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Plastically-driven variation of elastic stiffness in green bodies during powder compaction: Part I. Experiments and elastoplastic coupling

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ABSTRACT

Cold compaction of ceramic powders is driven by plastic strain, during which the elastic stiffness of the material progressively increases from values typical of granular matter to those representative of a fully dense solid. This increase of stiffness strongly affects the mechanical behaviour of the green body and is crucial in the modelling of forming processes for ceramics. A protocol for ultrasonic experimental investigation (via P and S waves transmission) is proposed to quantify the elastic constants (Young modulus and Poisson's ratio) as functions of the forming pressure. Experimental results performed in uniaxial strain allow for the introduction of laws that describe the variation of the elastic constants during densification. These laws are motivated in terms of elastoplastic coupling through the simulation of an isostatic pressure compaction process of alumina powder. A micromechanical explanation of the stiffening of elastic properties during densification is deferred to Part II of this study.

1. Introduction

The investigation of the mechanical properties of tablets and ceramic green bodies is important for the pharmaceutical and the ceramic industry, in the former case as dissolubility and related bioavailability of the drug is related to the density reached during pressing, while in the latter case as the control of ceramic pieces (which have to be handled without failure before firing) is crucial in enhancing the production of both traditional and structural ceramics. Therefore, the mechanical quality of the green bodies has been so far investigated as, first of all, related to density distribution and strength [3,6,16,19,27], while subsequent experimental campaigns have addressed also fracture toughness and the dependence of elastic properties on forming pressure. In particular, standard tests [10], pulse-echo [1,2] and bending resonance [18] in a long bar, and non-contact ultrasound with air [7,8,26], or water [22,23], have been employed to measure the elasticity of green bodies.

During cold mechanical densification of a ceramic granulate, the material experiences large plastic strain associated with the enlargement of the contact surfaces between grains (see Fig. 1), with a progressive gain in cohesion and elastic stiffening of the material.

In other words, compaction under pressure evolves the elasticity of a powder towards the stiffer elastic response of a fully dense material. This increase in stiffness is directly related to the development of plastic deformation, so that it can be modelled within the so-called 'elastoplastic coupling' constitutive framework, which was introduced by T. Hueckel [12–14] for rock-like materials and used to model and simulate the forming of ceramic powders [20,21,24,25]. In elastoplastic coupling, a dependence is introduced of elastic parameters on plastic deformation, so that the stiffening of the material during compaction can be modelled. However, for ceramic granulates the laws governing the dependence of elasticity on plastic strain have been until now only postulated using reasonable assumptions, but never calibrated against direct experiments. These experiments are one of the objectives addressed in the present article and were carried out by first defining and subsequently applying a testing protocol. The protocol defines a method of evaluating the elastic parameters of green bodies formed by uniaxial strain compaction through the measurement of ultrasonic P and S wave propagation speeds (Section 2).

In these experiments the Young modulus and Poisson's ratio increase after forming by 100% and 7%, respectively, while a greater variation is observed in the elastic bulk modulus K and in the Lamé coefficient λ. Three different laws are proposed to describe: (i) the compaction curves and the variations (ii) of the wave speeds and (iii) of the elastic parameters with the forming pressure. Finally, all the quantities which were found to be nonlinear functions of the forming pressure are shown to increase linearly with density when

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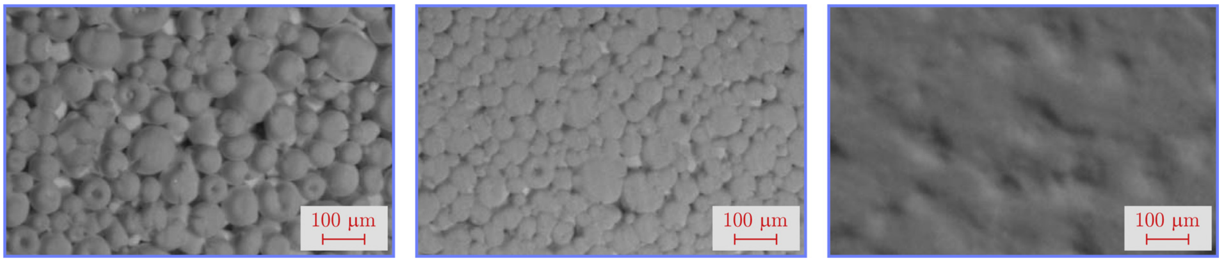

Fig. 1. During densification the contact area between the grains increases, as shown by the external surface of cylindrical samples of Martoxid KMS-96 alumina powder compacted under uniaxial strain at different pressures, from left to right: 1 MPa, 4 MPa, 100 MPa. Photos have been taken with a Nikon SMZ-800 optical microscope equipped with DSF1i camera head.

the latter increases from moderate to high values (but this linearity necessarily does not hold at low densities). Finally (Section 3) all the experimental findings are motivated in terms of elastoplastic coupling theory, through the simulation of an isostatic forming process. The development of a micromechanical model (in which the grains are idealized as elastoplastic circular cylinder or spheres) to explain the variation of the elastic stiffness in green bodies with the forming pressure is deferred to Part II of this study.

2. Ultrasound testing of elasticity during powder compaction

Two different powders have been investigated, one employed for structural and the other for traditional ceramics:

- a ready-to-press commercial grade, 96% pure, alumina powder (produced by Albermarle), namely, Martoxid KMS-96. This powder has particles of 170μm mean diameter, obtained through spray-drying, Fig. 2 (left);
- an aluminium silicate spray dried powder, labelled I14730, tested at two different water contents, namely, $w = 5.5\%$ and $w = 7.7\%$, corresponding to values used in the industrial forming of traditional ceramics. The powder has been manufactured by Sacmi S.C. (Imola, Italy) and has the granulometric properties reported in [5], Fig. 2 (right).

2.1. Experimental protocol

What follows is the experimental protocol that was defined for the measurement of the Young modulus and Poisson's ratio of the ceramic powder for different uniaxial strain values.

- 10 g of powder was weighed (using an Orma EB 200 scale, sensitivity ±0.0001 g) and was then poured into a stainless steel, cylindrical, single action die (30 mm diameter) and then settled using vibrations (for 20 s at 350 Hz). The mould was cleaned, before each compaction, with an ethanol soaked cloth.
- Various disk-shaped specimens were formed by uniaxial strain of the above powder in the mentioned mould to different loading pressures. This was achieved by imposing a constant rate displacement of 0.05 mm s^{-1} on the top of the mould, using a 100 kN electromechanical universal testing machine (Beta 100 from Messphysik Materials Testing); the applied loads were measured with a TC4 load cell from AEP transducers Italy (100 kN maximum load); the displacements of the crosshead of the testing machine was measured using a PY-2-F-010-S01M external displacement transducer from Gefran Italy; data were acquired with a NI CompactDAQ system interfaced with Labview 2013, from National Instruments.
- The thickness of each disk-shaped specimen was estimated using a Palmer caliper (from Mitutoyo, sensitivity ±0.001 mm), as the mean value of three measures taken at different points, according to the UNI EN 725-10:2008 (Advanced technical ceramics – Methods of test for ceramic powders – Part 10: Determination of compaction properties). The geometrical density of the green body was then calculated as the ratio between weight and volume of the disk-shaped specimens. At this stage, the density/pressure response of the material can be plotted (Fig. 5).
- The upper and lower surfaces of the disk-shaped specimens were covered with a layer of either pressure (Sonotech ultragel II ultrasonic couplant) or shear gel (Sonotech shear gel ultrasonic couplant), for use with pressure or shear wave transducers, respectively.
- The transmission mode technique was adopted in the experimental tests and both shear and longitudinal velocity *independent* measurements in the materials were made. An ultrasonic square wave pulser/receiver (Olympus 5077PR) unit combined with a NI PCI-5152 Digitizer/Oscilloscope was used with two normal incidence shear wave transducers (Olympus Panametrics NDT V151, frequency 0.5 MHz) and two pressure wave transducers (Olympus A102S, frequency 1 MHz).

  Two transducers were used one as transmitters and the other as receivers. The signals were measured using a NI PCI-5152 Digitizer/Oscilloscope, and a preamplifier (Olympus 5660B) was used between the receiving transducer and the receiver. In Fig. 3 the

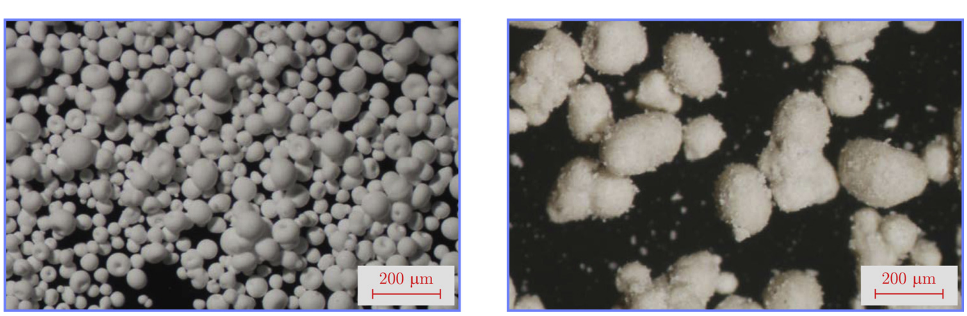

Fig. 2. Micrograph of the Martoxid KMS-96 powder (left) and of the I14730C powder at 5.5% water content (right). The photos have been taken with a Nikon SMZ-800 optical microscope equipped with DSF1i camera head. The scale bar is 200 μm.