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Experimental characterization of the behavior of granular visco-plastic and visco-elastic solids during compaction

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The microscopic behavior of viscous materials under compaction was studied, with focus on the evolution of the pore structure with increasing pressure, at different strain rates. Granular polyethylene glycol (PEG) and high density polyethylene (HDPE) were compacted with a compaction simulator up to different pressures, at two different strain rates. Compaction curves were constructed, and diametral strength tests were performed on the tablets. Scanning electron microscopy (SEM) was used to characterize the microstructure. PEG exhibited a visco-plastic behavior, as opposed to HDPE, which behaved visco-elastically. Observation of the pore structure revealed that PEG fractured and developed rate-dependent permanent deformation, resulting in good bonding and strong tablets. On the other hand, HDPE tablets contained large pores, even at high pressures, due to the considerable amount of springback after ejection, and their diametral strength was low. For PEG, the out-of-die microstructure was strongly dependent on the strain rate, while the HDPE structure was almost independent of it. In both cases, the diametral strength and the in-die density were dependent on the strain rate. However, the effect of strain rate on diametral strength was considerably more evident for PEG than for HDPE, due to their different nature and to the resulting pore structure.

1. Introduction

The powder compaction process is a common operation to several industries, including the pharmaceutical, ceramic, food, and household product manufacturing. In the pharmaceutical industry, the process acquires a special importance due to the fact that most of the medication doses -more than 80%- are administered in the form of tablets [1]. The technology available for tableting is vast and relatively simple. It provides for specific needs that range from aseptic environments for the manufacturing of low-demand products, to high production rates of over-the-counter drugs.

A wide variety of studies have been made to investigate the powder compaction process. Raw material properties and process conditions have been correlated to the bulk behavior of the powder as it undergoes compaction, and to the properties of the product tablet [2, 3]. Nevertheless, little is known about why and how such correlations are originated. The microstructure determines the properties of the tablet, and is a result of the properties of the powder and of its behavior under compaction. Studying the microstructure, therefore, provides direct evidence of the mechanisms that control the compaction process.

Observation of the internal structure of compacted granule beds has been done previously, especially in the field of ceramics. Scanning electron microscopy (SEM) is frequently used to look either at the pressed surface or the fracture surface of a compact [4–6]. Zhang *et al.* [7] and Naito *et al.* [8] used optical microscopy to study the internal structure of compacts with an immersion liquid technique proposed by Uematsu *et al.* [9, 10].

Only a few studies show a progression of the internal structure of compacts with increasing compaction force. For example, Lam and Kusakari [11] observed SEM micrographs of the fracture surface of hard and soft spray-dried granules compacted at several forces. They concluded that for hard granules, intergranular cohesion is improved by increasing the compaction force over a range of values wider than that for the case of soft granules. Uematsu *et al.* [10] used the liquid immersion technique to observe the structure of alumina compacts formed at increasing pressures.

The effect of strain rate, or compaction speed, on the properties of pharmaceutical tablets has been widely investigated. Materials that have a viscous nature respond differently when compacted at different strain rates, independently on whether they deform permanently (plastic nature) or recover after the load is removed (elastic nature). For materials that have a dominantly plastic behavior, such differences remain clearly evident even after ejection. For example, it has been reported that harder tablets are produced when the strain rate is decreased [12–14], due to the fact that the granules are given more time to develop permanent, or plastic, deformation and therefore to bond. Elastic materials, due to out-of-die recovery, may not show a pronounced microstructural difference after ejection. A detailed explanation can be derived from the observation of the microstructure of tablets in order to better understand the effect of strain rate on the compaction behavior of visco-plastic and visco-elastic materials.

This study provides insight into the microscopic behavior of viscous solids during compaction. With the use of SEM, the microstructure of the resulting tablets was studied, focusing on the evolution of the pore structure with increasing compaction pressure at different strain rates. Two different materials were used, in order to show through comparison, how the dominant deformation mechanism affects the powder's behavior during the compaction process, and the diametral strength of the tablets.

2. Experimental procedure

2.1. Materials

Two materials were analyzed in this study: polyethylene glycol (PEG) and high-density polyethylene (HDPE). Fig. 1 shows SEM micrographs of the powders as they were used in the experiments. Spray congealed PEG (Carbowax, PEG 8000, provided by Union Carbide) had a spherical, uniform shape, and a theoretical density of 1.09 g/cm^3. The powder was classified by sieving, and a range of sizes between 250 and 300 micrometers was collected and used in the experiments. The HDPE powder (Schaettifix 1820, Schaetti) had a nonuniform shape, and its theoretical density was of 0.95 g/cm^3. It was also classified by sieving and particles ranging in size from 200 to 400 micrometers were used. The theoretical densities were determined by helium pycnometry at 25°C.

2.2. Compaction experiments and characterization

The powders were compacted into tablets using a compaction simulator, the Rutgers Integrated Compaction Research System. A detailed description of the setup and operation of the simulator was reported by Celik and Marshall [15]. Single action pressing was used in a cylindrical die with a diameter of 10.3 mm. Sample weights of 0.5 g for PEG and 0.4 g for HDPE were compacted up to different pressures, ranging from 25 to 600 MPa, approximately. Two different strain rates (displacement speed of the upper punch) were used: 1 mm/s and 100 mm/s. Three repetitions were made for each pressing condition.

The force on the upper punch's contact surface and the strain displacement were collected as data from the simulator, and later used to construct compaction curves. The diametral strength of tablets compacted at different pressures and rates was measured with a tablet hardness analyzer (Vanderkamp, VK-2000; Vankel Industries, Inc.). To perform the measurements, the tablet is pressed diametrally between two solid surfaces until it fractures. The force needed to fracture the tablet is recorded as the diametral strength. SEM was used to study the microstructure evolution during the compaction process, through the observation of the pressed surface of the tablets, after ejection.

3. Results and discussion

3.1. SEM observations: effect of compaction pressure

Figs 2 and 3 show the SEM micrographs of the out-of-die pressed surface of PEG and HDPE tablets, respectively, compacted up to three different pressure levels (low, medium, and high), and at two different strain rates (1 mm/s and 100 mm/s). For PEG, the low, medium, and high pressure levels correspond to 36, 60, and 204 MPa, respectively. For HDPE, the levels are 60, 240, and 480 MPa, respectively. As expected, an increase in the compaction pressure caused the particles to get closer together, reducing the size of the pores and increasing the tablet's density. However, the different mechanical properties of the two materials triggered different densification paths, causing the pore structure to evolve uniquely in each case.

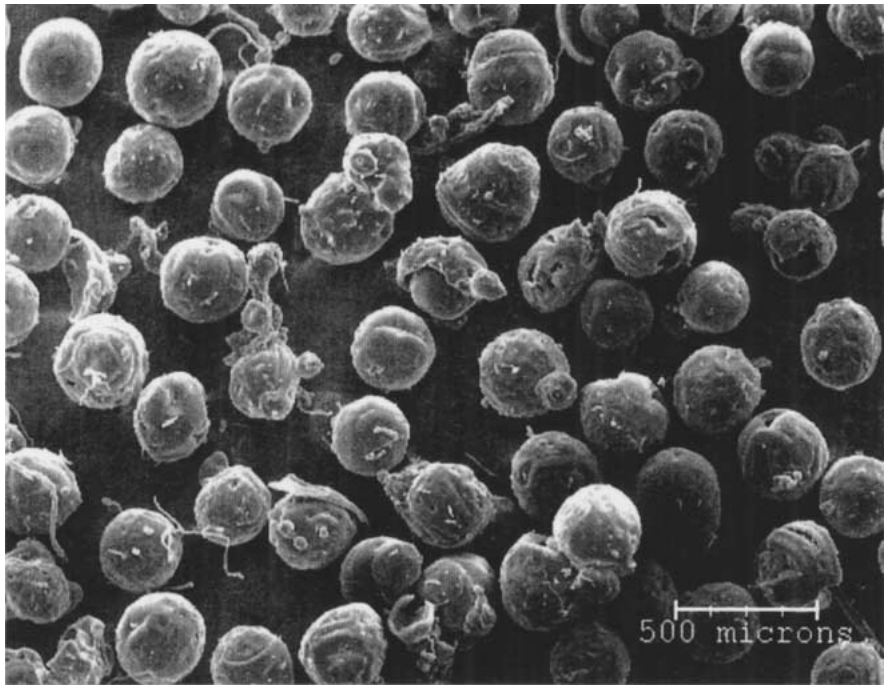
PEG showed rate-dependent permanent deformation, indicating a dominant visco-plastic behavior. It can be seen in Fig. 2 that, as the compaction pressure increased, the PEG particles fractured, and the smaller pieces rearranged, helping in the bonding process. It must be noted that the out-of-die pore structure is closed, especially at the highest pressure level, indicating good bonding as a result of permanent deformation. This is evidence that the amount of springback after ejection was small for PEG (see also Table I).

HDPE, on the other hand, behaved visco-elastically. It can be seen in Fig. 3 that the particles did not fracture; instead, they were flattened by the contact with the upper punch's surface. Although some flattening occurred, the deformation was mainly elastic because the bonding was poor and large pores remained, even at high pressures. No significant difference could be detected between the pore structure of the HDPE tablets formed at low and high pressures, indicating that the amount of springback after ejection was larger than that of PEG (see also Table I).

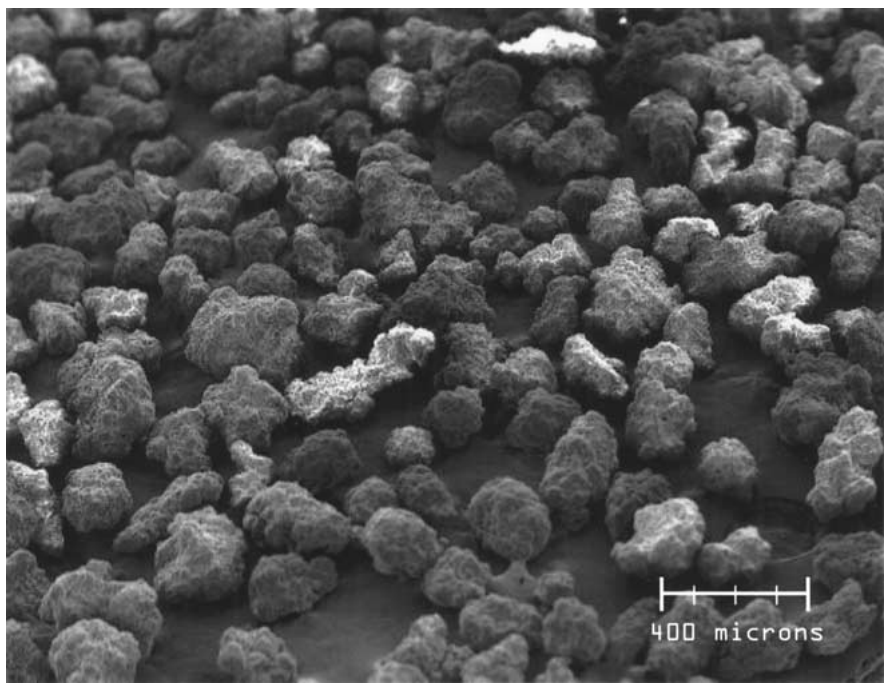
To support the above results, the exact amount of springback was calculated for both materials, as defined by

$$\text{Springback} = 1 - \frac{\text{Thickness}|^{@\ \text{max. pressure}}_{\text{in-die}}}{\text{Thickness}|_{\text{out-of-die}}}$$

Table I shows the average springback calculated from the compaction data obtained at 1 and 100 mm/s, with a maximum pressure of 500 MPa. As it was expected, the



(a)



(b)

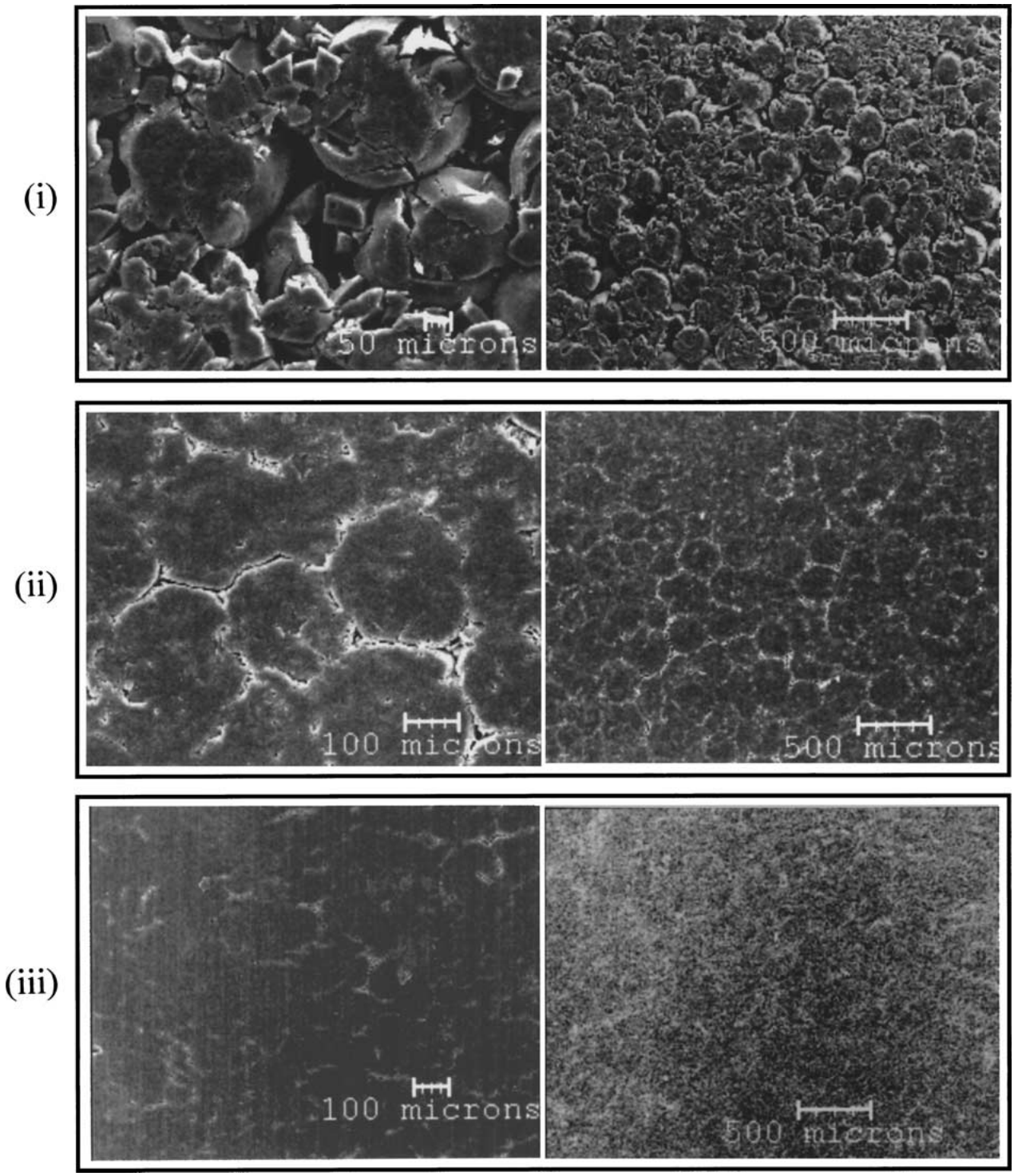
Figure 1 SEM micrographs of the powders used in the experiments: (a) PEG and (b) HDPE.

springback for HDPE, the elastic material, was larger than that of PEG.

3.2. SEM observations: effect of strain rate

For each material, the microstructure evolution responded differently to the strain rate. The out-of-die structure of PEG compacted at 1 mm/s (Fig. 2a) is noticeably more dense than when compacted at 100 mm/s (Fig. 2b). This agrees with the expected behavior according to the available literature [12–14], since a low compaction rate promotes additional densification for plastically deforming materials. Such a behavior takes place due to the longer time the material has to deform plastically. Therefore, one could expect that an elastic material would not show a pronounced difference in its out-of-die microstructure when compacted at different speeds, which was the case of HDPE. It can be seen in the SEM micrographs (Fig. 3a and b) that the structure of HDPE tablets showed no strong differences when compacted at different rates. Nevertheless, the viscous nature of both, HDPE and PEG, was reflected on the diametral strength measurements and on the in-die densification process.

Fig. 4 shows the effect of compaction strain rate on diametral strength. For both materials, the tablets were stronger when compacted at 1 mm/s than at 100 mm/s. However, such effect was stronger on PEG, the



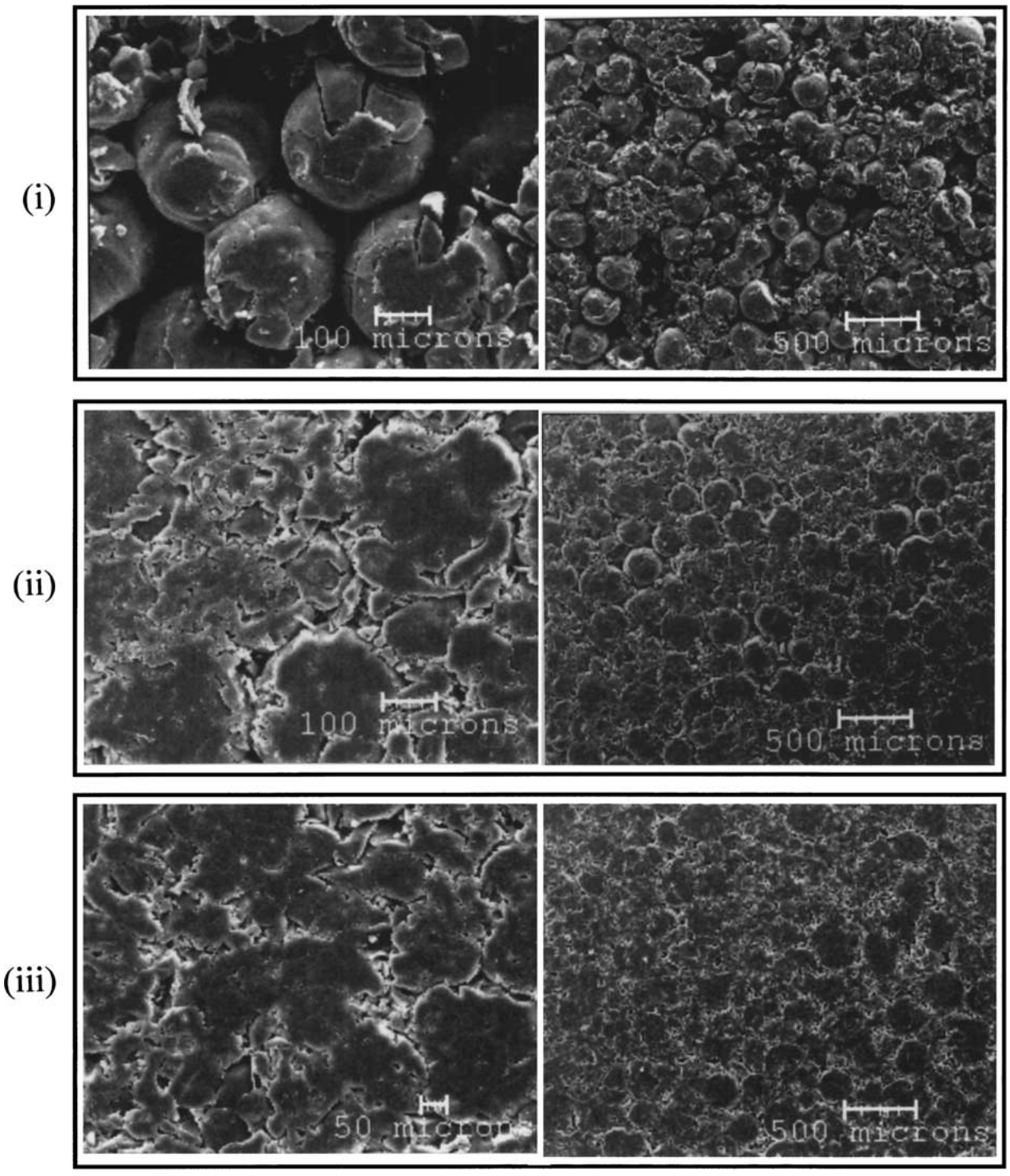
(a)

Figure 2 (a) PEG tablets' microstructure development with increasing compaction pressure, at 1 mm/s. Each set of SEM micrographs shows the upper surfaces of the ejected tablets compacted up to a specific pressure: (i) 36 MPa, (ii) 60 MPa, and (iii) 204 MPa. The micrographs on the left show higher magnifications of important details. (b) PEG tablets' microstructure development with increasing compaction pressure, at 100 mm/s. Each set of SEM micrographs shows the upper surfaces of the ejected tablets compacted up to a specific pressure: (i) 36 MPa, (ii) 60 MPa, and (iii) 204 MPa. The micrographs on the left show higher magnifications of important details. (*Continued*)

TABLE I Average springback for PEG and HDPE, calculated for a maximum pressure of 500 MPa

Material	Compaction rate (mm/s)	Average springback
PEG	1	0.0996
	100	0.1056
HDPE	1	0.2210
	100	0.2119

visco-plastic material, than it was on HDPE. The plots show that, on average, the PEG tablets compacted at 1 mm/s were almost twice as strong as those compacted at 100 mm/s, while the diametral strength values for HDPE tablets compacted at low and high rates were closer to each other. The clear difference in the PEG diametral strength measurements can be explained by looking, again, at the microstructure (Fig. 2a and b).



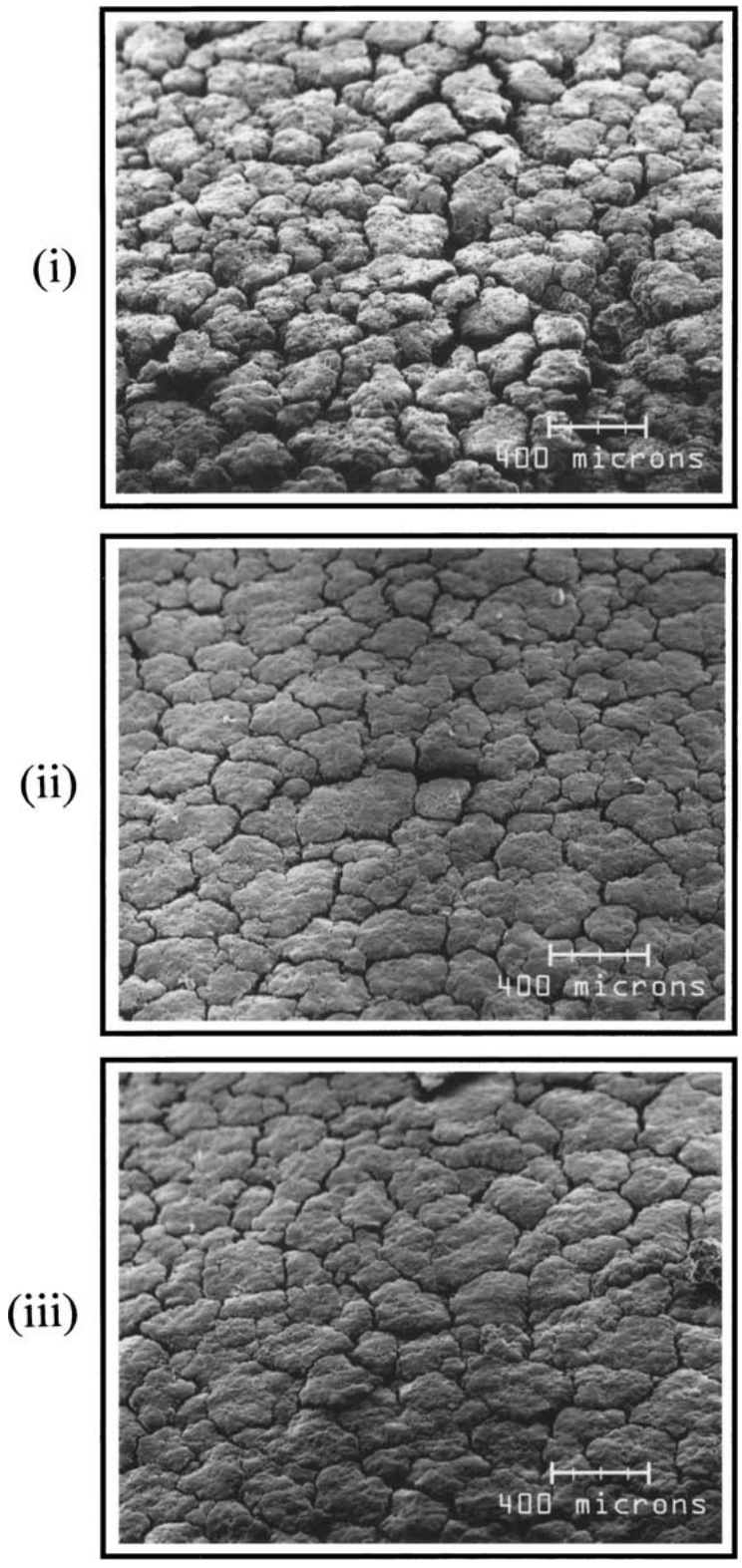
(b)

Figure 2 (*Continued*).

The pores in the tablets compacted at 1 mm/s are round, while the ones in tablets formed at a high rate are long and narrow, and resemble cracks along the particle boundaries. These longer, crack-like pores seem to have caused failure at considerably lower loads.

3.3. In-die behavior: compaction curves

A semilogarithmic plot of the density, or relative density, against the compaction pressure, as it develops throughout the process, is used to study the in-die behavior of the powder. From the resultant compaction curve (see Fig. 5), the different stages of the process can be identified and differentiated with a clarity that depends on the particular material which is under study. Stage I involves the beginning of the compaction process: as force is applied, the particles rearrange and fill in large packing voids, causing the density to increase slowly. In stage II a faster densification takes place, as the particles fracture (if the material is brittle), deform plastically and/or elastically, and bond, decreasing considerably the size of the voids. In stage III, all of the major deformation and bonding has been completed, so the density increases slowly as the few remaining

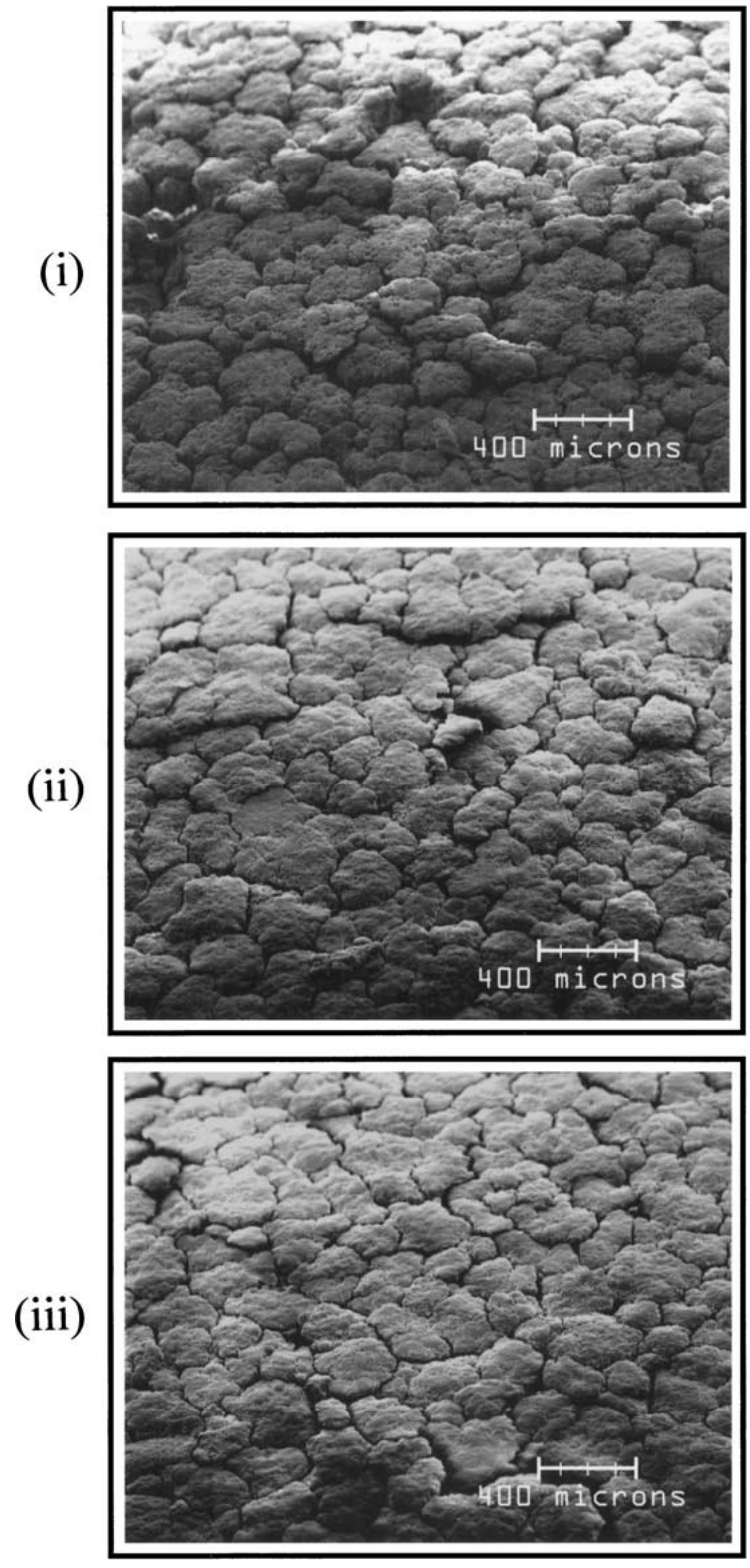


(a)

Figure 3 HDPE tablets' microstructure development with increasing compaction pressure, at different strain rates: (a) 1 mm/s and (b) 100 mm/s. Each set of SEM micrographs shows the upper surfaces of the ejected tablets compacted up to a specific pressure: (i) 60 MPa, (ii) 240 MPa, and (iii) 480 MPa. (*Continued*)

voids decrease in size. In this last stage, either plastic or elastic deformation may occur.

The in-die densification behavior of both, PEG and HDPE, showed strain rate dependence. Fig. 6 shows the compaction curves at different strain rates. The plots include the data from the three repetitions made for each condition. Stage I was omitted from the curves, since only stages II and III are the focus of the comparisons made throughout this study. For both materials, the in-die density was greater at 1 mm/s than at 100 mm/s. Clearly, this means that the different stages of the compaction process have more time to develop when subject to a lower strain rate, enhancing the densification process.



(b)

Figure 3 (*Coninued*).

For the case of HDPE, a mainly elastic material, it is important to look again at the springback results explained earlier (Table I). It is necessary to recall also that the compaction curves reflect the in-die behavior of the material, while the micrographs show the microstructure after the tablet has been ejected, and has had time to recover. In turn, and due to the springback after ejection, the micrographs of HDPE tablets show no considerable microstructural differences with respect to strain rate, even though the compaction curves do show differences. It is important to mention that the microstructure differences are not evident to the naked eye, but do have an impact on the strength of the tablets. However, such impact is relatively small, as compared to the case of PEG.

By looking at the compaction curves, it can be seen that the transition between stages II and III occurs approximately at 10 kN for PEG and 8 kN for HDPE.

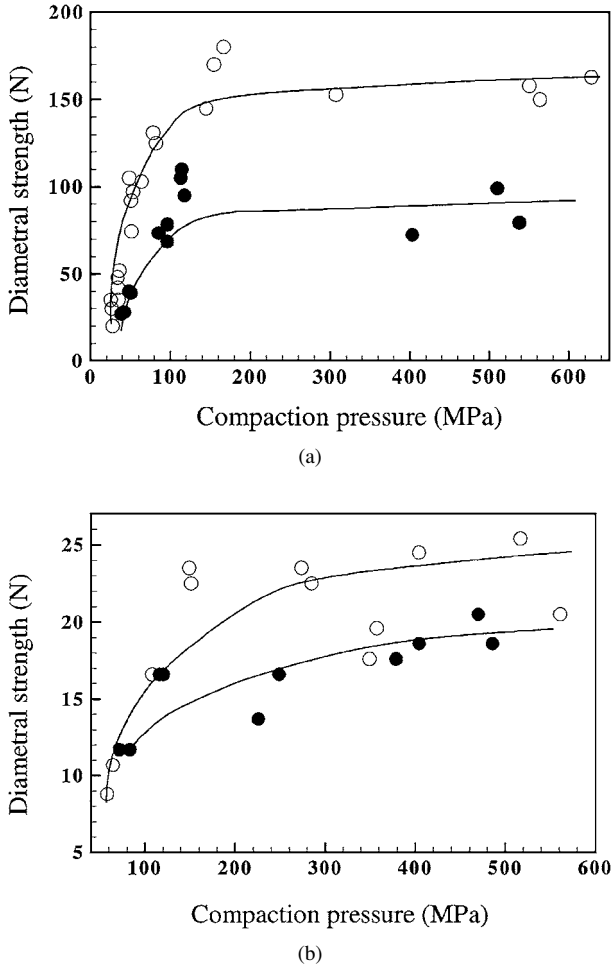


Figure 4 Diametral strength vs. compaction pressure for (a) PEG and (b) HDPE at different strain rates: (○) 1 mm/s and (●) 100 mm/s.

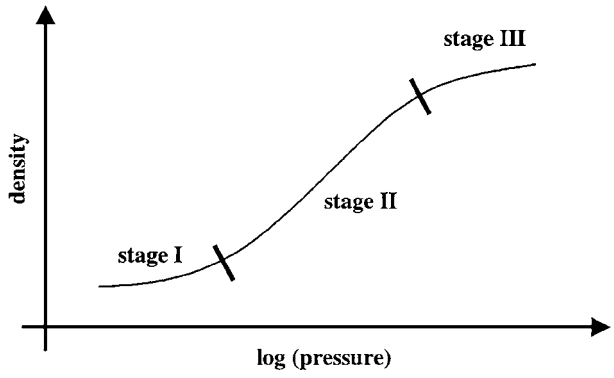


Figure 5 The compaction curve: a schematic representation of the compaction process.

In Fig. 4 it can be noted that these transition pressures also indicate the point at which the diametral strength becomes constant with respect to compaction pressure. This means that the deformation experienced by both materials during stage III was mainly elastic. Tablets pressed at the transition pressure or above recovered partially as the load was removed. So even though they were subject to higher loads, their diametral strength (out-of-die property) was not affected considerably after the stage II-III transition point.

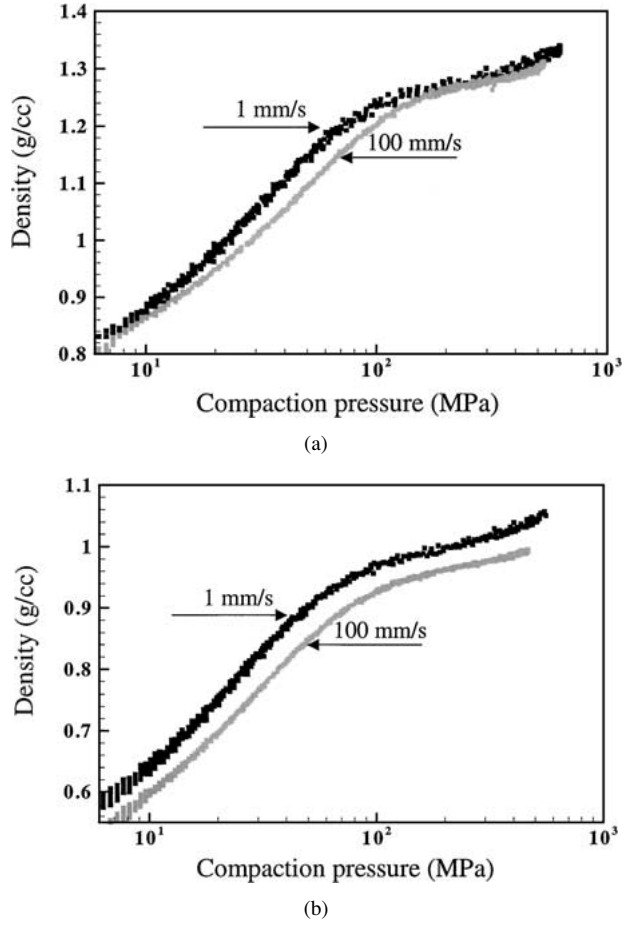


Figure 6 In-die density vs. compaction pressure for (a) PEG and (b) HDPE, at different strain rates.

4. Conclusions

The behavior of a particular powder under compaction is mainly determined by the material's mechanical properties. The densification path followed by the material as force is applied has a strong influence on the microstructure and, as a consequence, on the properties of the resulting tablet. Focusing on the out-of-die behavior of the finished tablets, the viscous nature of a material is more evident if the dominant deformation mechanism is plastic. The permanent deformation allows the differences to remain visible even after ejection. As was discussed, the visco-plastic material showed important differences in diametral strength when compacted at different strain rates. Such differences were noted in the SEM micrographs by looking at both, density and void structure. When elasticity is dominant, the springback after ejection may hide some of the changes that the powder went through during compaction. As a consequence, out-of-die properties seem similar, even when the powder was compacted at different strain rates.

In summary, PEG exhibited a visco-plastic behavior, undergoing greater densification during compaction to produce stronger tablets than the ones formed with HDPE, which behaved visco-elastically. The characterization of the pore structure through direct observation is an important tool that leads to the improvement of the powder compaction process, since it helps to understand the mechanical behavior of the materials involved.

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