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Compaction of a bed of fragmentable particles and associated acoustic emission

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Abstract :

The nuclear fuel of light water power reactors are manufactured by powder metallurgy. This is also the method that is used for the production of fuels containing minor actinides that have high activity and long life. Given their radiotoxicity, it is necessary to simplify the manufacturing process to the maximum, limiting dissemination and retention of matter. In addition, the fuel must have a mostly open porosity. Implementation of particles of a few hundred microns and controlled cohesion could meet this dual objective. However, it should be ensured that the mechanical strength of compacts before sintering is sufficient without adding binder. Thus, the phenomena that occur during the manufacture of compact are analyzed and quantified. It is shown that only a portion of the particles breaks upon application of a stress up to 600 MPa and it is possible to detect this fragmentation by acoustic emission.

Résumé :

Les combustibles nucléaires des réacteurs électrogènes à eau légère sont fabriqués par métallurgie des poudres. C'est également le procédé qui est retenu pour la fabrication des combustibles contenant des actinides mineurs à haute activité et à vie longue. Compte tenu de la radiotoxicité de ces derniers, il convient de simplifier au maximum le procédé de fabrication en limitant la dissémination et la rétention de matière. Par ailleurs, le combustible devra présenter une porosité majoritairement ouverte. La mise en œuvre de particules de l'ordre de quelques centaines de microns et de cohésion maîtrisée pourrait répondre à ce double objectif. Toutefois, il convient de s'assurer que la tenue mécanique des compacts avant frittage soit suffisante sans ajout de liant. Ainsi, les phénomènes qui se produisent lors de la mise en forme des compacts sont analysés et quantifiés. On montre que seule une partie des particules se fragmente lors de l'application d'une contrainte pouvant atteindre 600MPa et qu'il est possible de détecter cette fragmentation par émission acoustique.

Key words : granular material, compaction, UO₂, porosity, acoustic emission

1 Introduction

The current nuclear fuel manufacture implements a powder metallurgy process that comprises three main steps: preparation of powders, their compaction and sintering of the compact. It is also the reference process for the production of fuels containing minor actinides with high activity and long life, intended to be burnt in the fourth generation reactors. However, given the radiotoxicity of these fuels, they can be manufactured only in shielded cells. It is therefore necessary to simplify the manufacturing process as possible, by limiting dissemination and retention of nuclear matter. The technique that controls the process should be easy to implement and robust in a hostile and hardly reachable environment. In addition, to facilitate the release of helium during irradiation, a solution is to make a fuel having porosity essentially open, after sintering.

Thus, instead of micronic powders currently used for the production of fuels, the use of particles of a few hundred microns, graded in size, shape and cohesion, should help to limit the dissemination and retention of the nuclear matter. Such particles also facilitate the filling of press die. Nevertheless, the implementation of large particles without addition of organic binder can lead to compact that does not allow mechanical handling in industrial manufacturing process. An optimum between size, shape and cohesion of the particles

must be sought to obtain a compact with sufficient mechanical strength, while respecting the specifications of the sintered product. It is therefore appropriate to identify and quantify the mechanisms that occur during compaction according to the characteristics of compacted particles. More specifically, we are interested in highlighting the mechanism of particle fragmentation. We present the evolution of the compactness of a particle bed, depending on the applied stress. We then observe the microstructure of compacts with different compactness values. Lastly, we analyze the acoustic emission produced by the fragmentation of a single particle and that produced by a particle bed during compaction. This technique already used to monitor the compaction of pharmaceutical powders, has the advantage of being simple to be nuclear-oriented.

2 Material and experimental setup

Studies ^[1] to obtain calibrated particles directly during the manufacture of actinides oxides are in progress. However, the particles that are implemented here are obtained by mechanical granulation of UO_2 powder. They are obtained by compaction of a powder at 600 MPa; the elementary particles are submicron. The compacts are then crushed and size sorting is performed to retain only particles between 160 and 500 microns. These particles are then called granules. The density of the compact, determined by weighing and measurement, is 6.45 g/cm^3 , that corresponds to a compactness of 59%. The granules also have this density. They have a polyhedral shape as shown in FIG. 1a. Given the method for obtaining granules, some of them may have dimension greater than 500 microns. Their observation at higher magnification allows visualization of the powder particles constituting the granules (FIG. 1b and FIG. 1c). Links that bind these particles are Van der Waals attractions, electrostatic forces and capillary forces ^[2]. The dendritic shape of the powder particles also contributes to the cohesion of the granules ^[3].

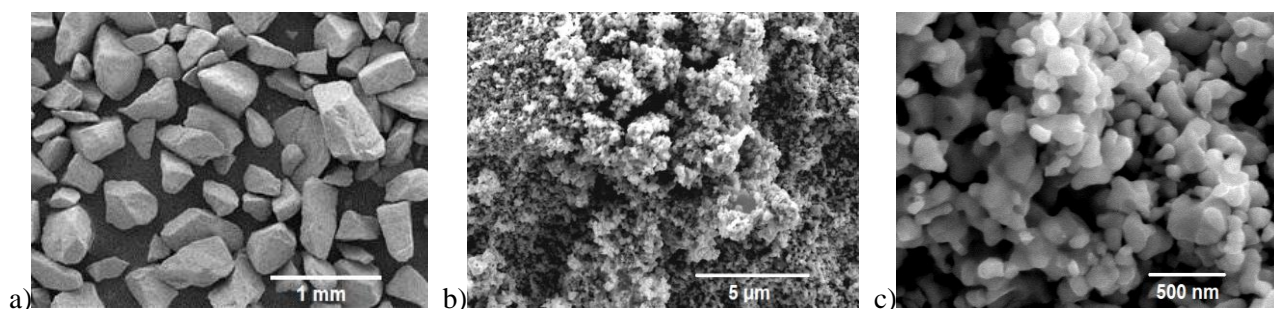


FIG. 1 - SEM observations of UO_2 granules (600MPa, 160-500 μm) at various magnifications

The granules are spilled into the press die and are compacted between two punches of 10 mm diameter (FIG. 2). The upper punch is movable and the lower punch is fixed. The die is mobile that allows ejection of the compact. Compaction is carried out at a speed of 0.1 mm/s for the movement of the upper punch until the desired applied stress. Then this pressure is maintained for 15 seconds before being reduced. During the ejection, a pressure of approximately ten times lower than the maximum applied stress is maintained on the compact to control the release of stored elastic energy during compaction and to avoid cracking or delamination of the compact ^{[4],[5]}. Force sensors (in blue in FIG. 2) are arranged directly on the punches and in the die. They record the force applied on the upper punch, the force transmitted to the lower punch and the radial force applied by the powder on the die. These three forces are used to calculate the friction coefficient between the granules and the die, and the ability of the granules to convert an axial force into a radial force. The average stress viewed by the compact is equal to the geometric mean of the applied stress and the transmitted stress ^[6]. Knowing the strengths, the position of the upper punch at any time and the compliance of the press, the variation in height of the compact as a function of stress can be calculated.

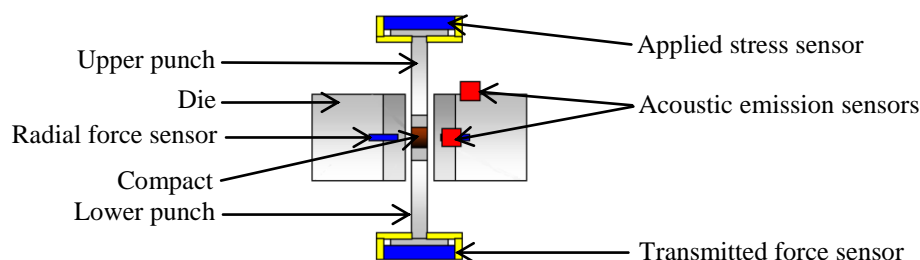
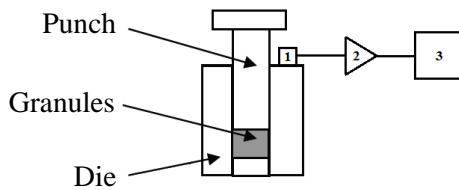


FIG. 2 - Diagram of the compression system

In addition, the die is equipped with two piezoelectric sensors (in red on FIG. 2). They record the acoustic emission during compaction using a device developed by Mistras company (FIG. 3). Sensors have a bandwidth between 100 kHz and 1 MHz. To enhance the signal transmission between the die and the sensor, they are fixed to the die by means of a spring that ensures a constant holding force. Silicon grease is used as a couplant. Before each test series, we test the quality of the sensor mounting by recording the acoustic emission produced by a pencil lead break as described in the norm “NF EN 1330-9”.



1. Acoustic sensor
2. Preamplifier (gain 40 dB)
3. Scanning system

FIG. 3 - Diagram of acoustic emission line

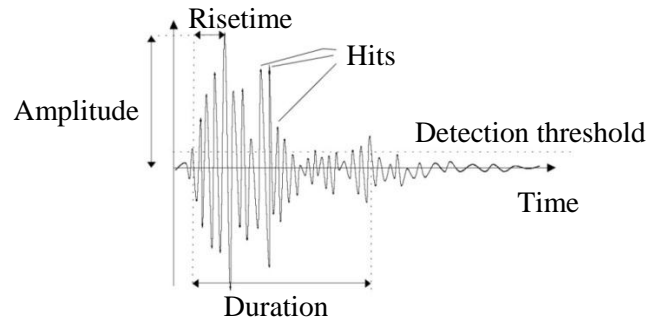


FIG. 4 - Burst signal parameters

FIG. 4 shows a typical burst signal of acoustic emission, and some associated parameters. The straight forward parameter is the number of hits, ie the number of pulses that exceed the detection threshold. However, it is not possible to associate the number of hits to a particular phenomenon because of the diversity of emission origins in the compact (friction, fragmentation) and possible spurious noise (background noise, electromagnetic radiation, mechanical vibrations related to the machine). However, an acoustic emission caused by a given mechanism will lead to a typical burst signal shape.

3 Results

3.1 Evolution of the porosity during compaction

Knowing the mass of granules introduced into the die, it is possible to continuously monitor the density of compact of UO_2 granules as a function of applied stress. The density after filling of the die is 3.0 g/cm^3 , that corresponds to a compactness of 47% of the stack of granules. The applied stress (FIG. 5) varies between 0 and 1 MPa (interval corresponding to the measuring accuracy of force) as the density is less than 3.9 g/cm^3 (60% compactness).

It is noted that the stress rapidly increases beyond about 40 MPa, that corresponds to a density in the die of 5.7 g/cm^3 . From 40 MPa, the difference between the applied and transmitted stresses also becomes significant. The density monotonically increases with stress. It is not possible to discern from this curve change in compaction mechanism. After ejection, the density of the compact compressed to 600 MPa is 6.80 g/cm^3 ; the rebound occurring during ejection is of the order of 8%. Moreover, we find that this density is greater than the measured density for the powder compacted to the same stress (6.45 g/cm^3).

Observation of a ceramographic section of compacts performed at different applied stresses allows to visualize the evolution of the microstructure (FIG. 7). After the application of a stress of 5 MPa, there are cracked granules and large porosity between the granules. Fragments of cracked granules will facilitate the rearrangement of granules. It seems that some granules are very fragmented, while others are almost not fragmented. When the stress increases from 5 MPa to a value between 100 and 300 MPa, fragmentation increases. However between 300 and 600 MPa, the size of the granules does not vary greatly. The difference in granular appearance on the picture of compacts formed at 5, 20 and 60 MPa and those formed at 100, 300 and 600 MPa is due to sample preparation. The former compacts are embedded in resin under vacuum and then polished, while the latter are thermally consolidated before being polished.

Even after an applied stress of 600 MPa, there are still some spaces between the granules that are not filled. These spaces are not due to granule wrenching during preparation of the sample, but to pores existing in the compact. It can be concluded that some granules may be subjected to high isostatic loads and low shear stress. They do not fragment while the porosity between the granules is then not completely reduced.

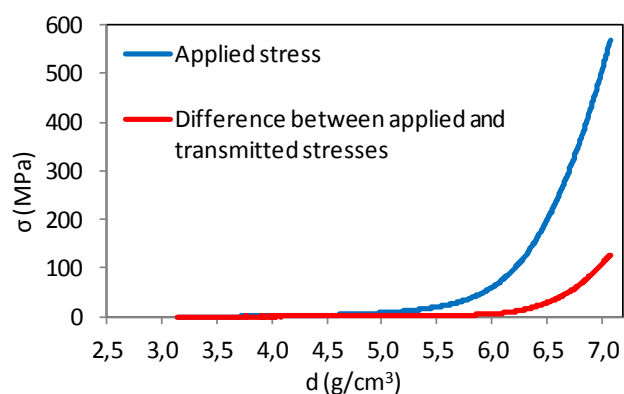


FIG. 5 – Evolution of the applied stress and the difference between the applied stress and the transmitted stress, as a function of the density

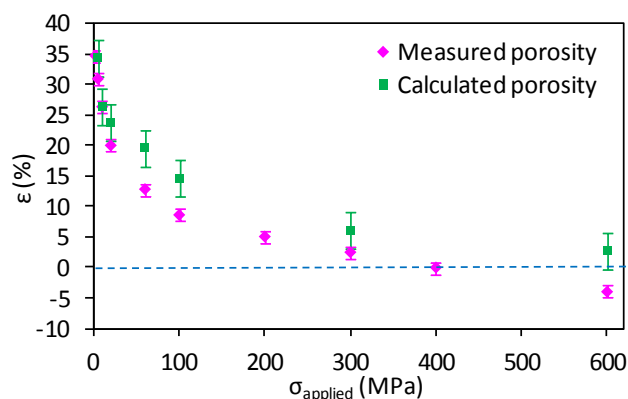


FIG. 6 – Evolution of the porosity calculated by weighing and geometric measurements and the porosity measured by image analysis, as a function of the applied stress

Quantification of granule size by image analysis is on progress. A first data analysis allowed to calculate the porosity between the granules. For low applied stresses, it can be assumed that the granules do not densify themselves. We can then calculate the porosity between the granules from the density of the compact and the density of granules (6.45 g/cm³).

As expected, porosity decreases as the stress increases (FIG. 6). For a stress of 400 MPa, the density of the compact is equal to that of the granules. The granules significantly densify on themselves at a stress of 400 MPa. Therefore, the assumption made to calculate the porosity between the granules is no longer valid. It is the reason of the negative calculated porosity between the granules of compacts made at 600 MPa. It remains true that the porosity rate thus calculated is comparable to that determined from the image analysis. For stresses below a hundred MPa, that should not lead to a significant densification of granules themselves, the observed differences may come from a too low sampling measurement by image analysis.

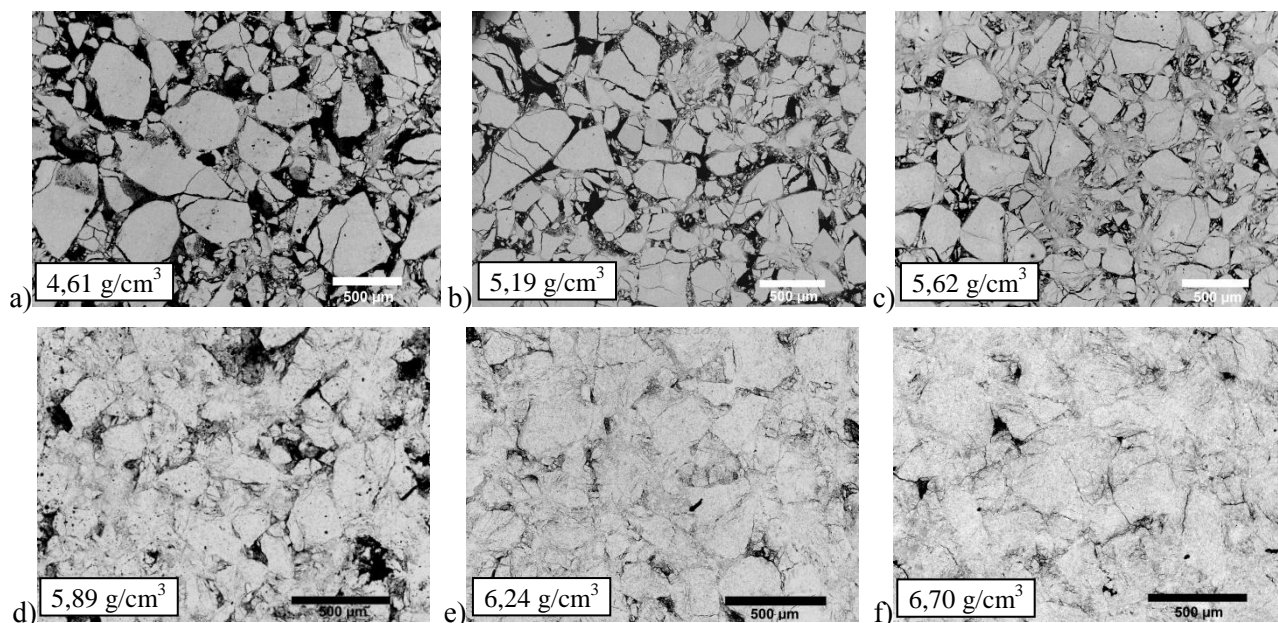


FIG. 7 - Ceramographic sections of compacts of granules performed at: a) 5MPa, b) 20MPa, c) 60MPa, d) 100MPa, e) 300MPa, f) 600MPa

Analysis by mercury intrusion porosimetry ^[7] of compacts would reveal the size of the pores. However, if this method is used to identify the stress from which the volume of pores between the granules becomes negligible, it cannot monitor/detect granules fragmentation. Only observations of the microstructure of compacts show that the granules fragmentation occurs for stresses below 300MPa.

3.2 Acoustic emission during the granules compaction

Acoustic emission is used in many processes as passive technique to monitor real-time processes that emit acoustic waves. In particular, this technique is implemented to detect and/or monitor cracks in materials. For example, G. Kerboul ^[8] followed the formation of cracks that sometimes occurs during the ejection of actinide powder compacts. In our case, the objective is to detect in situ fragmentation of granules to infer the evolution of the microstructure. Before following the acoustic emission during compaction of a bed of granules, we crushed one granule between two punches (FIG. 8) and simultaneously recorded the produced acoustic emission. The speed of movement of the punch is 500 μ m/min.

First force gradually increases, and then abruptly decreases. The granule then has a crack. The maximum force is 1.5 N \pm 0.7 N (dispersion obtained for a ten granules batch). The displacement to achieve the breaking strength is approximately 100 microns. It corresponds to the formation of flat surfaces on the granule in contact with the punches and deformation of the granule. During the increase in stress, no acoustic emission exceeds the threshold that was set at 25 dB. Upon breakage, a single event characterized by acoustic burst signal shown in FIG. 9a is detected. It has a shape similar to a graphite pencil lead break (FIG. 9b): a fast rise time followed by an exponential decay and duration of the order of 2 ms for both events. The shape of the burst recorded during the rupture of the granule is a typical characteristic of fragmentation. However, the amplitudes cannot be compared because the pencil lead break could not be performed directly on the punch holder granule (too small size punch).

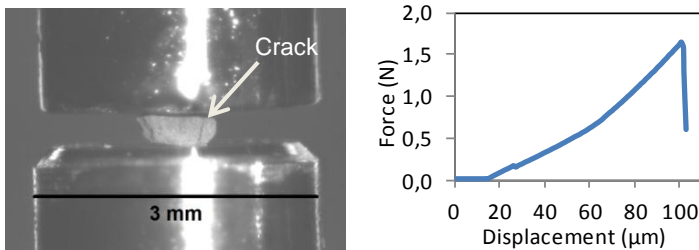


FIG. 8 – Illustration of a crushed UO₂ granule and load curve

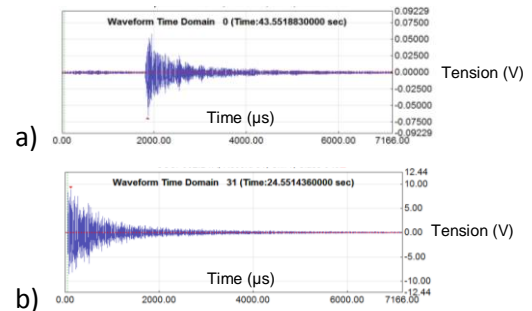


FIG. 9 – Emitted burst upon rupture of a) one granule, b) a pencil lead break

We also recorded the acoustic emission during the compaction of granules presented in § 3.1. As mentioned for the compaction of alumina powders ^[9], pharmaceutical powders ^[10] or sand ^[11], the number of hits increases as the density increases (ie when the stress increases) (FIG. 10). The number of hits increases exponentially until it reaches a plateau at a stress of about 35 MPa. When the stress exceeds 500 MPa, the number of hits increases again.

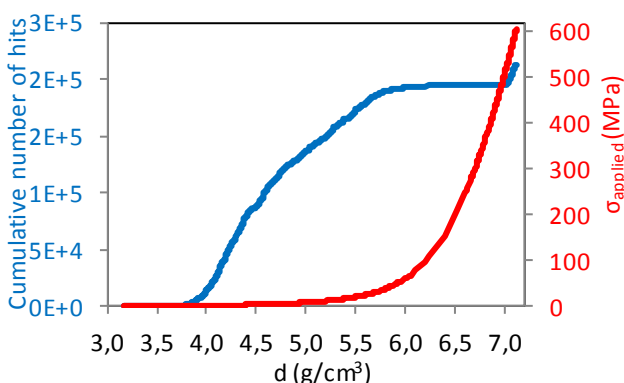


FIG. 10 – Cumulative number of hits and applied stress, as a function of compact density

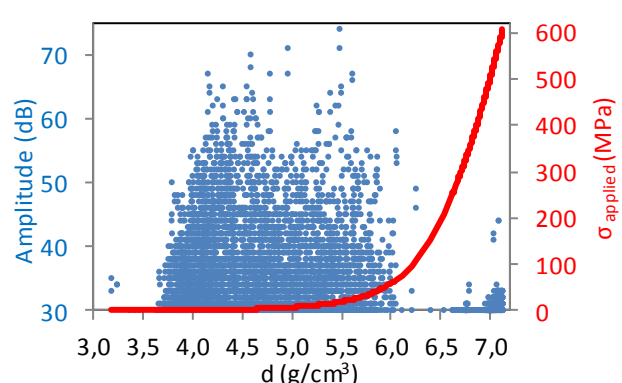


FIG. 11 - Maximum amplitude of burst signals and applied stress, as a function of the compact density

Each point plotted in FIG. 11 corresponds to the amplitude of a burst signal. The detection threshold was set at 30 dB. It is noted that burst signals of high amplitude appear as soon as a density of 3.6 g/cm³ (56%) is reached. Beyond 6.2 g/cm³, the amplitude of the burst does not exceed 30 dB. However, there is a slight increase in the amplitude of the burst when the stress exceeds 500 MPa. Between 3.6 g/cm³ and 6.2 g/cm³,

more than 80% of the burst signals, whose amplitude is greater than 35 dB, have a shape identical to that recorded in the rupture of a granule. They are characteristic of the granules fragmentation. On the other hand, the bursts observed at a stress greater than 500 MPa have a very different shape compared to those characteristic of fragmentation. The mechanism underlying these burst signals remains to be identified.

4 Conclusion

The decrease in porosity during the uniaxial compaction of a bed of granules between 160 and 500 microns obtained by compaction of a powder compact, then crushing and grain sorting, is due to rearrangements of granules and their fragmentation. Fragmentation allows further rearrangement without significant increase in the applied stress. For the high stresses, increase of compactness is mainly due to densification of the granules themselves.

The recording of the acoustic emission makes possible the in-situ determination of the beginning and the end of the granules fragmentation, based on the acoustic signature of this phenomenon. This is a promising and powerful tool for monitoring the compaction of powders through a multi-parameter analysis leading to pattern recognition. The observation of a ceramographic section of compact shows that some granules do not fragment, although the porosity between the granules is not completely filled. They are thus subject to a shear stress weaker than their cohesion.

These experimental results are in agreement with the numerical results obtained using a discrete element approach implementing dynamics of contacts ^[12]. Studies of the particle compaction having different cohesions and the influence of the characteristics of the granular packing on the compact porosity before and after sintering are in progress.

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