of Solids, Bulletin 123, Engineering Experiment Station, University of Utah, 1964.
[18] T.A. Bell, Powder handling-industrial needs in solids flow for the 21st Century, Powder Handl. Process. 11 (1999) 9–12.
[19] J. Schwedes, D. Schulze, Measurement of flow properties of bulk solids, Powder Technol. 61 (1990) 59–68.
[20] K. Saleh, P. Guigon, Mise en œuvre des poudres - Stockage et écoulement des silos, Techniques de l'ingénieur, 2012, p. J2255.
[21] W.-H. Wei, L.-Z. Wang, T. Chen, X.-M. Duan, W. Li, Study on the flow properties of Ti-6Al-4V powders prepared by radio-frequency plasma spheroidization, Adv. Powder Technol. 28 (2017) 2431–2437.
[22] A. D7891-15, Standard Test Method for Shear Testing of Powders Using the Freeman Technology FT4 Powder Rheometer Shear Cell, ASTM International, 2015.
[23] M. Li, M. Leturia, K. Saleh, Analysis of the periodic motion in a powder rheometer and development of a new flowability testing method, KONA Powder Part. J. 35 (2018) 160–171.
[24] R. Freeman, X. Fu, Characterisation of powder bulk, dynamic flow and shear properties in relation to die filling, Powder Metall. 51 (2008) 196–201.