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The incidence of pressure on the colour of compacted powders

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Abstract

This paper examines the effect of densification on the colour of compacted powders and considers whether the link is between colour and texture brought about by compaction pressure or by particle rearrangement. A methodology is established to qualify and quantify the colour and the texture of a free powder and its compact. This is done by measurements of the diffuse reflectance spectra (DRS), and experimental results are presented for the relation between densification characteristics and colour for compaction pressure of up to 30 kN.

Keywords: Compression; Compacts; Powders; Colour; Reflectance spectra

1. Introduction

The principal function of compaction is to give a form to powders by assembling the particles together under load. The volume reduction brought about by compression is due to particle rearrangement, deformation and fragmentation [1,2]. These mechanisms have an incidence on the texture and colour of the compacts. The colour of a compacted powder results from the individual properties of the particles (size, shape and mineralogy), the collective properties of the particles (size and shape distributions) and the solid-forming process (particle arrangement and surface states) [3].

A material can be characterised by two optical indices: the refractive index, n , and the absorption index, k [4]. In the case of the complex geometry of powders, apparent indices n_{app} and k_{app} are considered. In fact, a powder can be defined as a mixture of material and air; thus, changes in volume of a given amount of powder affect the apparent indices [5] as the ratio of solid to powder is changed. In particular, the absorption index k_{app} depends on the degree of voids in the system (air + material) [5]. The refractive index is related to interfacial phenomenon (air/material interfaces). If any new particles are formed due to fragmentation during compaction, the interfacial system will be changed with the appearance of new material/material interfaces resulting in a variation in n_{app} . In addition, the size of particles affects the scattering properties of the powder [3].

During a compaction process, the volume of the powder bed is reduced, air is expelled, and particles can be rear-

ranged, deformed and fragmented. As a consequence, there will be changes in the colour between the initial free powder and the final compact. These variations may be characterised by the diffuse reflectance spectra (DRS) that has already been used for other applications such as the evaluation of solid dosage forms [6] or the control of stability of tablets [7]. This paper attempts to link observations of changes in the colour of compacts with densification phenomena.

2. Materials and methods

2.1. Raw materials

The material used in this study is a mixture of 99% (w/w) of a white powder—75% (w/w) of microcrystalline cellulose (Avicel® PH105) and 25% (w/w) talc (Luzenac® Talc) with 1% (w/w) of a blue organic pigment phthalocyanin (Langdocyal® Bleu). The powder blend is prepared in an Erweka AR 402® drum mixer with a cylinder of 5-l capacity (17.5-cm diameter × 23-cm height) with a filling rate of 40% in volume, inclined about 45° to the horizontal plane and rotated at a speed of 15 rpm. The blend is obtained by successive dilution of the blue pigment in the white matrix. Two stages are required: a manual mixing stage and a machine mixing stage.

1. Firstly, all the pigment and an equal mass of the white matrix are put in a beaker and mixed. When the blend seems to be homogeneous, another similar amount of white matrix is added and mixed again. The operation is

Table 1
Material properties

| Material | True density (g/cm ³) | B.E.T. specific surface (m ² /g) | Weight median diameter d_{50} (μm) | Span ($(d_{90} - d_{10})/d_{50}$) | Bulk density (g/cm ³) |
|--|-----------------------------------|---|--------------------------------------|-------------------------------------|-----------------------------------|
| Micro-crystalline cellulose | 1.55 | 2.0 | 16 | 2.0 | 0.32 |
| Talc | 2.81 | 1.8 | 10 | 2.2 | 0.49 |
| Blue phthalocyanin | 1.71 | 59.4 | 3 | 2.3 | 0.33 |
| C ₇₅ T ₂₅ P ₁ | 1.74 | 3.6 | 18 | 1.9 | 0.35 |

repeated until 25% of the white matrix has been incorporated with the pigment.

- In the second stage, the remaining 75% (w/w) of the white matrix and the diluted colour blend are mixed in the drum at a speed of 15 rpm for 10 min. The mass of the final blend is calculated to give a filling level of 40% in volume. This blend is called C₇₅T₂₅P₁.

The properties of the materials are listed in Table 1. True solid density of the powders is determined by helium pycnometry (Micromeritics® AccuPyc 1330). Surface areas are measured by BET gas adsorption (Micromeritics® Asap 2010). Particle size is determined by dry powder laser diffraction (Malvern® Mastersizer). The weight median diameter is used to characterise particle size, but is not really significant as the cellulose and the talc are far from the spherical particles assumed in the calculations. Bulk density is measured in a measuring cylinder.

2.2. Compression

After mixing, the powder is compacted using an Instron 5567®-instrumented laboratory press at different loads of 1, 5, 10, 12.5, 15, 17.5, 20, 25 and 30 kN. Compacts of 3-cm diameter (7.1 cm²) are formed in a compression cycle at the speed of 10 mm/min, with a compact ejection speed of about 5 mm/min. The amount of material used in a test is

that required to fill the half volume (18.7 cm³) of the die and is calculated from the bulk density given in Table 1.

2.3. Diffuse reflectance spectra (DRS) analysis

The powder (as prepared in Fig. 1) and the compacts are characterised by colour and by analysis of their reflectance spectra. All the samples under study have a height greater than 7 mm.

The reflectance (R_λ) of a material, defined as the ratio of reflected light flux to incident light flux at each wavelength, is measured using a Spectrascan PR650® spectrophotometer. The reflectance measurement should be made with a diffuse incident light. However, as the apparatus used does not have an integrating sphere, measurements are performed in a small room with no windows and lit with two neon lights, thus eliminating fluctuations due to daylight. This lighting can be considered to be diffuse because the room is small and its walls diffuse the light. Moreover, reflectance is a relative measure and the error due to diffusion will occur during measurements of both the source and the sample.

The reflectance measurements are made in the range of visible wavelengths 400–700 nm as follows:

- The flux from a diffusing surface, supposed to have a reflectance equal to one, is measured. A reflectance equal to one means that the incident light flux is totally reflected and the measurement of the flux of that diffusing surface amounts to measuring the incident (source) light flux.
- The light flux from the sample is next measured in the same conditions.
- The reflectance, R_λ , is calculated by the ratio:

$$R_\lambda = \frac{\text{Flux}_{\text{Sample}}}{\text{Flux}_{\text{Diffusing surface}}} \quad (1)$$

- Five measurements are made for each sample and the mean value is used. The variation factor is majored to 1% on the measurement field (400–700 nm) and the

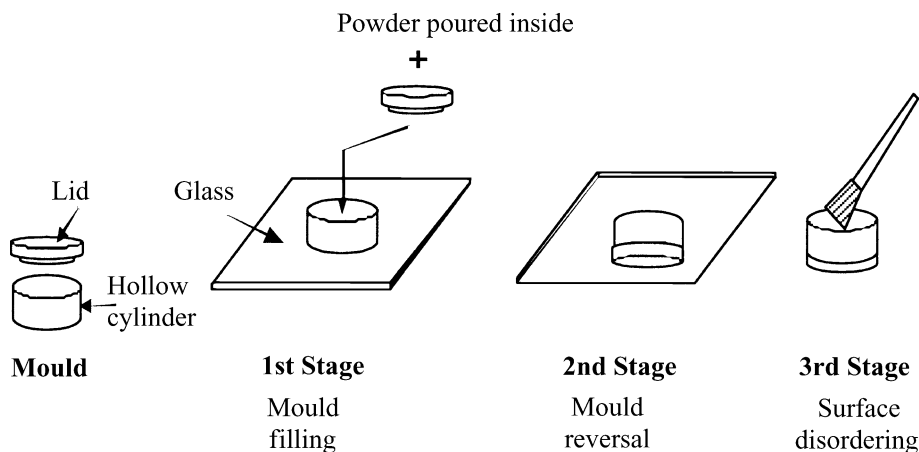


Fig. 1. Preparation of powder sample for measurement of the DRS.

Table 2
Correspondence between wavelength and the associated tint

| λ (nm) | 380–436 | 436–495 | 495–566 | 566–589 | 589–627 | 627–780 |
|-----------------------|---------|---------|---------|---------|---------|---------|
| Characteristic colour | Violet | Blue | Green | Yellow | Orange | Red |

relative error is estimated as being 0.72% for five measurements.

DRS can be related to the attribute of colour sensations in the case where the source is an achromatic light (a white light):

1. The area under the curve is proportional to the total energy reflected and is similar to luminance.
2. The width of the peak or the field under the curve is linked to the notion of saturation or “purity” of hue.
3. Finally, the peak position or the principal field determines the chromacity that gives the notion of tint (Table 2).

3. Results

The powder $C_{75}T_{25}P_1$ is compacted at different pressures. The compression cycle and the properties of the compacts are summarised in Fig. 2 and Table 3. The main characteristics of the powder and the compacts (consolidation pressure, relative density, calculated porosity and area under DRS) are given in Table 3. It can be seen that although the experiments are limited to low pressures of less than 42 MPa, the overall loss of porosity is around 50%. The area under the DRS, representing the luminance of the product, is seen to decrease with the increase of load. Firstly, the principal stage of the compaction cycle is examined by comparing the properties of the free powder and the compact. Then, different load ranges are studied: low (0–10 kN), medium (10–20 kN) and high (20–30 kN).

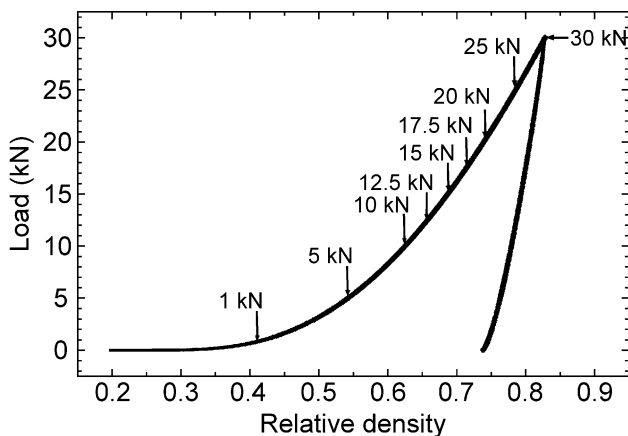


Fig. 2. Compression cycle of $C_{75}T_{25}P_1$ powder—evolution of the load vs. relative density.

3.1. Free powder compared with compact

DRS are surface measurements and, therefore, a change of the surface texture will have an influence on the reflectance measurement. Frodyma et al. [8] reported that using a high pressure to pack a sample causes 1–2% of variation in reflectance reading. In the case of a change from the state of a free powder to that of a compacted powder, we observe the same variation (Table 3) of 2% decrease for compact reflectance at the peak ($R_{\lambda, \text{peak}}$), but closer examination of the DRS (Fig. 3) shows the following:

1. The shape of the spectrum is modified. The characteristic peaks are practically the same, 2% of variation in reflectance at 452–454 nm, and the powder and the compact keep their blue colour. However, the difference between the two spectra is more important in the absorbent zone (for longer wavelengths around 550 nm and above) at around 7%.
2. In addition, the luminance, expressed by the area under the DRS, is lower for the compact than for the free powder. The compact is more absorbent than the powder.

All these observations correspond to the hypothesis that the reduction in volume caused by the load of 30 kN is significant enough to induce an increase in the apparent absorption index. The more the apparent absorption index increases, the less is the reflectance in the absorbent zone of the DRS. Moreover, deformation of particles under load creates a greater surface and a decrease in the light diffusion. These two phenomena (reduction in air volume and particle deformation) are responsible for the modifications in the DRS.

Table 3
Characteristics of the powder $C_{75}T_{25}P_1$ and its compacts

| Load (kN) | Pressure (MPa) | Relative density | Calculated porosity (%) | Area ₄₀₀ ⁷⁰⁰ under DRS (nm) | Peak (nm) | Width (nm) | Height, $R_{\lambda, \text{Peak}}$ |
|-----------|----------------|------------------|-------------------------|---|-----------|------------|------------------------------------|
| 0 | 0 | 0.20 | 80 | 193.9 | 456 | 300 | 0.79 |
| 1 | 1.4 | 0.4 | 60 | 193.8 | 468 | 300 | 0.79 |
| 5 | 7.1 | 0.50 | 50 | 194.3 | 452 | 300 | 0.79 |
| 10 | 14.1 | 0.56 | 44 | 194.7 | 464 | 300 | 0.80 |
| 12.5 | 17.7 | 0.60 | 40 | 188.0 | 464 | 300 | 0.79 |
| 15 | 21.2 | 0.62 | 38 | 188.8 | 468 | 300 | 0.79 |
| 17.5 | 24.8 | 0.64 | 36 | 187.6 | 468 | 300 | 0.79 |
| 20 | 28.3 | 0.66 | 34 | 186.4 | 468 | 300 | 0.79 |
| 25 | 35.4 | 0.69 | 31 | 184.6 | 468 | 300 | 0.79 |
| 30 | 42.4 | 0.72 | 28 | 180.7 | 452 | 300 | 0.77 |

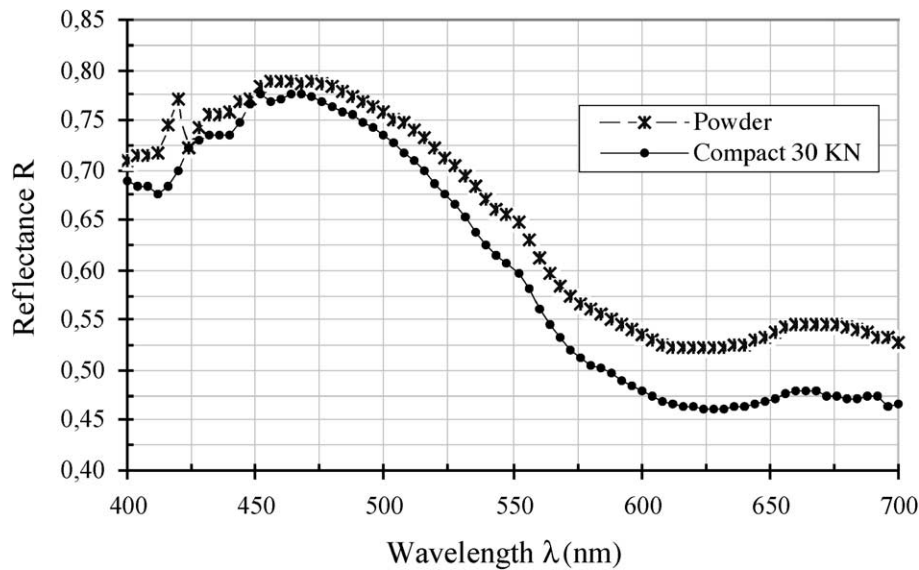


Fig. 3. DRS of $C_{75}T_{25}P_1$ powder and its compact made at 30 kN.

3.2. Variations at low loads

The above results show that there is a modification of the DRS due to changes in the surface state of the sample brought about by a load of 30 kN, which is the highest load used in our experiments, but still in the low pressure range (42 MPa). It is also interesting to study the effect of variation of compression load on DRS at even lower loads (Fig. 4 and Table 3).

The results show that all spectra are the same, whatever the load is. There is neither a change in the luminance (area under the DRS) nor in the general aspect of the spectra. Nevertheless, there is a large decrease in porosity of about 40% from the free powder state to compacted state (Fig. 2

and Table 3). This can be explained by the fact that DRS is a measurement of surface properties. To analyse a powder, we need to prepare a sample (Fig. 1) that necessarily involves packing the powder. Therefore, it can be supposed that when the compact is formed, the applied load (1, 5 and 10 kN) just contributes to deaerate the powder bed without disturbing the particle organisation at the surface. Moreover, at these low pressures, it can be assumed that the mechanisms involved are merely particle rearrangement with neither particle deformation nor particle fragmentation that would modify light scattering and the apparent refractive index. At these low pressures, the surface texture of compacts is not changed; therefore, the DRS is still the same and the colour is not changed.

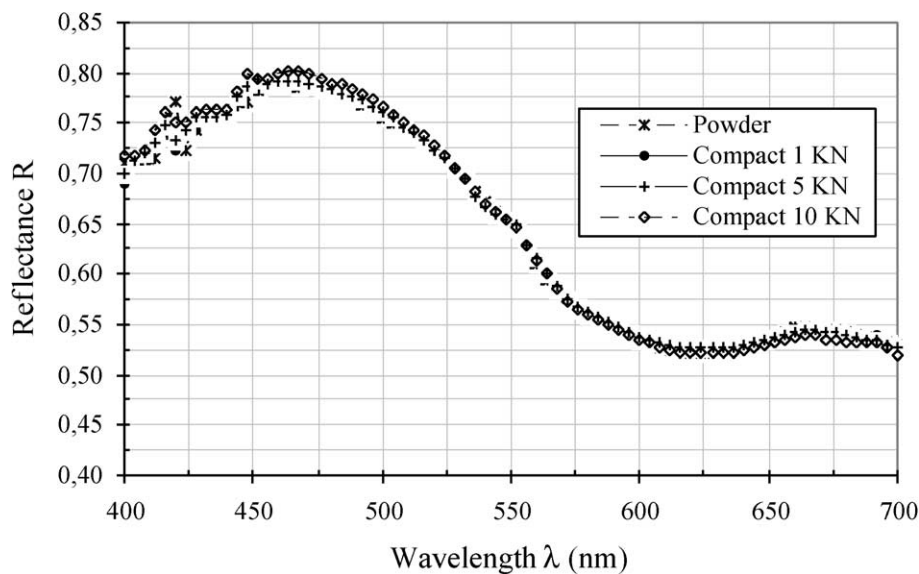


Fig. 4. DRS of $C_{75}T_{25}P_1$ powder and its compacts made at 1, 5 and 10 kN.

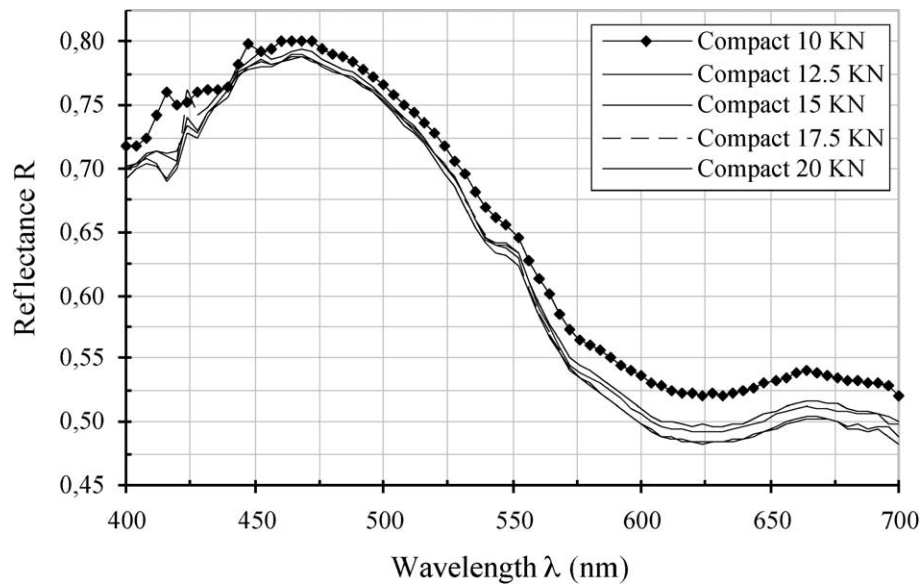


Fig. 5. DRS of $C_{75}T_{25}P_1$ compacts made at 10, 12.5, 15, 17.5 and 20 kN.

3.3. Variations at medium loads

If the consolidation load is increased (Fig. 5 and Table 3), it can be seen that:

1. The change in the spectrum in the absorbent zone (550–700 nm) appears above loads of 12.5 kN. The compact made at 12.5 kN is more absorbent, luminance is decreased, and the reflectance value is 4% less in the long wavelengths (600–700 nm). The reflectance spectrum varies only if there is a modification of the surface state. This phenomenon, from the point of view of the compression process, can be linked to compaction

mechanisms. Up to 10 kN, the powder is packed by deaeration and particle rearrangement of the powder bed. It may be assumed that at 12.5 kN, the maximum compacity without fragmentation is reached. Above this load, densification involves particle deformation and fragmentation. These phenomena cause changes in the particle size distribution and also bring about modifications in the pore size distribution at the surface of the compact. This modification of the surface texture of the compact causes a variation in the apparent optical indices and, therefore, a change in the DRS.

2. Compacts made at 12.5, 15, 17.5 and 20 kN have practically the same spectra. It can be seen that there is a

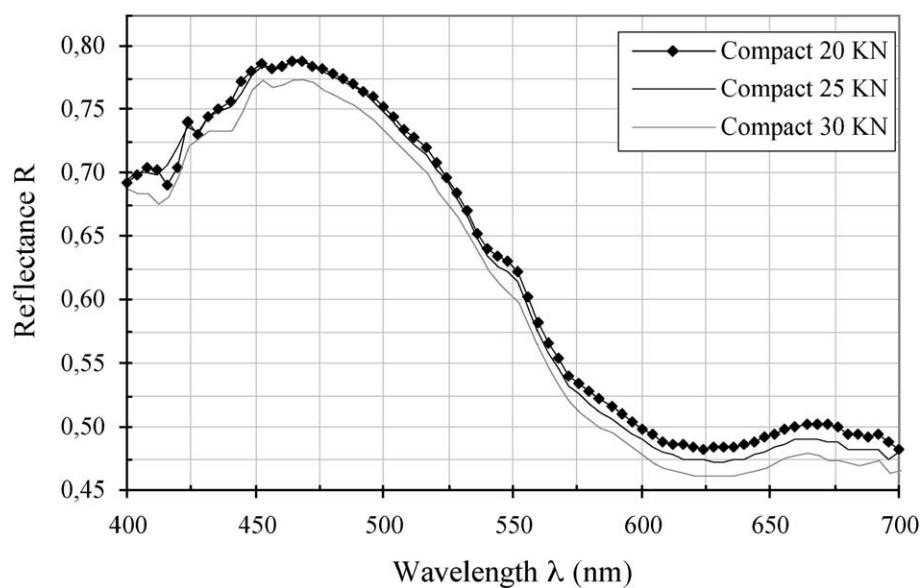


Fig. 6. DRS of $C_{75}T_{25}P_1$ compacts made at 20, 25 and 30 kN.

small variation in luminance with increase in pressure. This is almost certainly due to small variations in the surface porosity of the compacts. In fact, the overall reduction of pore volume of the compacts is around 6% in the load range from 12.5 to 20 kN.

3.4. Variations at high loads

It has been seen that the DRS of compacts formed at loads up to 12.5 kN does not vary with the value of the load. At the load of 12.5 kN, the DRS of the compact changes; however, for loads from 12.5 to 20 kN, the DRS does not vary very much. What happens when the consolidation load is increased above these values? The results are presented in Fig. 6 and Table 3. These show that the luminance of compacts slowly decreases, and the compacts are slightly more absorbent when pressure is increased.

This change in the luminance can be explained by the same compaction mechanisms as previously mentioned. The powder is essentially composed of microcrystalline cellulose, and cellulose is well-known for its aptitude for plastic deformation under load. It may be assumed that for the formulation used here, plastic deformation of particles is the predominant compaction mechanism at high loads. Moreover, a previous study [9] showed an affinity of the pigment for microcrystalline cellulose, where a pigment has the tendency to form agglomerates in the cellulosic matrix. The plastic deformation of cellulose, therefore, causes a dispersion of the pigment. As the pigment is more absorbent, this brings about a decrease of the luminance.

4. Conclusion

Consolidation pressure has an effect on the diffuse reflectance spectrum (DRS) of a powder. The DRS is a surface

measurement; therefore, if it changes, it means that there must be modifications in the surface texture. The compaction process involves several mechanisms: first, a reduction in the volume of the powder bed, then particle deformation and, finally, fragmentation. These modifications in particle size cause variations in pore size distribution inside and at the surface of the compact. These phenomena bring about a modification of the apparent optical indices (n_{app} and k_{app}) and, consequently, the DRS of the compacts.

Nomenclature

| | |
|---------------------|--|
| DRS | Diffuse reflectance spectrum |
| n, k | Optical indices: refractive and absorption indices, respectively |
| n_{app}, k_{app} | Apparent optical indices |
| R_λ | Reflectance |
| $R_{\lambda, peak}$ | Reflectance value at the peak wavelength |
| $Area_{400}^{700}$ | Area under the reflectance spectrum between 400 and 700 nm |

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