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Relaxation tests for the time dependent behavior of pharmaceutical tablets: a revised interpretation

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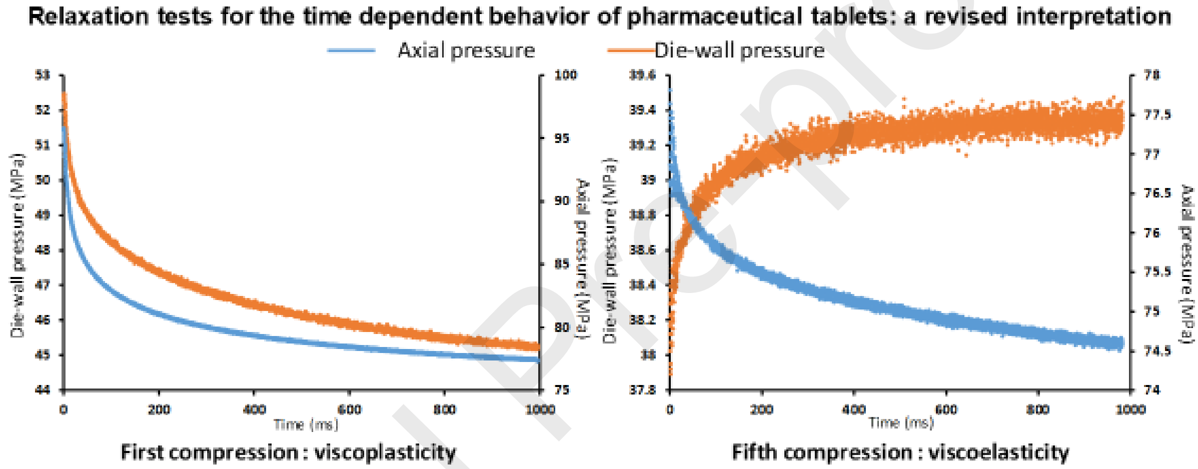

Abstract

Relaxation tests are often used in the pharmaceutical field to assess the strain rate sensitivity of pharmaceutical powders and tablets. These tests involve applying a constant strain to the powder in the die and then monitoring the stress evolution over time. Interpreting these tests is complicated because different physical phenomena, mainly viscoelasticity and viscoplasticity, occur simultaneously. These two phenomena cannot be distinguished by observing the evolution of the axial pressure alone, as it decreases in both cases. In this work, it was shown that monitoring the evolution of the die-wall pressure during relaxation can help separate the effects of these phenomena. Theoretical considerations revealed that during viscoplasticity, the die-wall pressure also decreases, whereas an increase in the die-wall pressure during relaxation indicates viscoelastic relaxation. This was confirmed experimentally using specially designed compaction cycles on four different pharmaceutical excipients. Experimental results indicated that at low pressure, viscoplasticity was predominant, whereas at high pressure, viscoelasticity became more prominent. These results suggest that at low pressures, relaxation tests can be used to assess the viscoplastic properties of different products. However, the use of high pressure should always be avoided as viscoelastic phenomena might become more significant, and the combination of both phenomena might compromise the interpretation.

Keywords: relaxation, tablet, viscoelasticity, viscoplasticity, SRS

1. Introduction

Pharmaceutical tablets are manufactured on an industrial scale using the die compression of powders. This process is also utilized in other industrial fields, such as producing ceramics or metals. One of the unique aspects of using this process in the pharmaceutical field, in addition to not being coupled with a sintering step as is generally the case for metals or ceramics, is the use of high compression speeds with punches that can travel as fast as 1 m/s. This is made possible by large rotary presses that can produce up to 1 million tablets per hour.

At such high speeds, it is not possible to neglect the influence of compaction speed, or compression kinetics, on the final properties of the tablet (Es-Saheb, 1992; Ruegger and Celik, 2000; Tye et al., 2005; Mizunaga et al., 2020). The sensitivity of a product to the compaction speed is often called strain-rate sensitivity (SRS)(Armstrong, 1989; Roberts and Rowe, 1985). During the development of a tablet, studying the SRS of a formulation can help anticipate scale-up problems, as developments are often conducted on smaller and consequently slower machines than those used in production.

Several physical phenomena can explain the SRS of a formulation. The most cited in the literature are viscoelasticity, viscoplasticity and air entrapment(Armstrong, 1989; Casahoursat et al., 1988; Çelik and Aulton, 1996; David and Augsburger, 1977; Katz and Buckner, 2013; Malamataris and Rees, 1993; Morehead, 1992; Rees and Rue, 1978; Rehula et al., 2012; Rippie and Danielson, 1981). It is complicated, from the literature, to determine which of these mechanisms is predominant during powder compression, as they are difficult to separate.

Among the methodologies proposed to study the SRS, several articles present the use of relaxation tests. A relaxation test consists in submitting a sample to a constant strain and follow the evolution of the applied stress. In the context of pharmaceutical tablets, these tests are generally performed on the tableting machine, i.e. in the die, by compressing the powder up to a certain pressure. Once the pressure is reached, the machine is stopped, and the evolution of the axial pressure is monitored(Shlanta Stephen and Milosovich George, 1964; David and Augsburger, 1977; Casahoursat et al., 1988; Rees J. E. and Rue P. J., 2011; Rehula et al., 2012; Mizunaga et al., 2021). Although all authors acknowledge that after compression is stopped, the axial pressure decreases, they do not always link this variation to the same phenomenon. For example, David et al.(David and Augsburger, 1977) claim that, using this test “Plastic flow in various materials has been evaluated”, linking the results to the viscoplastic behavior. Casahoursat et al.(Casahoursat et al., 1988) say that “the shapes of stress relaxation curves can be affected by air entrapped in the powder bed”. Finally, Rehula et al.(Rehula et al., 2012) state that “it is possible to assess viscoelastic properties of materials by means of the stress relaxation test.”. So, as it can be seen, it is difficult, from the literature, to understand exactly what is measured during a relaxation test.

To better understand the SRS, it would be of interest to determine if one of the proposed mechanisms is more predominant. As a first approach, the effect of air entrapment on relaxation tests can potentially be neglected. Indeed, recent studies on numerical simulation indicated that, even at very high speeds, the air pressure in the tablet at the compression top is around 1MPa (Klinzing and Troup, 2019), which is much lower than the actual pressure drops observed in the literature during relaxation tests. This is corroborated by another recent study that shows that the influence of air entrapment on force evolution during the loading phase is small (Vreeman and Sun, 2022).

Based on the previous discussion, the stress relaxation observed during compression should be more linked to viscoelasticity or to viscoplasticity. The aim of the present study is to try to find a way to identify if one of the two mechanisms is in fact dominant depending on the conditions used. For this purpose, we propose, in addition to monitoring the axial pressure, to study the evolution of the die-wall pressure. In the following text, we will first begin with some theoretical considerations about viscoelasticity and viscoplasticity. Experiments will be then presented to try to answer the question.

2. Theoretical considerations

As mentioned in the introduction, viscoelasticity and viscoplasticity are often conflated in the literature. For example, David et al.(David and Augsburger, 1977) clearly mention plastic flow (i.e. viscoplasticity) but use viscoelastic models to fit the data. So, we found it interesting to properly define both term and link the stress evolution in each case to the corresponding physical phenomena.

Viscoplasticity refers to plastic (irreversible) deformations that are time-dependent (in opposition to rate-independent plasticity) whereas viscoelasticity refers to elastic deformations (reversible) that are time-dependent. To establish a connection with the physical phenomena at play, we will first introduce a simple 1D scenario before generalizing it to the 3D case. This approach will enable us to investigate the evolution of the die-wall pressure during a relaxation test.

2.1. Simple 1D approach

To exemplify the difference between viscoelasticity and viscoplasticity, we will first use a simple 1D model. In all the text, compressive strains and stresses will be considered positive. Let's consider an elastoplastic solid that is submitted to a compressive strain ε. As the solid is elastoplastic, the applied strain will generate, in the solid, an elastic strain (ε^e) and a plastic strain (ε^p). It will also generate a stress (σ). If we suppose that the elastic behavior is governed by Hooke's law, with a Young's modulus E, the situation can be described by the two following equations:

$$\varepsilon = \varepsilon^e + \varepsilon^p \quad (1)$$

$$\sigma = E\varepsilon^e \quad (2)$$

This situation corresponds to the beginning of a relaxation test. Let's now consider the two cases of viscoelasticity and viscoplasticity. For the sake of the demonstration, we will consider that the strain was apply at an infinite speed (ideal relaxation test).

If the product is purely viscoelastic, it means that its Young's modulus depends on the strain rate. A classical representation can be made using Prony series (Lemaitre and Chaboche, 1990). This means that Young's modulus decreases with time. As we are performing a relaxation test, ε is constant, and as the product is not viscoplastic ε^p is also constant. As a consequence, using Eq. 1, we can deduce that ε^e is constant. Using Eq. 2, as E is decreasing and ε^e is constant, σ will decrease as a function of time, which is the expected trend.

Now if the product is viscoplastic, the plastic deformation will depend on time, meaning that ε^p will increase with time (more plastic deformation). As ε is kept constant, Eq. 1 indicates that ε^e is decreasing with time. As the product is not viscoelastic, E is constant (note that we neglect the variation of E with the density that are expected to be small). So, using Eq. 2, we can see that σ is also decreasing, as in the previous case, but for another reason.

This simple example shows why in both cases, we obtain a decrease of the applied stress. Even if the reason for the decrease is different in both cases, the result is the same and this explain why it is impossible to distinguish viscoelastic and viscoplastic phenomena using the applied stress in a 1D case.

2.2. 3D generalization

Let's now generalized the previous case in 3D in the particular case of a relaxation test performed in a cylindrical die, as done in the pharmaceutical field. For a 3D generalization, it is easier to separate the stresses and strains into volumic and deviatoric parts (Rippie and Danielson, 1981). Eq 1 can be separated into two equations, one for the volumic strain (ε_v) and one for the distorsionnal strain (ε_s) as follows:

$$\varepsilon_v = \varepsilon_v^e + \varepsilon_v^p \quad (3)$$

$$\varepsilon_s = \varepsilon_s^e + \varepsilon_s^p \quad (4)$$

The elastic equilibrium can be written using the bulk modulus (K), the shear modulus (G), the hydrostatic stress (p) and the Von Mises deviatoric stress (q) using the following equations:

$$p = K\varepsilon_v^e \quad (5)$$

$$q = 3G\varepsilon_s^e \quad (6)$$

In the particular case of a compression in a cylindrical die, p and q can be written as a function of the axial stress (σ_{ax}) and the radial stress (die-wall pressure, σ_{rad}) as:

$$p = \frac{\sigma_{ax} + 2\sigma_{rad}}{3} \qquad (7)$$

$$q = |\sigma_{ax} - \sigma_{rad}| \qquad (8)$$

In the same manner, ε_v and ε_s can be written as a function of the axial (ε_{ax}) and radial (ε_s) strains as:

$$\varepsilon_v = \varepsilon_{ax} + 2\varepsilon_{rad} \quad (9)$$

$$\varepsilon_s = \frac{2}{3}|\varepsilon_{ax} - \varepsilon_{rad}| \quad (10)$$

During loading and relaxation, the axial stress is greater than the radial stress. The absolute value can thus be omitted in Eq. 8. In the same way, during die compaction the axial strain is much higher than the radial strain so the absolute value can also be omitted in Eq 10.

Eq. 7 and 8 can be reversed to express the axial and radial stresses as a function of p and q leading to:

$$\sigma_{ax} = p + \frac{2}{3}q \quad (11)$$

$$\sigma_{rad} = p - \frac{q}{3} \quad (12)$$

As in the previous case, the idea is to understand the evolution of the stresses during a relaxation test. Both the axial and the radial stresses will be considered.

In the case of a viscoelastic behavior, as mentioned in the 1D scenario, evolution of the stress will be due to the evolution of the elastic constants, here K and G. Indeed, as the solid is not viscoplastic, there is no change in plastic strain and as the total strain is constant, the elastic strain is also constant. It is generally considered in the literature that the main part of viscoelasticity is due to the deviatoric behavior, i.e. the variation of the stresses is mainly due to variation of G (Rippie and Danielson, 1981). If we consider this limit case, i.e. G decreases but K remains constant, this means, thanks to equations 5 and 6, that during relaxation q will decrease and p will be constant. If we introduce these evolutions in Eq. 10 and 11 it means that, during the relaxation σ_{ax} will decrease and that σ_{rad} should in fact increase.

In the case of viscoplasticity, the evolution of the stress, is due to the evolution of the strains because, as the solid is not viscoelastic, the elastic moduli are constant. Due to the symmetry of the process and to the presence of the die, most of the strains are occurring in the axial direction. So, considering this hypothesis, the radial deformations can be neglected (both elastic and plastic). If so, ε_v and ε_s can be expressed directly as a function of ε_{ax} as follows:

$$\varepsilon_v \cong \varepsilon_{ax} = \varepsilon_{ax}^e + \varepsilon_{ax}^p \quad (13)$$

$$\varepsilon_s \cong \frac{2}{3}\ \varepsilon_{ax} = \frac{2}{3}\left(\varepsilon_{ax}^e + \varepsilon_{ax}^p\right) \quad (14)$$

In this case, Eq 5 and 6 can be rewritten as:

$$p = K\varepsilon_{ax}^e \quad (15)$$

$$q = 2G\varepsilon_{ax}^e \quad (16)$$

Introducing Eq. 15 and 16 in Eq.11 and 12 will then give:

$$\sigma_{ax} = \left(K + \frac{4}{3}G\right)\varepsilon_{ax}^{e} \qquad (17)$$

$$\sigma_{rad} = \left(K - \frac{2G}{3}\right)\varepsilon_{ax}^{e} \quad (18)$$

Note that the term $\left(K - \frac{2G}{3}\right)$ is equal to the first Lame coefficient, and is positive for Poisson's ratio between 0 and 0.5, which is the case for pharmaceutical powders. As explained in the 1D case, during viscoplastic flow, the plastic strain will increase and the elastic strain will decrease. Eq. 17 and 18 show that in this case, both the axial and the radial stresses should also decrease.

These theoretical considerations show that, during a relaxation experiment, the evolution of the die-wall pressure might make it possible to distinguish between viscoelastic or viscoplastic effects. Indeed, if axial pressure decreases in both cases, the limit cases studied before show that it could be possible to have an increase of die-wall pressure for viscoelasticity whereas die-wall pressure always decreases in the case of viscoplasticity. An increase of the die-wall pressure would thus indicate that viscoelasticity is the dominant behavior. It would thus now be interesting to look at the effective experimental behavior during compression.

3. Material and methods

3.1. Powders

Several classical pharmaceutical excipients were chosen for the sake of the demonstration: Microcrystalline cellulose "MCC" (Vivapur 12, JRS Pharma, Rosenberg, Bade-Wurtemberg, Germany), Starch "Sta" (Startab, Colorcon, etc.), Lactose monohydrate "Lac" (Excipress GR 150, ArmorPharma, Maen Roch, France) and mannitol "Man" (Pearlitol 200 SD, Roquette, Lastreme, France). Magnesium stearate (Ligamed MF-2-V, Peter Greven, Bad Münstereifel, Nordrhein-Westfalen, Germany) was used for internal lubrication. MCC and Sta were lubricated at 0.5% (W/W) and Lac and Man were lubricated at 1% (w/w). Lubrication was performed using a Turbula mixer (Type T2C, Willy A. Bachofen AG, Muttenz, Switzerland) for 5 minutes at 49 rpm.

3.2. Compaction experiments

All the compactions were performed using a compaction simulator Styl'One Evolution (Medelpharm, Beynost, France) which is a single station instrumented tableting machine. It is equipped with force sensors (strain gauges) on both punches (HBM 1-U93/50kN, response time 1µs) and on the die wall, and the displacements of the punches are monitored using incremental sensors.

Round flat punches with a diameter of 11.28 mm were used. The compaction cycles used were based on a saw-tooth profile with a punch speed of 20 mm/s. This profile was used to characterize the compressibility and tabletability of the different products with compaction pressure between 50 and 350 MPa. For the relaxation experiments, dwell-times were added at the compression top. All the compressions were symmetrical. For all the products, the filling weight was chosen to obtain a thickness of about 3mm at 150 MPa. The compaction event was set-up in order to have the center of the tablet in front of the die-wall pressure gauge.

Several relaxation experiments were performed using the parameters previously defined (i.e compaction speed of 20 mm/s, etc.). The first one was a multiple compression consisting of 5 successive compressions. The target thickness was set to reach 100 MPa for the first compression and kept constant for the four following compressions. For each of the 5 compressions, a relaxation time of 1 second was added at the compaction top.

Other experiments consisted of single compression with a relaxation time of 1 s at the compaction top. These experiments were performed at 50, 150 and 250 MPa.

The rational for each experimental condition will be explained in the results part below.

4. Results and discussion

4.1. Proof of concept using multiple compactions

In the theoretical part above, we showed that viscoelasticity and viscoplasticity could give different die-wall stress evolution during a relaxation experiment. The first idea was thus to see if this was indeed observed experimentally. For this purpose, an experiment was built to emphasize the different behaviors. To emphasize viscoplastic effect, it is important to perform the test in a pressure range were plastic deformation is happening to a large extent. This means that a range corresponding to overcompression (i.e. a range were an increase of pressure does not lead to a significant increase in density) should be avoided. The compressibility and tabletability profiles of the different products can be found in Figure 1. The curves are similar to what can be found elsewhere in the literature. At low pressure, the porosity decreases rapidly, meaning that a lot of plastic deformation is performed when the pressure increases. On the contrary when the pressure increases the porosity decrease less and less rapidly. The range where overcompression happened generally correspond to the range where a plateau is reached in the tabletability plots. According to Figure 1, no overcompression is seen on the studied pressure range for Lac, Man and Sta, as the tabletability profiles show a god linearity on the whole pressure range. In the case of MCC, above 200 MPa there is a clear tendency towards a plateau, even if it is not reached in the pressure range studied. Based on these results, for all the products used, a first choice of an axial pressure around 100 MPa was made, which is in a the range were plastic deformation is large. The idea was that, if plastic deformation is large, viscoplasticity should also be favored if it is present.

If, after the first compression, a second compression is performed until the same target thickness, the plastic deformation obtained during this second compression should be less, as most of it has already been performed during the first compression. And this should be particularly true if the compression is repeated several times. So during these repeated compressions, as less and less plastic deformation is occurring, viscoplasticity should be less and less and as a consequence, viscoelasticity should be favored.

To test this idea, a multiple compression cycle was used. It consisted in 5 successive compressions (which correspond to the maximum number of compressions allowed by the software used). For each compression event, a dwell-time of 1 s was added at the compression top to observe the relaxation behavior of the products. This protocol was applied to the different products and the results of the evolution of axial and radial pressures are presented in Figure 2. The axial pressure plotted represents the mean value between the pressures of the upper and lower punches.. As it can be seen, for all the products and for all the compressions, the axial pressure always drops during the relaxation time. Nevertheless, the importance of the drop was different depending on the products and on the compaction event. This is coherent with the theoretical development above. It can also be seen that the pressure drop is always more important for the first compression than for the others. This means that the time dependent phenomena are more important for the first compression. This also seems logical, as the viscoplastic phenomena should be more important in the first compression. It can also be noted that, even if the same thickness set point was given for all the compressions, the maximal axial pressure decreases from one compression to the other. This is obviously partially due to the relaxation during the dwell-time. Note that if only viscoplastic effect would take place we could expect that maximum pressure obtained during reloading to be the same as the one obtained at the end of the previous relaxation. This is not strictly true. This difference could mostly be due to the presence of viscoelastic phenomena but also to other phenomena that can occur during the unloading and relaxation between two compression events.

The evolution of the die-wall pressure shows a different behavior. For the relaxation during the first compression, the die-wall pressure decreases for all the products, which is coherent with a viscoplastic relaxation. Nevertheless, this behavior changes for the following compression. This change depends on the product but they all follow the same trends which is a transition between a clear decrease in the die-wall pressure to an increase of the die-wall pressure. To better visualize this effect, a focus on the relaxation of the fifth compression is presented in Figure 3.

As it can be seen in Figure 3, for the fifth compression, all the products show an increase of the die-wall pressure at the beginning of the relaxation which is coherent with a viscoelastic behavior as explained above. Nevertheless, depending on the product, the profiles are different. Lac and Man show an increase of the die-wall pressure during the whole relaxation. For MCC and more importantly for starch, this first increase seems to be followed by a small decrease afterwards.

The difference between these two behaviors can be interpreted by the fact that, maybe for MCC and Sta, plastic deformation still occurs, even if to a much lower extent, during the fifth compression. This possibility is backed-up by the observation of the evolution of the die-wall pressure during the different compression which is emphasized in Figure 4. On the second compression, Lac, Man and MCC exhibit a similar behavior with a slight increase at the beginning followed by a slight decrease. This behavior is less and less pronounced when the number of compressions increases. For Sta, the decrease is clearly more marked for the first compression and the increase at the beginning of relaxation is only slightly visible for the last compression.

These results confirmed the theoretical considerations presented below and show that the evolution of the die-wall pressure during relaxation can make it possible to better identify the mechanisms occurring during a relaxation experiment. Obviously, viscoelasticity and viscoplasticity are not mutually exclusive. Both phenomena happen at the same time, but the evolution of the die-wall pressure can give an idea of what is the dominant mechanism that is happening during a relaxation experiment. Results show that for the first compaction viscoplastic relaxation is predominant whereas viscoelasticity tends to become predominant when the number of compressions increases. Depending on the product it can take more compression for the plastic deformation and thus the viscoplastic behavior to decrease.

The experiments performed in this part made it possible to emphasize two important points. The first is that, as mentioned above, the evolution of the die wall pressure can indicate what is the dominant mechanism during a relaxation experiment. The second point is that, based on the relaxation curves obtained for the first compression, it can be deduced that, at 100 MPa, for the products studied, the dominant relaxation mechanism is viscoplasticity. This can be linked to the fact that, for all the products, this pressure corresponds to a state of the powder where it is still possible to perform a large plastic deformation, i.e. to a state where it is still possible to decrease significantly the porosity. Obviously, this will not be true if the pressure is increased to a value where the rate of porosity reduction with increasing pressure begins to be small. It would thus be interesting to see what is the stress evolution obtained when a high pressure is used for the relaxation text.

4.2. Case of a relaxation at high pressure (low porosity)

The idea was here to use an axial pressure as high as possible to reach a pressure range were most of the plastic deformation has already been performed and the rate of porosity reduction with increasing pressure is as small as possible. Due to the limitations of the accessible range for the die-wall pressure (limit around 200MPa), an axial pressure of 250 MPa was chosen. Relaxation experiment, using only one compression, where thus performed for all the products at this pressure. The results of the evolution of axial and radial pressure for these experiments can be found in Figure 5.

As expected, for all the products, the axial pressure presents a decrease during relaxation. For the die-wall pressure, there is a clear increase at the beginning for all the products, sign of a viscoelastic behavior. Nevertheless, afterwards for Lac and more significantly for Man, this increase is followed by a decrease. This behavior is similar to the one observed previously for the second compression at 100 MPa. This suggests that even at 250 MPa, plasticity is still occurring for these two products, which is consistent with the tabletability profile discussed above (no plateau observed) . The decrease is less marked for MCC and Sta. In the case of MCC, it was mentioned above, based on the tabletability plot, that above 200 MPa the powder was approaching the overcompression range (tendency towards a plateau on the tabletability plot). This can explain the small plastic deformation. In the case of Sta, it was shown that the powder was still away from overcompression. Nevertheless, it is also well-known that Sta is very viscoelastic (Desbois et al., 2020; Meynard et al., 2022). This high viscoelasticity might in fact compensate the residual viscoplasticity to some extent. These explanations are difficult to prove, but they emphasize the fact that, at this pressure, relaxation experiments are difficult to interpret. This will be discussed in the last part of this paper.

4.3. Interpretation of relaxation tests

The results presented above made it possible to confirm the theoretical considerations explained above. The use of die-wall pressure makes it possible to better identify the dominant time dependent phenomenon occurring during a relaxation test. At low pressures, where the rate of porosity reduction with increasing pressure is small, viscoplastic phenomena are dominant. When the pressure increases, as less plasticity is possible, viscoelastic phenomena begin to be detectable and might in fact dominate especially near the overcompression range. This means that, depending on the pressure at which the relaxation is performed, different phenomena are seen and moreover, different products might not be subject to the same phenomena as some might be closer to the overcompression range.

To illustrate this fact, relaxation curves for the four products are presented in Figure 6. To better compare the products, the evolution of the axial pressure during a relaxation experiment (1 compression) was normalized with respect to the maximal pressure which was taken equal to 50, 150 and 250 MPa depending on the experiment. Several comments can be made. First, the total relaxation is much more important at low pressure (Figure 6a) than at high pressure (Figure 6c). This means that viscoplastic phenomena, which are dominant at low pressure promote a larger stress relaxation than viscoelastic phenomena that are more dominant at high pressure. Second, depending on the pressure used to perform the relaxation, the order of the curve is different. If starch presents the highest relaxation and Lac the lowest at 50 MPa, at 250 MPa the highest relaxation is for Man and the lowest for MCC. The reason for this is clear considering the previous discussion. At low porosity, viscoplasticity is dominant and the products are ranked based on their viscoplastic behavior. At high pressure, viscoelasticity is important and the ratio between viscoelasticity and viscoplasticity is dependent on the product. The relaxation of MCC, which is close to overcompression, is mainly due to viscoelasticity whereas Man still shows a significant viscoplasticity and thus a larger relaxation. The interpretation of these curves in terms of SRS is thus very tedious as different mechanisms are in fact compared.

Considering these comments, it is possible to propose a way to use the relaxations curves. In fact, relaxation experiments should always be performed at low pressure, where viscoplastic phenomena are predominant. At these low pressures, the results can be used to assess the viscoplastic properties of different products. High pressures should always be avoided, as viscoelastic phenomena may become more significant, and the combination of both phenomena could compromise the interpretation. Finally, relaxation test should not be used to study the viscoelasticity of powders as it is difficult to avoid the presence of viscoplastic relaxation and that, according to the results presented, the relaxation due to viscoplasticity are of a greater order of magnitude than the one due to viscoelasticity. For viscoelasticity, other methodologies should be used as proposed in the literature(Desbois et al., 2020; Meynard et al., 2022).

5. Conclusion

In this article, it was shown, first using theoretical considerations and then using experiments, that the evolution of the die-wall pressure can make it possible to differentiate viscoplasticity from viscoelasticity during relaxation tests. Indeed, an increase of the die-wall pressure during relaxation is a sign of the predominance of viscoelastic deformations. This information made it possible to show that, at low pressure where the rate of porosity reduction with increasing pressure is small, the relaxation tests are mainly influenced by viscoplasticity. In contrast, as the pressure increases, relaxation phenomena are increasingly influenced by the viscoelastic behavior of the powder.

Relaxation test should thus only be used at low pressure, to study the viscoplasticity of powders. At high pressure viscoelasticity begins to be detectable during relaxation. Nevertheless, it might be impossible for most of the products to reach a pressure where viscoplasticity becomes really neglectable compared to viscoelasticity. The study of viscoelastic properties of powders should thus be made using other methodologies.

Nomenclature

ε: strain

ε^e : elastic strain

ε^p : plastic strain

ε_v: volumic strain

ε_v^e: volumic elastic strain

ε_v^p: volumic plastic strain

ε_s: distortionnal strain

ε_s^e: distortional elastic strain

ε_s^p: distortional plastic strain

ε_{ax}: axial strain

ε_{ax}^e: axial elastic strain

ε_{ax}^p: axial plastic strain

ε_{rad}: radial strain

σ: stress

σ_{ax} : axial stress

σ_{rad} : radial stress

p: hydrostatic stress

q: Von Mises deviatoric stress

E: Young's modulus

K: bulk modulus

G: shear modulus

Legend to figures

Figure 1: compaction behavior of the different products: a) compressibility (porosity vs axial pressure), b) tabletability (tensile strength vs axial pressure)

Figure 2: Evolution of axial and die-wall pressure for 5 consecutive relaxation tests made on : a) Lac, b) Man, c) MCC and d) Sta

Figure 3: Evolution of axial and die-wall pressures for the 5th compression on: a) Lac, b) Man, c) MCC and d) Sta

Figure 4: focus on the evolution and die-wall pressure for 5 consecutive relaxation tests made on: a) Lac, b) Man, c) MCC and d) Sta

Figure 5: Evolution of axial and die-wall pressure for a relaxation tests made at 250 MPa on : a) Lac, b) Man, c) MCC and d) Sta

Figure 6: relaxation curve for the different products. On each graph, the vertical axis corresponds to the axial pressure divided by the maximal axial pressure. Maximal axial pressures are: (a) 50MPa, (b) 150MPa and (c) 250 MPa.

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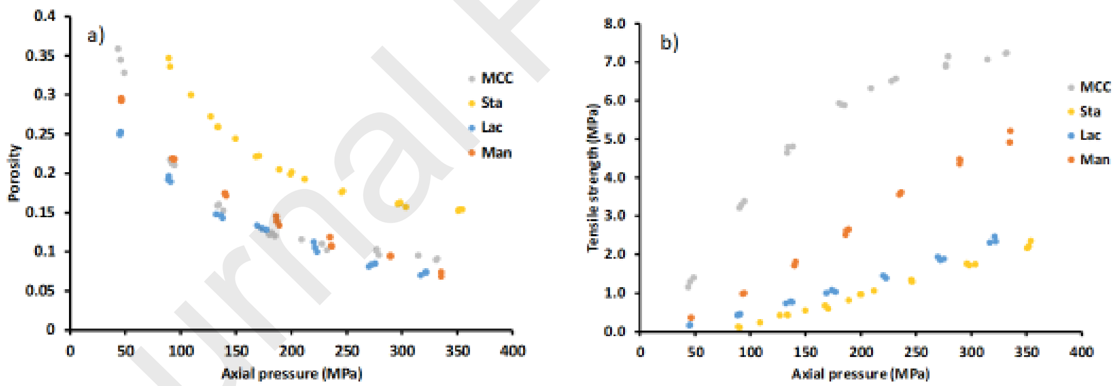

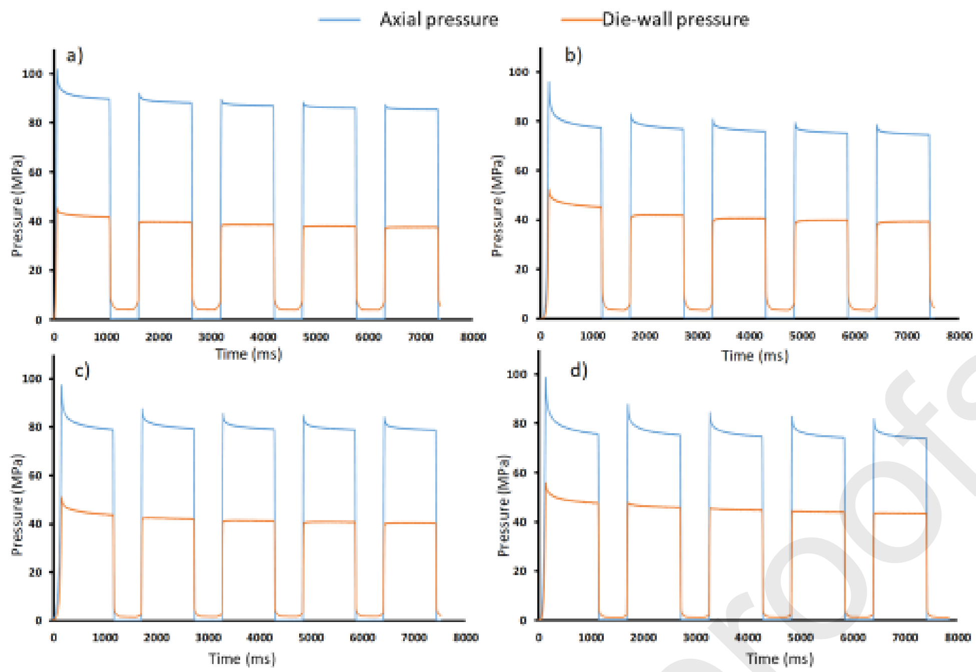
Axial pressure
Die-wall pressure
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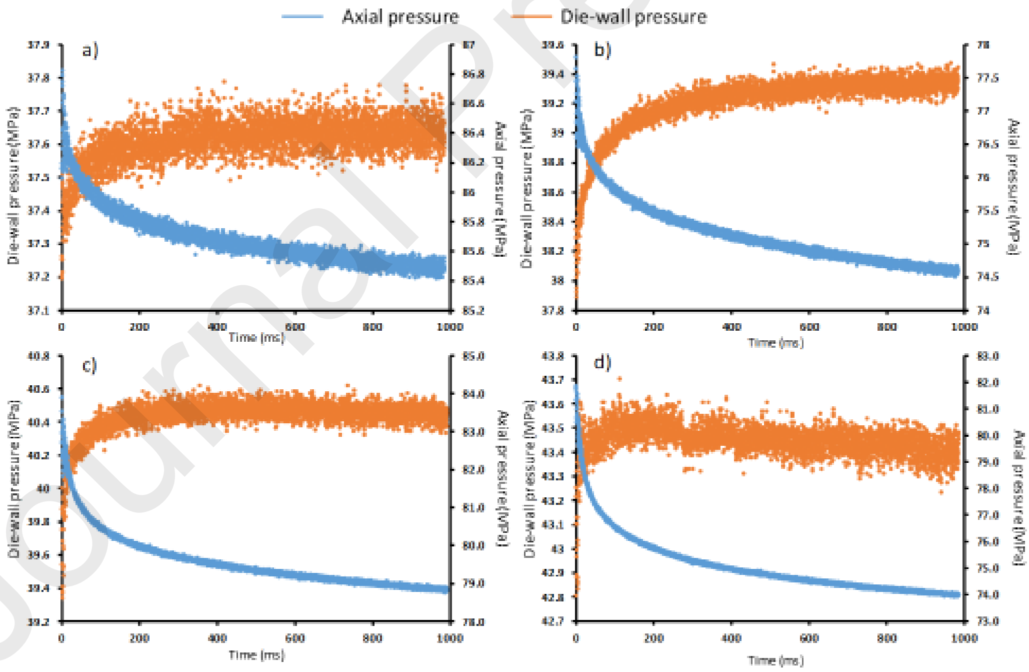
Pressure (MPa)
Time (ms)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Die-wall pressure
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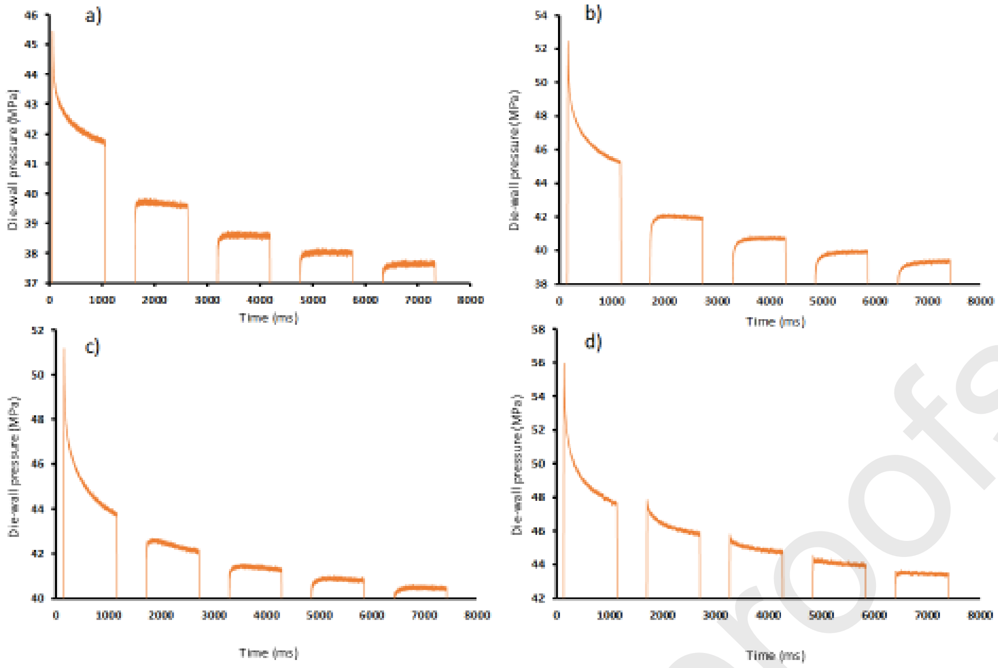
Die-wall pressure (MPa)
Axial pressure (MPa)
Time (ms)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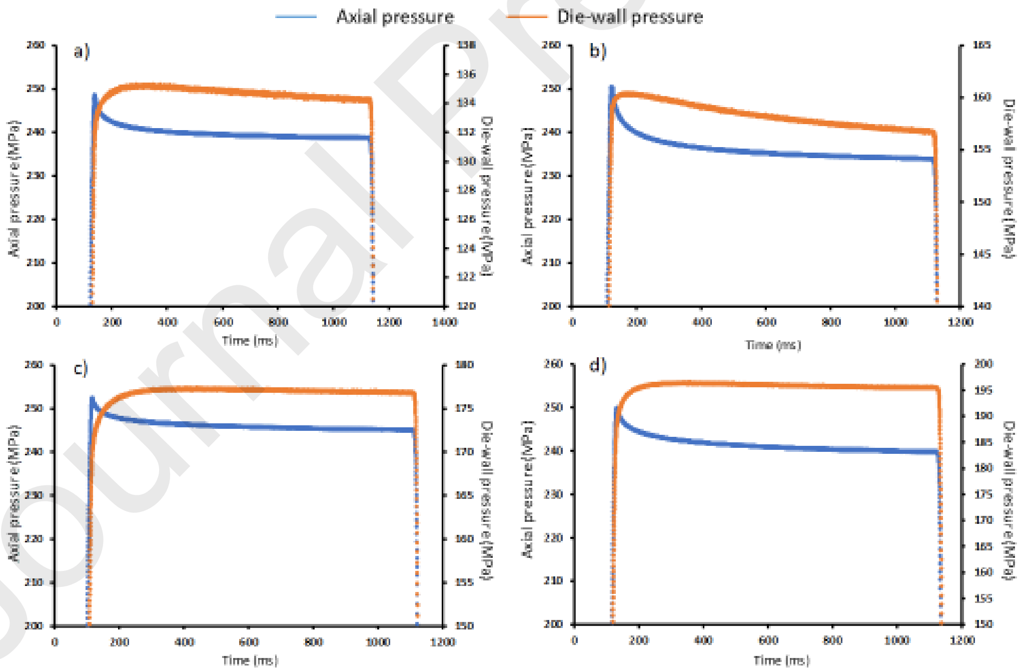
Time (ms)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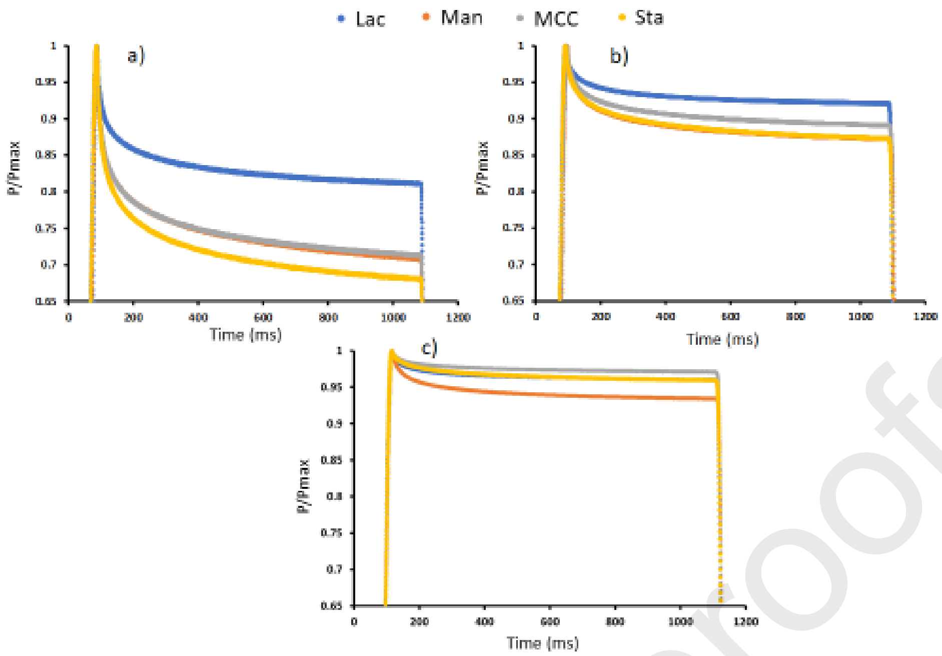

Declaration of interests

☒ 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.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: