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Identification of the fragmentation of brittle particles during compaction process by the acoustic emission technique

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ABSTRACT

Some nuclear fuels are currently manufactured by a powder metallurgy process that consists of three main steps, namely preparation of the powders, powder compaction, and sintering of the compact. An optimum between size, shape and cohesion of the particles of the nuclear fuels must be sought in order to obtain a compact with a sufficient mechanical strength, and to facilitate the release of helium and fission gases during irradiation through pores connected to the outside of the pellet after sintering. Being simple to adapt to nuclear-oriented purposes, the Acoustic Emission (AE) technique is used to control the microstructure of the compact by monitoring the compaction of brittle Uranium Dioxide (UO_2) particles of a few hundred micrometers. The objective is to identify *in situ* the mechanisms that occur during the UO_2 compaction, and more specifically the particle fragmentation that is linked to the open porosity of the nuclear matter. Three zones of acoustic activity, strongly related to the applied stress, can be clearly defined from analysis of the continuous signals recorded during the compaction process. They correspond to particle rearrangement and/or fragmentation. The end of the noteworthy fragmentation process is clearly defined as the end of the significant process that increases the compactness of the material. Despite the fact that the wave propagation strongly evolves during the compaction process, the acoustic signature of the fragmentation of a single UO_2 particle and a bed of UO_2 particles under compaction is well identified. The waveform, with a short rise time and an exponential-like decay of the signal envelope, is the most reliable descriptor. The impact of the particle size and cohesion on the AE activity, and then on the fragmentation domain, is analyzed through the discrete AE signals. The maximum amplitude of the burst signals, as well as the mean stress corresponding to the end of the recorded AE, increase with increasing mean diameter of the particles. Moreover, the maximum burst amplitude increases with increasing particle cohesion.

1. Introduction

The nuclear fuels of light water power reactors are currently manufactured by a powder metallurgy process that consists of three main steps: preparation of the powders, powder compaction, and sintering of the compact. This process will also be used for the production of fuels containing long-lived minor actinides (such as americium) to transform them into short-lived or stable nuclides in a fast reactor. Given their radiotoxicity, these fuels need to be manufactured in hot cells. Therefore, it is necessary to simplify the manufacturing process as much as possible, thereby limiting the dissemination and retention of the nuclear matter.

In addition, in order to facilitate the release of helium and fission gases during irradiation, a majority of the pores of the fuels must be connected to the outside of the pellet after sintering. However, this open porosity must also permit mechanical handling in the industrial manufacturing process. An optimum between size, shape and cohesion of the particles of the nuclear fuels must be sought in order to obtain a compact with a sufficient mechanical strength [1–3], while respecting the specifications of the sintered product.

Using brittle particles of several hundred micrometers seems to be a good solution to obtain both compacts with open porosity and limitation of the dissemination of the nuclear matter. These particles may be obtained by Calcined Resin Microsphere Pelletization process [4]. Our work is concerned with brittle Uranium Dioxide

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(UO_2) particles, obtained by mechanical granulation of UO_2 powder, whose size ranges between 160 and 500 μm. These particles will be called "granules" hereafter.

The aim of our work is also to propose a technique that controls the process that should be easy to implement and robust in a hostile and hardly reachable environment. As the density of the medium monotonically increases with the applied stress (Fig. 1), it is impossible to highlight a change in compaction mechanism that could inform on the amount of porosity. Therefore, we propose to control the microstructure of the compact by monitoring the compaction of the brittle particles using the Acoustic Emission (AE) technique. This technique has the advantage of being simple to adapt to nuclear-oriented purposes.

The AE technique is a powerful tool dedicated to structure health monitoring (the term "structure" having to be understood here in an overall meaning). This technique has found so many applications in civil engineering [5–7], industrial pharmacy [8], geophysics [9,10], and materials science [11–16], that it is impossible here to review all the books and articles related to this topic. The AE technique is generally used to monitor real-time processes that emit acoustic waves. It is also used to detect and/or monitor defaults and cracks in materials [17–19], and to monitor the compaction of metallic [20,21], pharmaceutical [22,23] or ceramic [24] powders. Indeed, the final step of the process (*i.e.* the ejection of the compact) may create defaults inside the compact that can be detected through the comparison of the number of the acoustic events and AE energy rate between the undamaged compacts and the damaged counterparts. Some works also focus on the correlation between mechanisms during compaction (*e.g.* deformation, fragmentation, friction, etc.) and acoustic signatures [25]. Nevertheless they usually consider continuous signals and the evolution of the number of counts, rarely the discrete signals and the evolution of parameters such as amplitude, waveform, rise time, time duration, and frequency. Moreover, the AE technique is strongly dependent on the structure and the material of interest, and the analogies should be done with caution. To our knowledge, studies concerned with UO_2 are very rare and focused essentially to the detection of defaults during the material compaction using amplitude and V_{rms} analysis [24,26]. In our case, the objective is to identify *in situ* the mechanisms that occur during the UO_2 compaction, and more specifically the particle fragmentation that is linked to the open porosity of the nuclear matter, in order to infer the evolution of the material microstructure. In our work continuous as well discrete signals are considered and relevant descriptors of the fragmentation are sought. In a previous paper [27] we put in evidence the acoustic signature of fragmentation of a single UO_2 granule under compaction. Here we present some additional results and we focus more specifically on the acoustic signatures

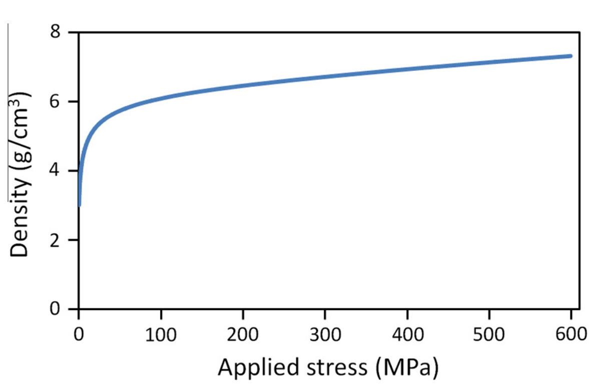

Fig. 1. Evolution of the density of the UO_2 granules as a function of the stress applied during the compaction process.

of the mechanisms occurring in a bed of granules. The main difficulty lies in the fact that the wave propagation strongly evolves during the compaction process, since the UO_2 medium is a loose granular medium at the beginning of the process and becomes a consolidated porous medium at the end.

The paper is organized as follows. Section 2 describes the properties of the UO_2 particles as well as the experimental set-up, namely the compression system coupled to the AE set-up. In Section 3 we present some new results concerning the acoustic signature of fragmentation of a single granule. Section 4 is focused on the AE during the compaction of a bed of granules. More specifically, we emphasize the impact of the granule size and cohesion on the AE activity, and then on the fragmentation domain.

2. Material and experimental setup

2.1. Uranium Dioxide (UO_2) granules

UO_2 compacts were obtained first by compaction of a powder at 600 MPa, whose elementary particles are submicron. These compacts were then crushed, and size sorting was performed to keep only particles with diameter ranging between 160 and 500 μm. These particles are called granules.

The compact density determined by weighing and measurement was 6.49 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. 2a. Their observation at higher magnification allows visualization of the elementary particles constituting the granules (Fig. 2b). Links that bind these particles are Van der Waals attractions, electrostatic forces, and capillary forces. The dendritic shape of the elementary particles also contributes to the cohesion of the granules. Some granules were thermally consolidated at 700, 1000 and 1200 °C, respectively, under argon-5% hydrogen atmosphere. At about 800 °C UO_2 starts sintering. Some solid links are thus formed between particles, which increases the mechanical strength of the granules (Fig. 3).

2.2. Compression system and acoustic emission line

The granules were poured into the press die and were compacted between two punches with a diameter of 10 mm (Fig. 4). The upper punch could move and the lower punch was fixed. The die was mobile, which allowed ejection of the compact. The upper punch moved at a speed of 0.1 mm/s until reaching the desired applied stress. During the ejection, a pressure approximately ten times lower than the maximum applied stress was maintained on the compact, in order to control the release of the stored elastic energy during compaction, and hence to avoid cracking or delamination of the compact. Force sensors (in blue[2] in Fig. 4), located directly on the punches, recorded the force applied on the upper punch and the force transmitted to the lower punch. Both forces are generally used to calculate the ability of the granules to convert an axial force into a radial force. The mean stress (σ_{mean}) viewed by the compact is equal to the geometric mean of the applied stress and the transmitted stress. Knowing the mass of granules added in the die, the position of the upper punch at any time, the strengths, and the small deformation of the press, we could calculate the variation in density of the compact as a function of stress.

Furthermore, the die was equipped with two piezoelectric sensors (in red in Fig. 4). They recorded the Acoustic Emission (AE) during compaction using a device developed by Mistras® Company. AE sensors (Mistras® μ30) of diameter 10 mm were used;

[2] For interpretation of color in Fig. 4, the reader is referred to the web version of this article.