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Tablet strength, porosity, elasticity and solid state structure of tablets compressed at high loads

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Abstract

The effects of applying high loads to a powder bed on the properties of compacts of sodium chloride, sucrose and polyethylene glycol were studied. The porosity (apparent zero) and tensile strength levelled off when a high enough load was applied. This levelling off corresponded to a pronounced elasticity, and for polyethylene glycol, an irreversible increase in particle density and melting point was detected. Tablet elasticity (volume increase during decompression) is discussed in terms of two contributing factors. Firstly, rupture of bonds between tabletting particles and, secondly, a pronounced volume expansion of the particles (crystal lattice). The latter mechanism was believed to be important only for polyethylene glycol. The possibility of utilizing the tensile strength at zero porosity as an intrinsic strength value for the tabletted materials was also tested. Significant differences between different size fractions were obtained even when tablets compressed to zero porosity were compared. This was especially pronounced for sodium chloride.

Keywords: Tablet; Compaction load; Porosity; Tensile strength; Elastic recovery; Solid state structure

1. Introduction

During consolidation of a powder bed a reduction in porosity takes place. This reduction in compact volume brings the particles into close proximity to each other. The reduced distance between the particles facilitates creation of bonds and makes the particles stick together into a coherent compact.

Three different bonding types are normally considered in direct compression of pharmaceutical materials: intermolecular forces, solid bridges and mechanical interlocking (Führer, 1977). Van der Waals forces are probably the most important type of intermolecular force, responsible for holding the particles together in a tablet. Hydrogen bonds and electrostatic forces are other examples of forces that act over a distance between particles. The nature of solid bridges depends on the chemical structure of the material and these forces arise when there is real contact between particles. Bonding by hooking or twisting of particles depends on the surface texture and shape of the particles. The dominating bond type is rarely known, but probably depends on various factors

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including the degree of compression and the inherent properties of the material. In the high porosity range, the principal attractions between the particles for most pharmaceutical materials have been suggested to be intermolecular forces (Karehill and Nyström, 1990; Nyström et al., 1993), whereas in the low porosity range it cannot be excluded that solid bridges are also of increasing importance.

Mechanical properties, such as Young's modulus and fracture mechanics parameters, have been determined for pharmaceutical materials (Mashadi and Newton, 1987; Roberts and Rowe, 1987; Bassam et al., 1990; York et al., 1990; Newton et al., 1993). This is mostly done by extrapolation of compact measurements to the corresponding value for a compact porosity of zero, which is considered to correspond to a material property, rather than just a characteristic of the specimen (Mashadi and Newton, 1987). Knowledge of these properties could make it possible to predict the compaction behaviour of a pharmaceutical material (Bassam et al., 1990). The prerequisite for such an extrapolation is probably that the principal physical structure of the compact is also retained in the low porosity range covered by extrapolation.

By applying high pressures to a powder bed, low porosities of the resulting compact can be achieved. When the porosity of the tablet is close to zero, the structure of the tablet should be different from the structure at 'normal' porosities (5–25%). For most pharmaceutical materials, where a compact can be described as a dispersion of solid units in a continuous air phase (Nyström et al., 1993), the final porosity reduction may eventually represent a transformation to a new physical structure, where the solid constitutes the continuous phase. Thus, the bonding structure of the resulting compact may also be altered.

The solid state properties of the material can be affected when high pressures are applied to a powder bed. There have been several reports concerning an increase in the true density under compression, of directly compressible materials (Ponchel and Duchêne, 1989; Blattner et al., 1986; Pedersen and Kristensen, 1994). This phenomenon is mostly reported for materials with low melting points, known to form compacts of low porosities, e.g. aspirin and polyethylene glycols. This effect on the solid state properties of the materials is reported under pressure, and seems to be reversible when the load is reduced.

The aim of this study was to evaluate the effect of applying a range of high compaction loads on the tensile strength and porosity of the formed compacts. Firstly, it was investigated if it is a feasible technique to use the strength at zero porosity as an intrinsic tablet strength. This concept has been used by applying extrapolation to obtain a single unique value for the tensile strength of the material. Secondly, it was tested whether specific compaction loads could be identified, above which no further reduction in porosity or increase in strength was obtained. Thirdly, it was tested if compaction at very high loads had a tendency to increase the elastic recovery of the compacts formed. It was also investigated if such a mechanism resulted in irreversible changes in solid state properties.

2. Materials and methods

2.1. Materials

Three different materials were studied (Table 1). For sodium chloride (crystalline, puriss,

Table 1
Size and surface characteristics of the test materials. Average values for three measurements and the corresponding standard deviations in parentheses

Material	Sieve size fraction (μm)	Volume specific surface area (cm^2/cm^3)
Sodium chloride	180–355	262 (5)[a]
Sodium chloride	20–40	3356 (47)[b]
Sucrose	180–355	280 (1)[a]
Sucrose	20–40	3131 (58)[b]
Polyethylene glycol	90–180	–[c]
Polyethylene glycol	40–80	1856 (13)[b]
Polyethylene glycol	20–40	2329 (23)[b]

[a] Measured with a transient permeameter according to Eriksson et al. (1990).

[b] Measured with a Fisher sub sieve sizer (USA).

[c] Not measured.

Table 2
Effect of compaction at 1200 MPa on solid state properties, melting point, heat of fusion and density. Average values of three measurements and the corresponding standard deviations in parentheses

Material	Melting point (°C)		Heat of fusion (J/g)[a]		Density (g/cm^3)[b]	
	Raw material	Tablet	Raw material	Tablet	Raw material	Tablet
Sodium chloride	–[c]	–	–	–	2.154 (0.0002)	2.155 (0.0006)
Sucrose	190.1 (0.2)	190.5 (0.6)	129.8 (2.4)	131.9 (3.5)	1.587 (0.0004)	1.576 (0.0014)
Polyethylene glycol	59.7 (0.1)*	61.8 (0.6)	187.0 (0.7)	188.4 (2.0)	1.073 (0.0001)*	1.213 (0.0005)

[a] Measured with DSC 20, Mettler, Switzerland.

[b] Measured with helium pycnometry for the coarsest fraction of the materials compacted at 1200 MPa (Accurpyc 1330, Micromeritics, USA).

[c] Not measured.

*Paired t-tests give significant differences ($p < 0.05$) between raw material and tablet.

Kebo-lab, Sweden) and sucrose (crystalline, Svenskt socker AB, Sweden) the coarse fraction, 180–355 μm, was prepared by dry sieving (Retsch, Germany). The 20–40 μm fraction of sodium chloride was obtained by milling with a mortar mill (Retsch KM 1, Germany), and for the preparation of sucrose, a pin disc mill (Alpine 63C, Alpine AG, Germany) was used. The milled material was then air classified (Alpine 100 MZR, Alpine AG, Germany) to prepare the desired fraction. For the third material used, polyethylene glycol 8000 (crystalline powder, Aldrich-Chemie, Germany), the raw material was air classified to obtain 40–80 μm and 20–40 μm size fractions. A fraction of 90–180 μm was also prepared by dry sieving. Sodium chloride and polyethylene glycol mainly undergo volume reduction by plastic deformation, while sucrose is a more fragmenting material (Nyström et al., 1993). The apparent particle densities (B.S. 2955, 1958) of all the materials were determined by helium pycnometry (Table 2) (Accurpyc 1330, Micromeritics, USA). The volume specific surface areas of the test materials were determined by permeametric methods (Table 1). All the materials were stored for at least 2 days at a relative humidity of 40% (Nyqvist, 1983) before compaction.

2.2. Compaction

An instrumented single punch press (Korsch EK 0, Germany) was used. The material was weighed on an analytical balance and manually poured into the die. Lubrication was performed before every compaction by compression of a loose powder plug of magnesium stearate. The maximum upper punch pressure during compression was recorded for each tablet and at least five tablets were prepared for each load. A deviation not exceeding 4% from the desired compaction load was accepted. The compressions of sucrose and sodium chloride were performed at 50 (or 75 for the coarse fraction), 100, 200 and 300 MPa with a tablet diameter of 11.3 mm. Compaction was also performed at 300, 500, 800 and 1200 MPa with a punch diameter of 5.65 mm for sodium chloride and sucrose. For polyethylene glycol, compaction was performed at 50, 100, 300, 500, 800 and 1200 MPa, and the diameter of the compacts was 5.65 mm. The thicknesses of the sodium chloride and sucrose compacts were in all cases 4.0 mm ± 3%, by adjustment of the distance between the punches and the weight of the material. For the polyethylene glycol compacts the distance between the punches was kept constant by applying a constant pressure to a metallic cylinder, and the different loads were obtained by varying the amount of material filled into the die. The thickness of the compacts of polyethylene glycol subsequently varied between 3.30 and 4.40 mm.

2.3. Determination of tensile strength

The tablets were stored for not less than 2 days at 40% relative humidity before diametral compression tests were carried out at a speed of 4 mm/min (Holland C50, Great Britain), and the tensile strength of the compacts was calculated according to Fell and Newton (1970). The compacts failed in tension, except for the compacts prepared at the highest compression loads where some splitting under the diametral compression test occurred, indicating shear and compressive failure, especially for sucrose, 20–40 μm at 800 and 1200 MPa.

2.4. Determination of porosity

The porosities of the formed compacts were calculated from the apparent particle density (Table 2), dimensions and weight of the tablet.

2.5. Extrapolation of tensile strength to zero porosity

The logarithm of the tensile strength of the specimen was plotted as a function of porosity. A linear regression to zero porosity was performed (Ryskewitch, 1953), and the theoretical tensile strength at zero porosity was calculated.

2.6. Determination of elastic recovery

The porosity was recorded as a function of pressure every millisecond during compression and decompression (Armstrong and Haines-Nutt, 1972; Duberg and Nyström, 1986). Corrections for deformation of the punches were also performed.

The elastic recovery during the tablet processing ($E_{\%}$) was calculated:

$$E_{\%} = \frac{T_{\max} - T_{\min}}{T_{\min}} \times 100 \quad (1)$$

where $T_{\min}$ is the minimum thickness during compression and $T_{\max}$ is the maximum thickness of the compact obtained during decompression. Here, the decompression phase was followed until the upper punch left the tablet (at an upper punch pressure of 2 MPa). The elastic recovery was determined for two size fractions of the test materials, and average values for three tablets are presented. Data for the evaluation of elastic recovery were recorded at 300 and 1200 MPa for the materials, and the tablet diameter was in all cases 5.65 mm. Usually it is preferable to use porosity data for calculation of elastic recovery, due to the increased information it gives about the effect of elasticity on bonding between particles (Duberg and Nyström, 1986), i.e. the porosity is a more sensitive measure for expressing the risk of rupture of bonds during decompression of the compact. In this study, however, elastic recovery was calculated from changes in thickness of the compact, because of the negative porosities obtained in some cases.

2.7. Characterization of solid state properties as measured by DSC, helium pycnometry and X-ray analysis

The melting point and heat of fusion for the raw material and tablets prepared with a compression load of 1200 MPa were compared. A small piece of material was cut from the tablet and used for characterization. These measurements were made for polyethylene glycol and sucrose, and the coarsest fractions of the materials were used. The melting point of sodium chloride could not be determined with the apparatus used (DSC 20, Mettler, Switzerland). A heating rate of 10°C/min was used and the measurements were carried out in aluminium pans (open system). As an additional characterization of eventual changes in solid state properties, the densities of tablets compressed at 1200 MPa were determined with a helium pycnometer (Accupyc 1330, Micromeritics, USA) after 2 days storage at 40% relative humidity and compared with the densities of the raw materials. The raw material of polyethylene glycol and a tablet of the same material were analysed by X-ray diffraction with Cr K_{α} radiation with the aid of an XDS2000 diffractor (Scintag, USA). The experimental procedure has been described in more detail elsewhere (Sjökvist et al., 1989).

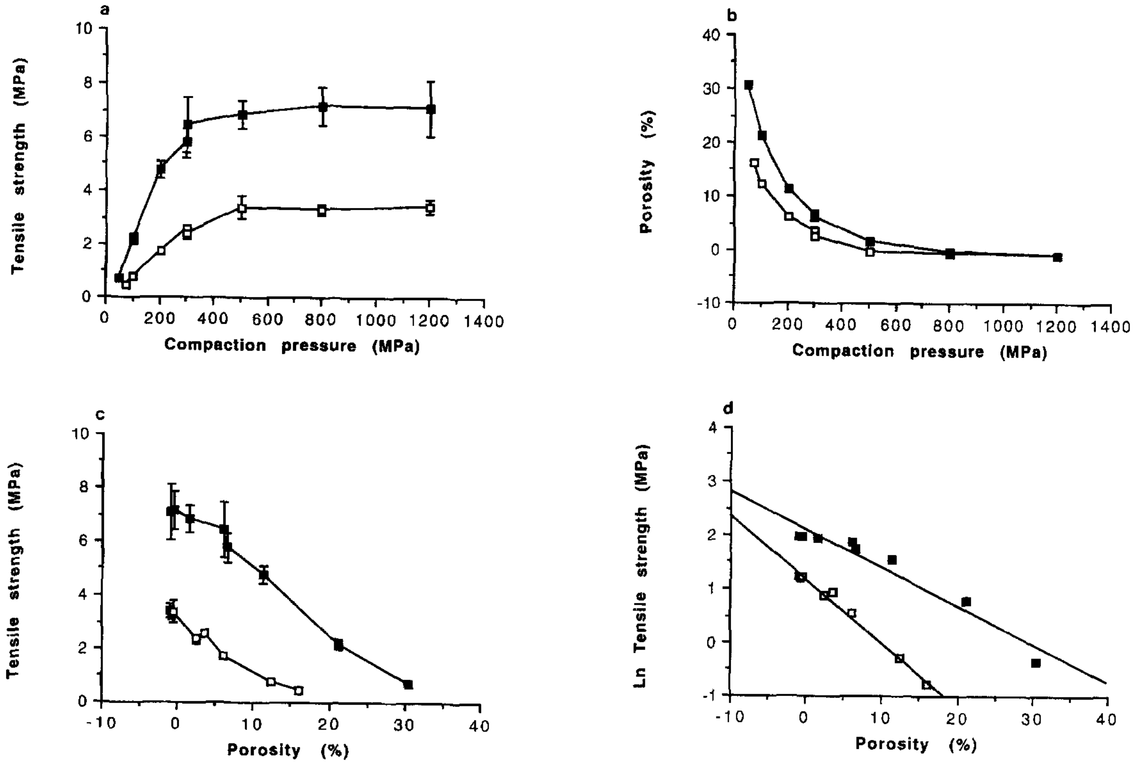

Fig. 1. Tensile strength of sodium chloride tablets. Sodium chloride size fractions: 180–355 μm (□) and 20–40 μm (■). Confidence intervals for $p = 0.05$ are given in (a)–(c). (a) Compaction pressure as a function of tensile strength. (b) Compaction pressure as a function of porosity. (c) Porosity as a function of tensile strength. (d) Porosity as a function of the natural logarithm of tensile strength. Correlation coefficients for the 180–355 and 20–40 μm size fraction are 0.992 and 0.966.

3. Results

3.1. *Effect of compaction pressure on tensile strength and porosity of the compact*

For all the materials, porosity values, or apparent porosity values, close to zero were reached. Zero porosity was reached both at pressure, i.e. under compression, and after ejection and 2 days additional storage of the compacts (Fig. 1b, 2b and 3b). However, the compaction pressure for obtaining zero porosity varied between the materials. Also, for the tensile strength, higher compression loads resulted in a continuous increase in tensile strength until a certain value was reached, after which a higher compression load did not result in any further increase in tensile strength (Figs. 1a, Fig. 2a and 3a). To some extent the thresholds in compression load resulting in zero porosity (Figs. 1b, 2b and Fig. 3b) coincide with the load needed to give a plateau in tensile strength. For polyethylene glycol these plateaux were reached at a compaction pressure of approximately 300 MPa and for sodium chloride and sucrose the corresponding value was approximately 500 MPa. The plateau in tensile strength for the finest size fraction of sodium chloride was reached at approximately 300 MPa. For the highest compression loads, 800 and 1200 MPa, capping tendencies were observed for sucrose, especially for the 20–40 μm fraction. This capping tendency could be seen during testing of diametral compression strength (Fig. 2a) as a moderate decrease in strength with increasing compaction load (Nyström et al., 1978).

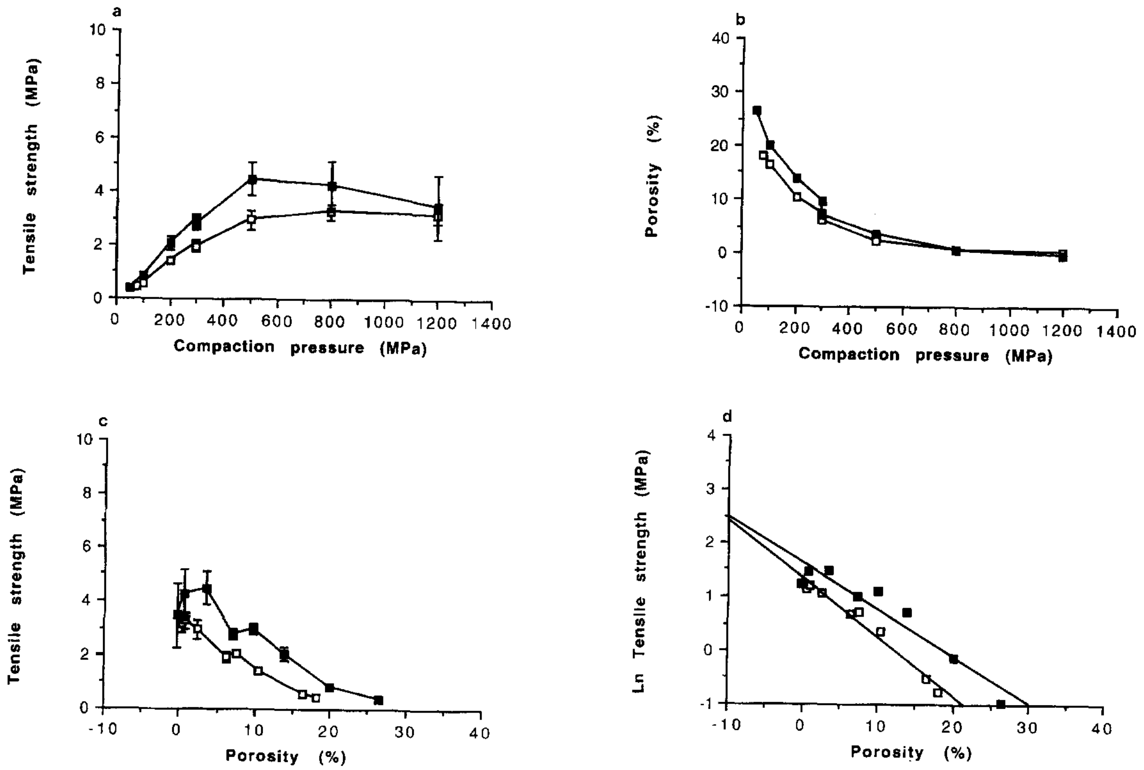

Fig. 2. Tensile strength of sucrose tablets. Sucrose size fractions: 180–355 μm (□) and 20–40 μm (■). Confidence intervals for $p = 0.05$ are given in (a)–(c). (a) Compaction pressure as a function of tensile strength. (b) Compaction pressure as a function of porosity. (c) Porosity as a function of tensile strength. (d) Porosity as a function of the natural logarithm of tensile strength. Correlation coefficients for the 180–355 and 20–40 μm size fraction are 0.986 and 0.952.

3.2. Extrapolation/compaction to zero porosity

The sodium chloride systems correlated reasonably well with the extrapolation procedure according to Ryskewitch (1953), i.e. the data adjusted to a straight line (Fig. 1d). For sucrose the correlation was not as good, probably due to the observed capping tendencies (Fig. 2d). Generally, for polyethylene glycol the deviation from a straight line was considerable (Fig. 3d). It can be seen from Figs. 1d, 2d and 3d that the different size fractions yield different values of tensile strength at zero porosity. The largest difference between the size fractions is seen for sodium chloride and polyethylene glycol. The two size fractions of sucrose gave similar profiles, and the differences in tensile strength between the two size fractions at zero porosity was minute. It is thus uncertain if the extrapolation procedure or even the direct compaction of tablets to zero porosity could be utilised to estimate an intrinsic strength value of a material. The extrapolation is especially doubtful for the polyethylene glycol system because of the apparent negative porosities obtained during compression, even at low loads.

3.3. Effect of compaction at high loads on elasticity and solid state properties

Generally, an increase in compaction load resulted in an increased elastic recovery for the test materials (Table 3). No major effect of particle size on the elastic properties of the compacts was seen. The elastic recovery was largest for polyethylene glycol and smallest for sodium chloride.

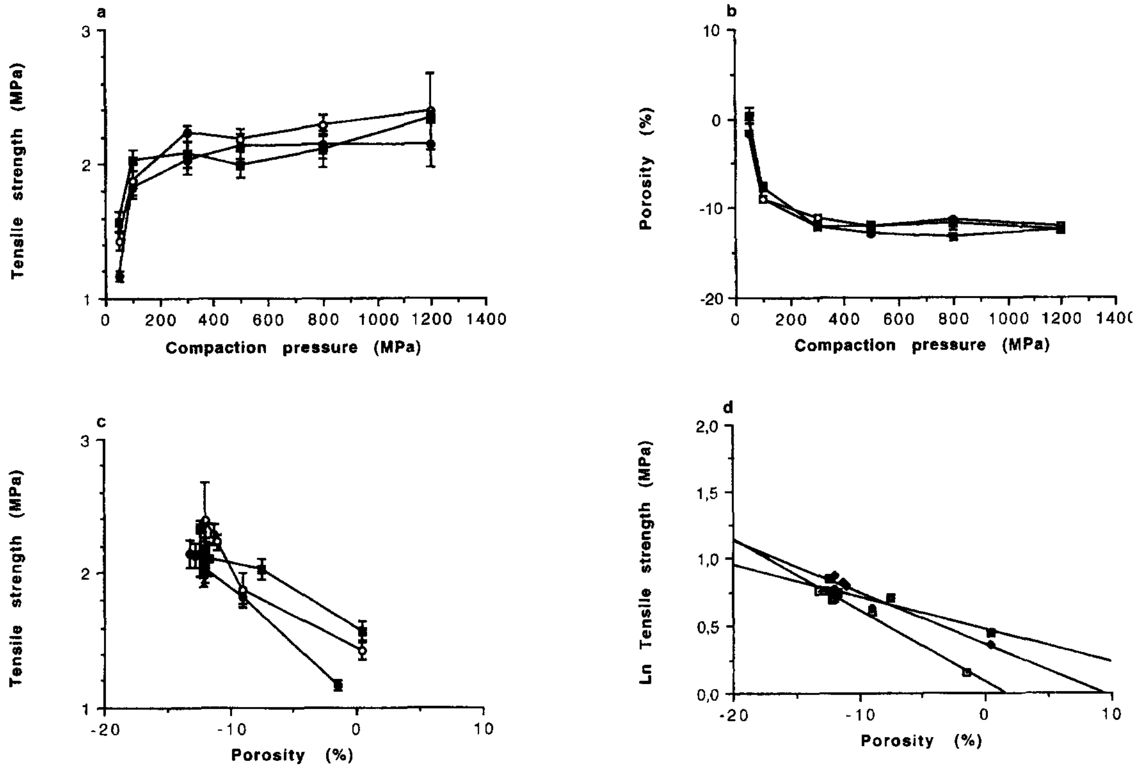

Fig. 3. Tensile strength of polyethylene glycol tablets. Polyethylene glycol size fractions: 90–180 μm (●), 40–80 μm (○) and 20–40 μm (■). Confidence intervals for $p = 0.05$ are given in (a)–(c). (a) Compaction pressure as a function of tensile strength. (b) Compaction pressure as a function of porosity. (c) Porosity as a function of tensile strength. (d) Porosity as a function of the natural logarithm of tensile strength. Correlation coefficients for the 90–180, 40–80 and 20–40 μm size fraction are 0.995, 0.969 and 0.909.

The melting point and heat of fusion for polyethylene glycol increased after compaction (Table 2), indicating an effect of the compression procedure on the solid state properties of the material. The effect on the solid state properties of polyethylene glycol was also supported by density measurements, where an increase in density of the material was detected (Table 2). The structure of the polyethylene glycol (Craig and Newton, 1991) was too disordered to obtain any information on the effect of compaction on the volume of the unit cell of the material by X-ray analysis.

No significant effect of compaction at 1200 MPa on particle density, melting point or heat of fusion could be seen for sodium chloride and sucrose (Table 2). For sucrose there was a tendency that the density of the material decreased due to compaction.

4. Discussion

When applying high loads during tabletting, low porosity compacts may be obtained (Figs. 1b, 2b and 3b). Further increase in compaction load gave no corresponding volume reduction within the compact, due to the limited space for additional fragmentation and plastic deformation. The energy thus supplied by an increase in compaction load was stored as elastic energy, i.e. during decompression, elastic recovery of the particles took place and this restoration also resulted in breakage of bonds that were formed between tablet particles during compression. This elastic recovery resulted in a plateau in both tensile strength and porosity (Figs. 1a, 1b, 2a, 2b, 3a and 3b). Even a reduction in the tensile strength was obtained for sucrose (Fig. 2a), due to capping.

Table 3

The effect of high compaction load on tensile strength and elastic recovery. Standard deviations are given in parentheses. For the tensile strength data $n = 5$ and for the elasticity data $n = 3$

Material	Size fraction (μm)	Tensile strength (MPa)			Elastic recovery[b] (%)		
		Compacted at 300/500 MPa[a]	Compacted at 1200 MPa	Strength ratio[c]	Compacted at 300 MPa	Compacted at 1200 MPa	Elasticity ratio[c]
Sodium chloride	180–355	3.39 (0.34)	3.40 (0.20)	1.00	0.68 (0.10)	2.30 (0.10)	3.39
Sodium chloride	20–40	6.85 (0.43)	7.09 (0.84)	1.04	1.37 (0.11)	1.31 (0.09)	0.96
Sucrose	180–355	2.94 (0.27)	3.14 (0.22)	1.07	2.11 (0.05)	2.86 (0.68)	1.36
Sucrose	20–40	4.47 (0.50)	3.45 (0.98)	0.77	3.28 (0.36)	3.91 (1.08)	1.19
Polyethylene glycol	90–180	2.04 (0.07)	2.14 (0.19)	1.05	4.77 (0.19)	11.8 (0.39)	2.48
Polyethylene glycol	20–40	2.09 (0.19)	2.35 (0.18)	1.13	4.51 (0.07)	13.1 (0.30)	2.95

[a] At 300–500 MPa the porosity almost reached zero and subsequently the tensile strength values levelled off.

[b] Relative increase in compact thickness as calculated from Eq. (1).

[c] Ratio of data obtained at 1200 and 300/500 MPa, respectively.

Table 4
Tensile strength values at zero porosity (or the value closest to zero porosity), and at maximum apparent negative porosity values. Standard deviations for $n = 5$ are given in parentheses

Material	Size fraction (μm)	Tensile strength[a] (MPa)	
		At 'zero' porosity values	At minimum porosity values
Sodium chloride	180–355	3.39 (0.34)	3.40 (0.20)
Sodium chloride	20–40	7.16 (0.55)	7.09 (0.84)
Sucrose	180–355	3.14 (0.22)	3.14 (0.22)
Sucrose	20–40	3.45 (0.98)	3.45 (0.98)
Polyethylene glycol	90–180	1.17 (0.05)	2.14 (0.23)
Polyethylene glycol	20–40	1.57 (0.06)	2.35 (0.18)

[a] From Figs. 1c, 2c and 3c.

In this study extremely low porosities (apparently zero porosity) of the specimens were obtained. The high compression loads applied (up to 1200 MPa) may explain why such low porosities have been reached, even after ejection and storage of the compacts for 48 h (Figs. 1b, 2b and 3b). These high compaction loads can affect the structure of the raw material. For some materials an increase in true density has been reported under pressure (Ponchel and Duchêne, 1989; Blattner et al., 1986; Pedersen and Kristensen, 1994). This effect could probably explain the apparently negative porosities observed. For acetylsalicylic acid it was reported that the volume of the unit cell was affected under pressure, and that this reduction in volume was reversible (Pedersen and Kristensen, 1994). For the materials included in this study apparent zero porosity or even apparent negative porosities were reached. Even if the absolute values of the porosities are to some extent uncertain, due to limited precision in the determination of the thickness of the compacts (mainly due to the varying thickness over the tablet surface), extremely low porosities were reached, even after ejection of the tablets.

For polyethylene glycol, the increase in melting point, heat of fusion and density after compaction (Table 2), indicated an influence on the solid state properties of the material. The increase in density can probably be explained by an irreversible decrease in the volume of the unit cell, but this was not possible to confirm by X-ray diffraction. This irreversible effect is possibly due to the high compaction load used for preparation of the compacts. Some less probable explanations for the increase in density of polyethylene glycol are related to the use of gas pycnometry. Here, the increase in true density registered may depend on the incapability of the helium pycnometry method to register the true density of the material due to inclusions of air within uncompressed particles. When enough load is applied to these particles, the included air may be squeezed out, and would thus give a false increase in density. However, when measuring the density of tablets, there may also be closed pores within the compacts, which would correspond to an even higher true density of the compact formed. There was a slight tendency that the apparent density of the sucrose particles decreased after compaction (Table 2), which can be due to amorphisation of the crystalline material when compacted at high loads (Ek et al., submitted), but this effect was not significant in this study.

Due to the fact that apparent negative porosities for the tablets cannot be reached without affecting the structure of the materials, the apparent negative values obtained must correspond to a value close to true zero porosity (Table 4). Sodium chloride and sucrose show no difference in these two porosity values and a corresponding levelling off in tensile strength, indicating that the minimum porosity is reached at zero porosity. For materials with similar properties to polyethylene glycol, an alteration of the solid state properties of the materials may be expected when compacted, indicated by the continuous decrease in apparent negative porosity to a plateau level and an increase in tensile strength (Fig. 3b).

The materials included in this study form compacts with different structures, i.e. different bonding types and bonding surfaces. The forces holding sodium chloride particles together in a tablet are believed to be mainly intermolecular forces, but solid bridges are probably of importance as well (Nyström et al., 1993). This means that a more or less continuous transformation of bonding type is expected, and probably the change in bonding structure is not too pronounced. Compacts of sucrose are probably held together mainly by intermolecular forces, which indicates that when the porosity of the compact is reduced and approaching zero, a change in physical structure should take place due to fusion of particles. If, eventually, zero porosity is reached the tablet may be seen as a large single crystal. For such an extreme compact the stresses created during diametral compression should probably then result in failures going through the tablet particles instead of around them, as may be expected at higher porosities. However, in this study no drastic change in bonding properties at the higher loads for any of the materials was observed (Figs. 1a, 2a and 3a). This was explained on the basis that the failure in the compact during strength measurement was going around particles and that the separation distances between the particles remained approximately constant for compaction loads exceeding 300–500 MPa. Thus, the minute volume of air probably left within the compacts can cause considerable restriction in bonding. The bonding forces between the particles could also be hindered further by contamination of the particle surfaces (Dollimore and Heal, 1961).

Thus in theory, the tensile strength determined at zero porosity should represent a measure of the material's strength (Mashadi and Newton, 1987; Newton et al., 1993), independent of processing parameters, such as particle size, compression speed and dimensions of compacts. However, in this study a considerable effect of the particle size was seen for tablets compressed at zero porosity (Figs. 1c, 2c and 3c). Besides the explanation given above, that even a minute volume of air could drastically reduce the interparticulate bonding, another explanation for this could be that the different size fractions may represent different materials with varying strength characteristics, and that the tensile strength at zero porosity is in reality different for the different size fractions (Newton et al., 1993), due to, for example, differences in initiation and propagation of cracks. The strength value obtained for tablets compressed to zero porosity (in this study corresponding to compaction loads exceeding 300–500 MPa) may then be seen as some type of intrinsic tablet strength for each respective starting material. However, it must be considered that one single, chemical compound did not deliver one single value. On the contrary, as seen especially for sodium chloride, it seems that the preprocessing of the starting material (e.g., milling to different particle sizes) will give bonding properties that will not be equalised by compression, even if zero porosity is reached. The obtained maximum tensile strength value for sodium chloride in this study (approximately 8 MPa) is much lower than the values reported in the literature (approximately 100 times higher) for tensile strength of whisker crystals (Macmillan, 1972). These whisker crystals are almost perfect with an extremely low concentration of defects in the crystal structure, and reported values (experimental and theoretical) are quite similar. Due to the non-perfect crystals forming the compact, a lower value is expected and the interparticle voids and contamination on the particle surfaces further reduce the tensile strength of the compact. Thus, the results illustrate that the use of a strength value at zero porosity is incapable of delivering one, single measure for the strength of materials.

An increase in compaction load will increase the elastic deformation of the particles, which can be seen as an elastic recovery of the formed compact (Table 3). Elastic recovery then probably depends on two factors. Firstly, the regaining of some of the original distance between atoms and ions may occur, the elasticity then depends on the elastic recovery of the particles themselves. Secondly, processing parameters such as friction during ejection can result in rupture of bonds between tablet particles. For sodium chloride and sucrose the elastic recovery is probably related mainly to such a rupture of bonds, i.e. results in an increased average distance between the tablet particles. On the other hand, for polyethylene glycol, the energy applied during compression is used to alter the solid state properties of the material when compacted (Table 2). Thus, during compression, the volume of the particles is decreased due to the changes in the unit cell dimensions. Since this is to some extent a reversible process, a subsequent increase in cell dimensions during compression results in a pronounced elastic recovery for the polyethylene glycol system when ejected (Pedersen and Kristensen, 1994).

5. Conclusion

It does not seem possible to compress tablets to zero porosity, to obtain a single measure for the strength of a material. In this study, different size fractions gave different values at zero porosity. This may be due to the different inherent properties of the size fractions, but will more probably be due to the fact that, in the pressure range applied, no complete displacement of air was possible. Thus, minute amounts of air build up thin films around the tablet particles, thus strongly decreasing the compact strength. Subsequently, the existence of such minute air gaps between the tablet particles will result in the failure created during strength testing going around the particles, rather than through them. Different size fractions of the compacted material will thus represent different bonding areas in the final compact even when zero porosity is almost reached. If zero porosity is reached, extremely high compaction loads are needed, higher than used in this study, to remove the gas and contamination layers surrounding the tablet particles. When these films or layers are removed, a pronounced increase in strength of the compact can be expected.

For the materials included in this study, the tensile strength of the compacts level off at 300–500 MPa. In this interval, porosity values close to zero are reached or the porosity stops decreasing (for polyethylene glycol, apparent negative porosity values are reached). When the applied load exceeded 300–500 MPa, no further strength increase was obtained, but an increased elastic recovery was recorded, which resulted in rupture of bonds created during compression. This increased elastic recovery resulted in a levelling off of tensile strength. For sucrose, there was even a decrease in tensile strength with a decrease in compaction pressure. Probably, measurement of axial tensile strength (Nyström et al., 1978) would have given a more pronounced decrease in strength after compaction with a load above 300–500 MPa, as suggested by Ek et al. (submitted).

During compaction, an increase in the density of the crystals is obtained if a certain compaction load is applied, supplying the energy needed for reducing the distance between the ions or molecules in the crystal structure. This effect may be reversible for some materials and irreversible for others, as seen clearly for polyethylene glycol in this study.

It seems that the sensitivity to changes in solid state properties induced by compaction varies between materials. For several pharmaceutical materials an increase in true density can probably be detected. Some materials may show irreversible changes in solid state properties and for other materials a reversible effect can be monitored. The stress needed to develop these changes varies, depending on the properties of the material. Materials with a low melting point are probably more sensitive to these effects, e.g. polyethylene glycol where an increase in the particle density after compaction at 1200 MPa is detected, indicating an increase in crystallinity for the material when compressed at high loads. Also, materials with higher melting points can be expected to be affected by compaction, but the compaction probably results in a decrease in crystallinity, i.e. amorphisation of the material as shown by others for microcrystalline cellulose. The effect of compaction on solid state properties can be examined by density measurements and thermal analysis.

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