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Mechanisms for post-compaction changes in tensile strength of sodium chloride compacts prepared from particles of different dimensions

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Abstract

The changes in mechanical strength of sodium chloride compacts, prepared from particles of different dimensions, were followed during a post-compaction storage period of up to 4 days at different relative humidities. This revealed that the increase in tablet strength can occur by two different mechanisms referred to as mechanism α and β. Compacts of fine particulate sodium chloride were shown to increase in tablet strength in the presence of moisture by means of a mechanism described as molecular rearrangement at the surface of the particles. For this mechanism, mechanism α, the increase in tablet strength could be time delayed, with respect to the formation of the compacts, by storing the compacts at 0% relative humidity. Compacts of coarse particulate sodium chloride were shown to increase in tablet strength at all relative humidities, mechanism β, but the presence of moisture in the compact slowed down the process of increasing tablet strength. These changes in tablet strength were postulated as due to either a visco-elastic particle deformation (stress relaxation), where the relative positions of the particles in the compacts are changed continuously after ejection from the die, or a stabilisation of bonds which had already been formed during compression.

Key words: Post-compaction tensile strength increase; Bond formation process; Visco-elastic deformation; Solid bridge stabilization; Molecular rearrangement; Mechanism α; Mechanism β

1. Introduction

Arguments have been presented, in the literature, indicating that the physical character of a compact of pharmaceutical materials, prepared under normal tableting conditions, can be described in terms of an aggregate of individual but strongly cohering particles, (Alderborn and Glazer, 1990; Karehill and Nyström, 1990a). Thus, the tensile strength of such a specimen theoretically is governed by the sum of the bonding forces of all individual interparticulate bonds in the failure plane of the compact. Based on this physi-

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cal model of a compact, attempts have been made to explain how variations in the properties of the material, e.g., particle dimensions (Alderborn and Nyström, 1982), and variations in the compaction process, affect the mechanical strength of the specimen in terms of changes in the interparticulate bonding characteristics of the compact. In such studies, different experimental procedures concerning the time which elapses between compaction and strength analysis and the storage conditions (primarily the relative humidity) during this storage period have been used. At the same time, a number of studies (Rees and Shotton, 1970; Bhatia and Lordi, 1979; Rue and Barkworth, 1980; Down and McMullen, 1985; Ahlneck and Alderborn, 1989; Karehill and Nyström, 1990b; Alderborn and Ahlneck, 1991) have demonstrated the existence of a change in the mechanical strength of tablets during storage. An interpretation of this observation is that interparticulate bonds are formed or disturbed in the compact without the application of any mechanical force. One factor of importance for such a bond formation process seems to be the presence of water in the material, due to sorption of water from the environment.

When compacts are exposed to atmospheres with high relative humidities, multimolecular adsorption of water can occur followed by condensation of water in the pores of the compact. It has been suggested (Ahlneck and Alderborn, 1989; Luangtana-Anan and Fell, 1990; Karehill and Nyström, 1990a) that the presence of condensed water in the pores of a compact will reduce the bonding force, due to intermolecular attraction forces, between particles in the compact and thus decrease the compact strength. Furthermore, fluctuations in the humidity of the environment to which the compacts are exposed, can cause desorption of water which can lead to crystallisation of material dissolved in sorbed water. The consequence might be the formation of interparticulate bonds and an increased compact strength (Ahlneck and Alderborn, 1989).

Under constant ambient conditions below a humidity at which condensation occurs, two mechanisms responsible for a post-compaction strength increase have been suggested in the literature for compacts of non-hygroscopic materials. The first mechanism can be described as a continuing deformation of particles during a certain time period after the compression phase (Rees and Rue, 1978; Karehill and Nyström, 1990b). The result of such a visco-elastic deformation (also referred to as stress relaxation) will be the formation of new interparticulate bonds and a subsequent increased coherency of the specimen. The second mechanism was described as the formation of new interparticulate bonds due to the repositioning of amorphous surface material resulting in the concentration of material at zones of interparticulate contact, i.e., at the interparticulate junctions (Ahlneck and Alderborn, 1989; Alderborn and Ahlneck, 1991; Elamin et al., 1994). The incidence of the latter process has been shown to be related to the relative humidity of the storage environment and thus the amount of sorbed water at the particle surfaces. The role of water is thus to increase the mobility of the molecules, probably by transforming amorphous material from a glassy to a rubbery state.

The mechanisms suggested have been based primarily on observations of the physical characteristics of sodium chloride compacts. However, the dimensions of the sodium chloride particles, as well as their mechanical treatment before compaction, varied in the studies. Thus, it is still unclear whether or not there is more than one mechanism responsible for changes in tablet strength during storage of compacts of non-hygroscopic materials, such as sodium chloride, and the aim of this paper was to elucidate this issue. A study of the mechanical strength of sodium chloride compacts, stored under different relative humidities, for different storage times and prepared from sodium chloride particles with different dimensions, was considered a suitable experimental strategy in this context. A fundamental knowledge of the mechanisms responsible for post-compaction strength changes of compacts is necessary in order to establish prerequisites (material, process and environment related factors) for such changes to occur and thus provide an increased understanding of the performance of materials used in tableting.

2. Materials and methods

2.1. Preparation of powders

A series of powders of sodium chloride (crystalline puriss., Kebo, Sweden) was prepared in the following way:

The size fractions 20–40 and 40–60 μm were prepared by milling the raw material in a pin disc mill (Alpine 63C, Alpine AG, Germany) followed by air classification (Alpine 100 MZR, Alpine AG, Germany). The size fractions 90–125, 212–300A and 425–500 μm were prepared by dry sieving of the raw material with ordinary laboratory sieves. The fraction 212–300B μm was prepared by milling the size fraction > 500 μm gently in a mortar followed by dry sieving.

All powders were stored at 0% relative humidity (in desiccators over phosphorus pentoxide) and room temperature for not less than 7 days before compaction.

2.2. Compaction and determination of tablet tensile strength

Compacts were prepared in an instrumented single punch tablet machine (Korsch EK 0, Germany) equipped with 1.13 cm flat-faced punches at maximum upper punch pressures of 75 and 150 MPa. The distance between the punch faces at the lowest position of the upper punch was in all cases 3 mm at zero pressure (the compaction pressures were controlled by varying the amount of powder filled into the die). External lubrication with magnesium stearate suspension (1% w/w in ethanol) was performed before each compression. The powder for each compact was weighed individually on an analytical balance and poured manually into the die. The motor of the machine was then started, when the upper punch was at its highest position. The motor was stopped and the flywheel arrested manually, directly after the compression process and the compact was finally removed from the die table.

The diametral compression strength of the tablets (Holland C50, U.K. or Erweka TBH 28, Germany) was measured immediately after compaction ($t = 15$–20 s) or the tablets were transferred directly to mini-desiccators (glass vials of diameter 2.5 cm and height 6 cm, one tablet in each) for storage at four relative humidities: 0, 33, 57 and 75%. The relative humidities in the different desiccators were controlled by phosphorus pentoxide or by a saturated salt solution (Nyqvist, 1983). The salt solutions were gently stirred during the storage periods. After storage times of 5, 10, 15, 60 and 5760 min (i.e. 4 days), the diametral compression strength was determined.

In addition, the diametral compression strengths of compacts of size fractions 20–40 and 425–500 μm were measured after storage periods including a transfer of compacts between 0 and 57% relative humidity in the following way. The tablets were stored at 0 or 57% relative humidity for 5760 min and then transferred to 57 or 0% relative humidity, respectively, and stored for a further period of 5760 min. Finally, the compacts were transferred back to the first relative humidity and stored for another period of 4320 min (i.e., 3 days).

In all cases, the tensile strength of the compacts was calculated according to Fell and Newton (1970). The results presented are means of five determinations.

2.3. Compaction and determination of tablet surface area

Compacts were prepared in the instrumented single punch tablet machine by compressing the powder in a special die, placed on the die table, by rotating the fly wheel of the tablet machine by hand as described earlier (Alderborn at al., 1985). No lubrication of the die was used in this case. For all size fractions, compacts were prepared at 75 MPa and for the size fraction 425–500 μm, compacts were also prepared at 100 MPa.

The air permeability of the tablets was measured with the Blaine apparatus immediately after compaction ($t \approx 45$ s) or after storage of the compacts (placed in the die) in a desiccator at 0% relative humidity or at ambient relative humidity (approx. 57%) for 15 and 60 min. Only compacts prepared from the size fractions 20–40 and 425–500 μm were stored in this way. The specific surface area of the compacts was calculated from the permeability equation (Alderborn at al., 1985). The results presented are the means of five determinations.

Table 1
Powder and tablet permeametry surface area together with calculated values for absolute and relative changes in surface area per pressure unit for all size fractions

Size fraction (μm)	Powder surface area (cm^{-1})	Tablet surface area [a] (cm^{-1})	ΔS/MPa [b] (cm^{-1} MPa^{-1})	ΔS_{rel}/MPa [c] (MPa^{-1})
20– 40	4883	5509	8.35	0.002
40– 60	2929	3781	11.36	0.004
90–125	700 [d]	1436	9.81	0.014
212–300 A	314	570	3.14	0.011
212–300 B	373	815	5.89	0.016
425–500	168	454	3.81	0.023

[a] Measured approx. 45 s. after compaction. Compaction pressure 75MPa.
[b] Absolute change in surface area per unit pressure.
[c] Relative change in surface area per unit pressure.
[d] Estimated from mean particle size in the sieve fraction and a surface-to-volume shape factor of 7.5.

3. Results

3.1. Dimensions and compaction characteristics of particles

Microscopy examination of the powders showed that the unmilled particles generally had a cubical shape with a relatively smooth surface texture. The two finest size fractions had particles with a less defined but fairly regular shape. The relatively coarse but milled particles (212–300

Table 2
Tensile strength and 95% confidence interval of tablets compacted at 75 MPa: results for all size fractions under different storage conditions (time and relative humidity)

RH (%)	Storage time (min)	Size fractions 20–40 μm		40–60 μm		90–125 μm		212–300 A μm		212–300 B μm		425–500 μm	
		Tensile strength (MPa)	Confidence interval (MPa)	Tensile strength (MPa)	Confidense interval (MPa)	Tensile strength (MPa)	Confidense interval (MPa)	Tensile strength (MPa)	Confidense interval (MPa)	Tensile strength (MPa)	Confidense interval (MPa)	Tensile strength (MPa)	Confidense interval (MPa)
0	0	0.73	0.07	0.50	0.07	0.69	0.06	0.17	0.08	0.44	0.07	0.17	0.08
	5	0.74	0.09	0.49	0.05	0.72	0.06	0.36	0.02	0.64	0.10	0.31	0.10
	10	0.67	0.09	0.55	0.08	0.71	0.06	0.33	0.02	0.62	0.05	0.28	0.17
	15	0.66	0.09	0.53	0.10	0.70	0.07	0.32	0.05	0.64	0.03	0.36	0.08
	60	0.69	0.13	0.54	0.06	0.72	0.05	0.37	0.08	0.66	0.09	0.39	0.08
	5760	0.67	0.14	0.11	0.03	0.69	0.06	0.39	0.06	0.61	0.03	0.40	0.08
33	5	0.85	0.14	0.61	0.05	0.68	0.05	0.34	0.03	0.60	0.05	0.28	0.11
	10	0.84	0.16	0.70	0.07	0.67	0.01	0.33	0.05	0.60	0.04	0.37	0.08
	15	0.84	0.18	0.69	0.10	0.72	0.08	0.37	0.08	0.57	0.03	0.36	0.11
	60	1.05	0.28	0.81	0.17	0.73	0.13	0.32	0.02	0.61	0.03	0.38	0.06
	5760	1.51	0.20	1.15	0.16	1.07	0.15	0.35	0.04	0.64	0.04	0.37	0.07
57	5	1.29	0.13	0.93	0.03	0.75	0.09	0.33	0.04	0.62	0.06	0.25	0.06
	10	1.54	0.22	1.03	0.10	0.78	0.08	0.31	0.02	0.63	0.07	0.29	0.03
	15	1.42	0.33	1.30	0.16	0.84	0.08	0.27	0.08	0.63	0.03	0.36	0.05
	60	1.85	0.01	1.40	0.23	0.86	0.15	0.31	0.03	0.64	0.10	0.33	0.10
	5760	2.72	0.16	2.48	0.12	1.48	0.11	0.30	0.04	0.71	0.07	0.43	0.08
75	5	1.68	0.29	1.41	0.09	0.96	0.15	0.18	0.07	0.51	0.06	0.12	0.05
	10	1.51	0.11	1.34	0.05	0.83	0.11	0.19	0.07	0.45	0.05	0.21	0.10
	15	1.40	0.28	1.46	0.06	0.95	0.16	0.17	0.02	0.42	0.09	0.13	0.07
	60	1.04	0.13	1.10	0.13	0.79	0.04	0.15	0.04	0.38	0.11	0.17	0.06
	5760	0.75	0.06	– [a]	–	– [a]	–	– [a]	–	0.18	0.02	– [a]	–

[a] Not measurable.

μm B) were more irregularly shaped compared to the other particles. The milling procedure also gave a slightly increased powder surface area compared to fraction 212–300 μm A (Table 1). It seems reasonable that the milled particles would also show, on a molecular level, a disordered structure on the fracture surfaces (Kontny et al., 1987; Ahlneck and Zografi, 1990). Such a disordering might affect the potential of the material to form interparticulate bonds when compacted and also affect the incidence of post-compaction strength changes during storage (Elamin et al., 1994), as will be discussed below.

Compacts formed at 75 MPa generally showed a greater tablet surface area compared to the powder surface area (Table 1). It seems thus reasonable that the sodium chloride particles, although they generally can be described as a material which compresses by deformation (De Boer et al., 1978; Duberg and Nyström, 1986), showed some fragmentation during the compression process (Down, 1983; Down and McMullen, 1985; Alderborn and Glazer, 1990). There is a tendency that a decreased original particle size increased the absolute increase in surface area per applied pressure unit while the converse applied for the relative increase in surface area per pressure unit (Table 1). This is consistent with earlier observations (Alderborn and Nyström, 1985). A reduction in original particle size also generally gave an increased tablet strength, measured immediately after compaction (Tables 2 and 3).

A comparison between fractions 212–300 μm

Table 3

Tensile strength and 95% confidence interval of tablets compacted at 150 MPa: results for all size fractions under different storage conditions (time and relative humidity)

RH (%)	Storage time (min)	Size fractions											
		20–40 μm		40–60 μm		90–125 μm		212–300 A μm		212–300 B μm		425–500 μm	
		Tensile strength (MPa)	Confidence interval (MPa)	Tensile strength (MPa)	Confidence interval (MPa)	Tensile strength (MPa)	Confidence interval (MPa)	Tensile strength (MPa)	Confidence interval (MPa)	Tensile strength (MPa)	Confidence interval (MPa)	Tensile strength (MPa)	Confidence interval (MPa)
0	0	1.95	0.08	1.94	0.13	1.29	0.08	0.75	0.09	1.08	0.09	0.60	0.03
	5	1.90	0.24	2.36	0.45	2.12	0.15	1.00	0.10	1.60	0.21	0.84	0.07
	10	2.02	0.17	2.50	0.36	2.12	0.31	1.10	0.06	1.80	0.09	0.99	0.17
	15	1.76	0.29	2.42	0.16	2.26	0.18	1.06	0.05	1.77	0.14	0.98	0.05
	60	2.24	0.61	2.22	0.19	2.34	0.14	1.05	0.07	1.87	0.01	1.06	0.04
	5760	2.05	0.30	2.58	0.14	2.21	0.35	0.97	0.13	1.83	0.20	0.87	0.09
33	5	2.10	0.20	2.55	0.51	2.15	0.21	0.87	0.11	1.36	0.09	0.77	0.08
	10	2.06	0.08	2.28	0.32	2.11	0.15	0.95	0.05	1.58	0.09	0.86	0.03
	15	2.20	0.15	2.45	0.37	2.13	0.28	0.97	0.10	1.66	0.13	0.91	0.13
	60	2.19	0.15	2.73	0.50	2.10	0.17	1.02	0.08	1.67	0.11	1.04	0.22
	5760	2.92	0.38	4.27	0.30	3.03	0.27	0.86	0.20	1.96	0.09	0.89	0.10
57	5	2.59	0.22	2.38	0.27	1.77	0.09	0.73	0.06	1.24	0.09	0.71	0.06
	10	2.86	0.50	2.75	0.34	1.84	0.12	0.78	0.03	1.38	0.07	0.70	0.05
	15	2.88	0.27	2.70	0.19	1.94	0.17	0.76	0.11	1.37	0.03	0.72	0.15
	60	3.31	0.12	3.28	0.30	2.21	0.21	0.77	0.05	1.60	0.10	0.82	0.10
	5760	4.88	0.56	4.31	0.22	2.88	0.38	1.01	0.06	2.11	0.43	0.90	0.05
75	5	2.90	0.55	2.66	0.15	1.72	0.14	0.74	0.04	1.19	0.05	0.65	0.06
	10	2.65	0.25	2.98	0.15	1.86	0.12	0.72	0.07	1.30	0.06	0.66	0.08
	15	2.58	0.25	3.14	0.23	1.96	0.12	0.71	0.04	1.27	0.04	0.65	0.07
	60	1.79	0.21	3.10	0.18	1.96	0.09	0.65	0.03	1.35	0.11	0.69	0.07
	5760	– [a]	–	0.78 [a]	0.05	1.28	0.07	– [a]	–	1.03	0.06	– [a]	–

[a] Not measurable.

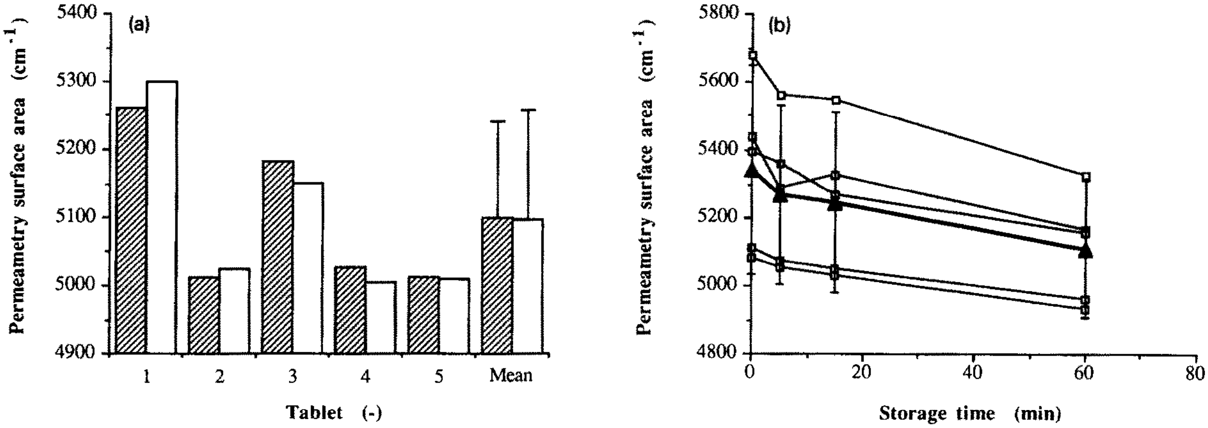

Fig. 1. Permeametry tablet surface area for sodium chloride tablets compacted of size fraction 20–40 μm. The tablets were compacted at 75 MPa. (a) (▨) Tablets stored at 0% relative humidity for 0 min; (□) tablets stored at 0% relative humidity for 15 min. (b) Tablets stored at 57% relative humidity for 60 min; (□) tablet surface area for individual tablets; (▲) calculated mean values of tablet surface area for five tablets and corresponding 95% confidence intervals.

A and B shows that the milling procedure tended to increase the degree of fragmentation of the particles during compression, which agrees with the findings of Wong and Pilpel (1990). A reasonable explanation is that the increased irregularity facilitates fracturing of particles. In addition, the milling procedure might induce defects, such as small cracks, in the particles which can initiate a fracture process and hence, promote particle fragmentation during compression.

3.2. Post-compaction changes in compact surface area

As a general measure of the pore structure of the compact, the air permeability of the compacts, recalculated as tablet surface areas, was determined. The sensitivity of such a method with respect to the detection of changes in the pore structure of powders and compacts is not defined. Furthermore, if the structure of the upper sur-

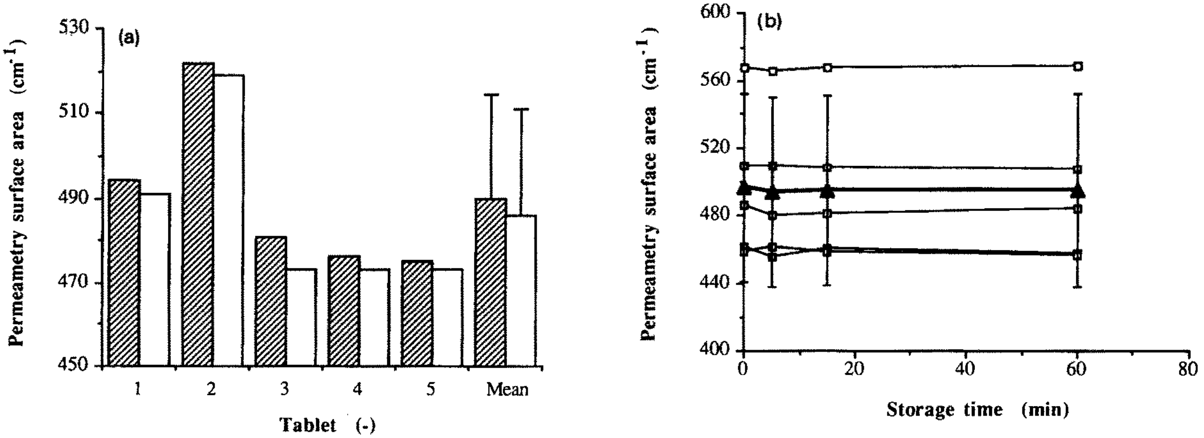

Fig. 2. Permeametry tablet surface area for sodium chloride tablets compacted of size fraction 425–500 μm. The tablets were compacted at 100 MPa. (a) (▨) Tablets stored at 0% relative humidity for 0 min; (□) tablets stored at 0% relative humidity for 15 min. (b) Tablets stored at 57% relative humidity for 60 min; (□) tablet surface area for individual tablets; (▲) calculated mean values of tablets surface area for five tablets and corresponding 95% confidence intervals.

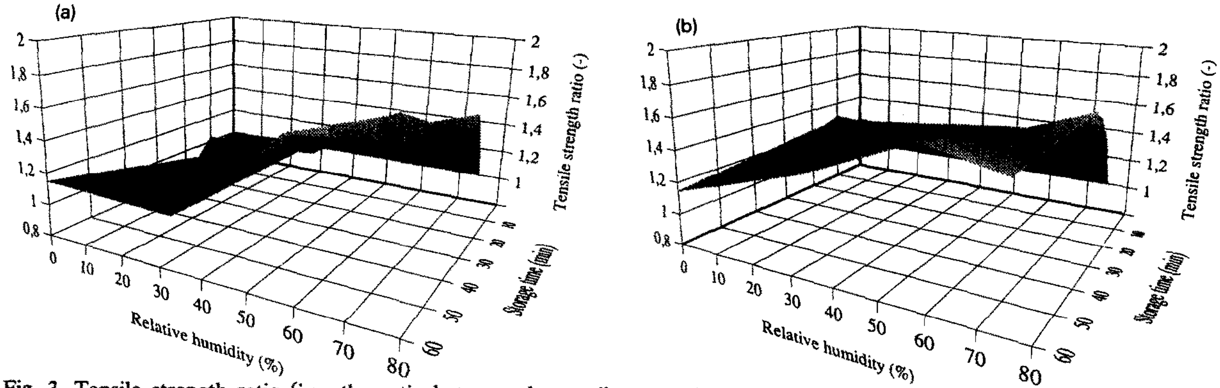

Fig. 3. Tensile strength ratio (i.e., the ratio between the tensile strength measured after storage and measured directly after compaction) for sodium chloride tablets as a function of storage conditions (time and relative humidity). The tablets were compacted at 150 MPa. (a) Size fraction 20–40 μm; (b) size fraction 40–60 μm.

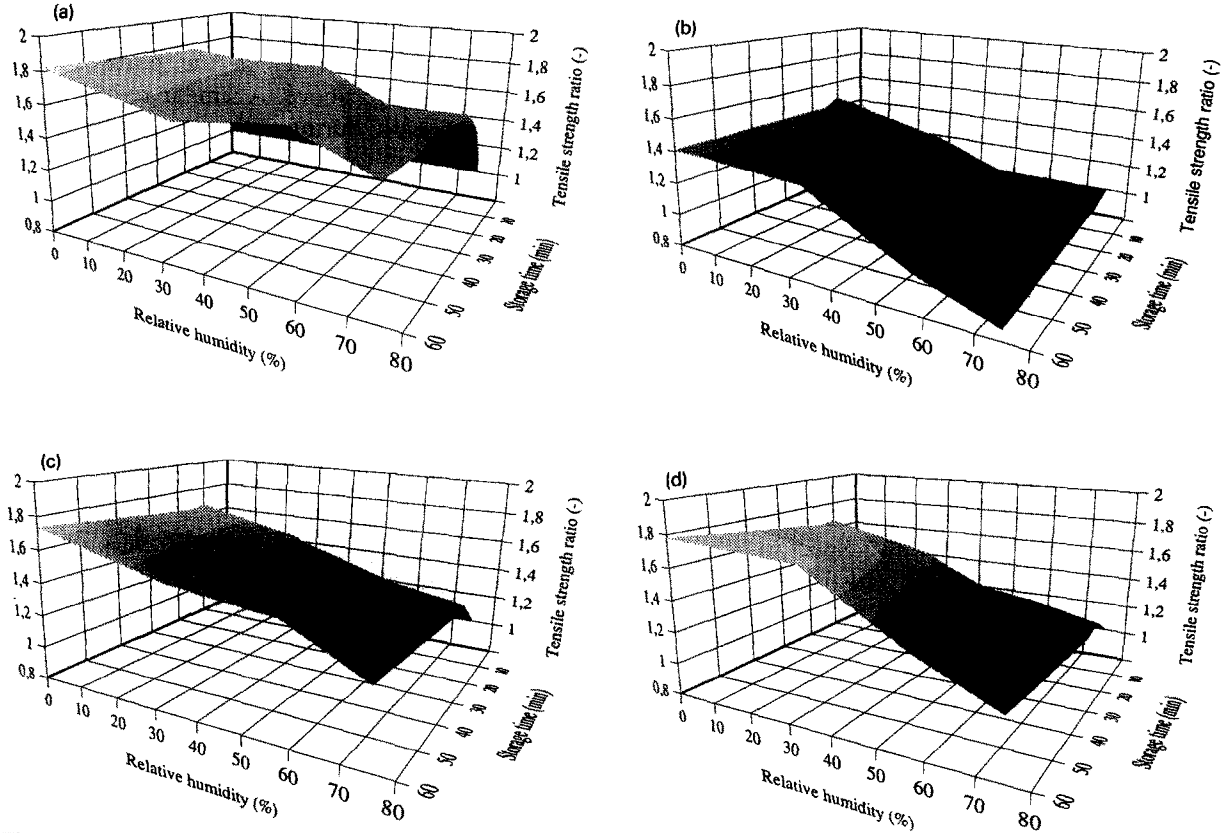

Fig. 4. Tensile strength ratio (i.e., the ratio between the tensile strength measured after storage and measured directly after compaction) for sodium chloride tablets as a function of storage conditions (time and relative humidity). The tablets were compacted at 150 MPa. (a) Size fraction 90–125 μm; (b) size fraction 212–300A μm; (c) size fraction 212–300B μm; (d) size fraction 425–500 μm.

face of the compact is atypical compared to the structure of the interior of the compact (Train and Hersey, 1960), an altered physical structure of the interior of the compact might occur which is not detectable with the permeametry technique. However, due to the need to assess the pore structure during a relatively short period of time directly connected to the compaction process, methods which require a sample preparation procedure, such as gas adsorption and mercury intrusion techniques, could not be used.

Changes in the tablet surface area with storage time were followed for compacts of two of the size fractions, 20–40 and 425–500 μm (i.e., the two extreme fractions with respect to particle size) under storage conditions of 0 and 57% relative humidity. The storage times were 15 and 60 min, respectively, since major changes in the tensile strength occurred during these periods (Table 2 and 3).

No detectable changes in the pore structure during storage of compacts of the coarse fraction, stored at either humidity, and for compacts of the fine fraction, stored at 0% relative humidity, were obtained by air permeability analysis (Fig. 1 and 2). Thus, it seems that the physical structure of these compacts is more or less unchanged after the compaction process, although very small changes in the relative positions of particle surfaces within the compact might occur, which are not detectable with the permeability technique.

For tablets of the fine fraction stored at 57% relative humidity (Fig. 1b), there is a tendency for the compact surface area to decrease continuously during the storage period. The observed changes in surface area (with respect to the calculated mean values) are not statistically significant, but there is a similar trend for each individual compact supporting the view that the decreased surface area corresponds to a real change in the physical character of the compact. This tendency is also supported by its agreement with earlier observations on compacts formed of fine particulate sucrose and sodium chloride (Ahlneck and Alderborn, 1989; Alderborn and Ahlneck, 1991). Thus, the results indicate that during storage of these compacts at 57% relative humidity, the compact pore structure, i.e., the geometry of the pores in terms their size and tortuosity, has changed slightly, which is manifested as an increased air permeability.

3.3. Post-compaction changes in compact tensile strength

For compacts prepared of the two finest size fractions, the tensile strength seemed not to change during storage at 0% relative humidity (Tables 2 and 3), as also found by Elamin et al. (1994). However, in humid atmospheres, the tensile strength increased during the storage period. For tablets stored at 33 and 57% relative humidity, the increase continued over the 4 day storage period, while for tablets stored at 75% relative humidity, the tensile strength reached a peak level and decreased thereafter. This reduction in compact strength after a certain storage time is probably a result of multilayer sorption followed by condensation of water in the pores of the tablet, as discussed above. A similar reduction in compact strength during storage at the critical relative humidity for sodium chloride (i.e., 75%) was valid for compacts of all size fractions used in this study.

In Fig. 3, the relationship between the post-compaction changes in tablet strength (expressed as the ratio between the tensile strength mea-

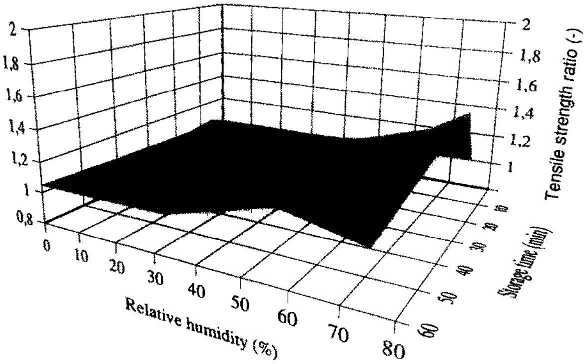

Fig. 5. Tensile strength ratio (i.e., the ratio between the tensile strength measured after storage and measured directly after compaction) as a function of storage conditions (time and relative humidity). Sodium chloride tablets compacted of size fraction 90–125 μm. The tablets were compacted at 75 MPa.

sured after storage and measured directly after compaction), the storage time and the relative humidity during the storage is presented. Compacts of both 20–40 and 40–60 μm particles showed a qualitatively similar relationship between those variables, thus the tendency is that a process causing the tablet strength to increase with time is initiated, when water from the environment is sorbed by the material.

For compacts of the coarser particles, another pattern with respect to the relationship between compact strength, storage time and storage relative humidity was obtained (Fig. 4). The compact strength increased during storage independently of the relative humidity of the environment provided that the humidity was below the critical relative humidity. Furthermore, during the first 60 min of storage, the increase in compact strength was most dramatic at 0% relative humidity and an increased relative humidity reduced the magnitude of the increase (Fig. 4). However, when the compacts were stored for a longer time, up to 4 days and below 75% relative humidity, the compact strength reached similar values which were independent of the storage humidity (Fig. 6 and Tables 2 and 3). Thus, for the four coarsest size fractions of sodium chloride, sorbed water is not a prerequisite for a post-compaction increase in compact strength to occur. On the contrary, instead of promoting a process causing the for-

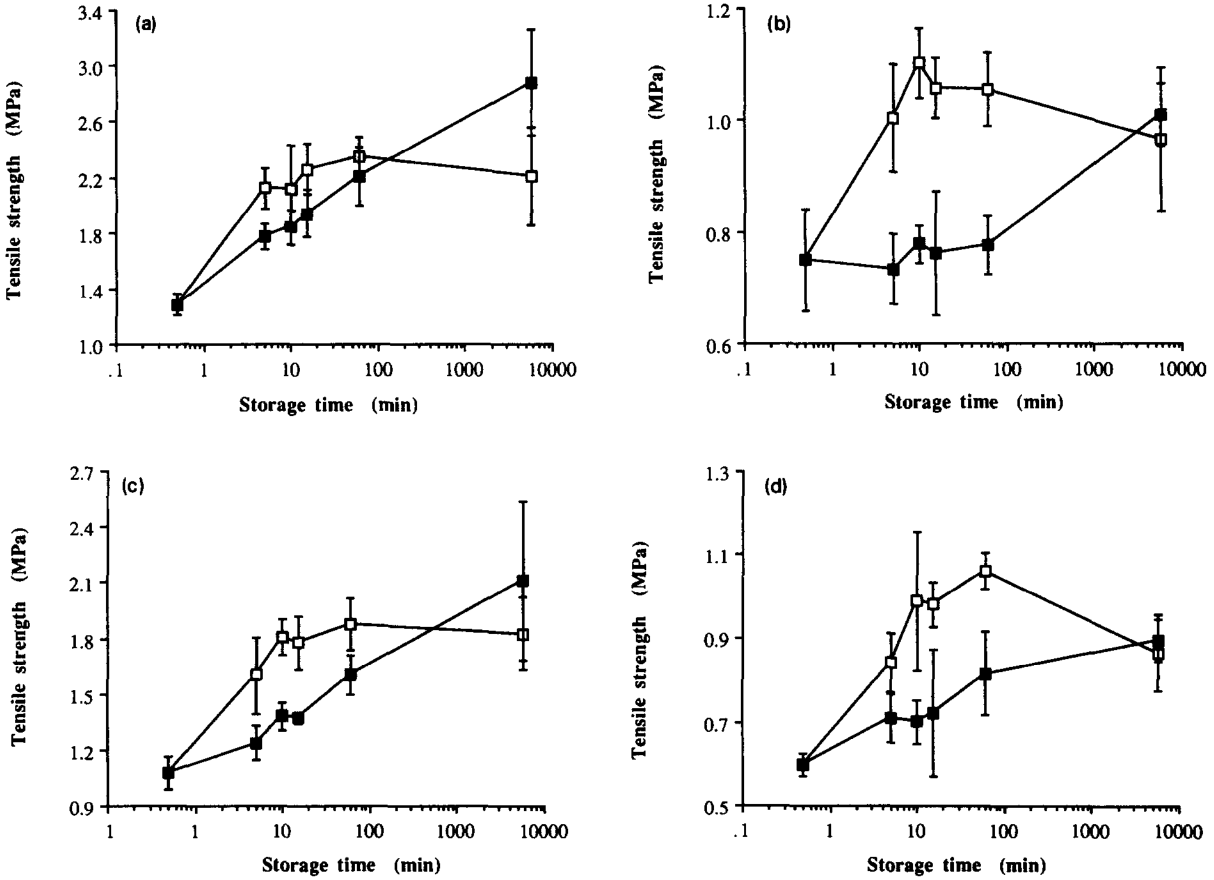

Fig. 6. Tensile strength of sodium chloride tablets as a function of storage time for tablets stored at different relative humidities. The tablets were compacted at 150 MPa. Tablets stored at (□) 0% relative humidity and (■) 57% relative humidity. (a) Size fraction 90–125 μm; (b) size fraction 212–300A μm; (c) size fraction 212–300B μm; (d) size fraction 425–500 μm.

mation of interparticulate bonds, sorbed water seems to slow down the tablet strength increase, although the final compact strength was similar.

Thus, a comparison between the graphs in Fig. 3 and 4 indicates that for compacts of sodium chloride, the increase in tablet strength during storage of compacts can occur by two different mechanisms. This is further supported by the results obtained from the studies on post-compaction strength changes during storage of the compacts, which involved transfers of compacts between atmospheres of different relative humidities (Fig. 7 and 8). In the discussion below, these mechanisms will be refered to as mechanisms α and β.

For compacts of the fine particulate sodium chloride (20–40 μm) (Fig. 7), a transfer of compacts from 0 to 57% relative humidity after 4 days of storage initiated a process resulting in an increase in compact strength. During this second storage period of 4 days, a compact strength was reached which was similar to the compact strength after a single storage period of 4 days (i.e., directly after compaction) at 57% relative humidity. Thus, the results support the observation that for this material, the presence of water in the atmosphere which surrounds the compact is necessary in order to initiate a process which causes the tablet strength to increase. However, the potential of the material to form new interparticulate bonds is preserved in the compact during storage at a low relative humidity, i.e., a bond formation process can be time-delayed and initiated when the material is exposed to water a certain time period after the formation of the compact.

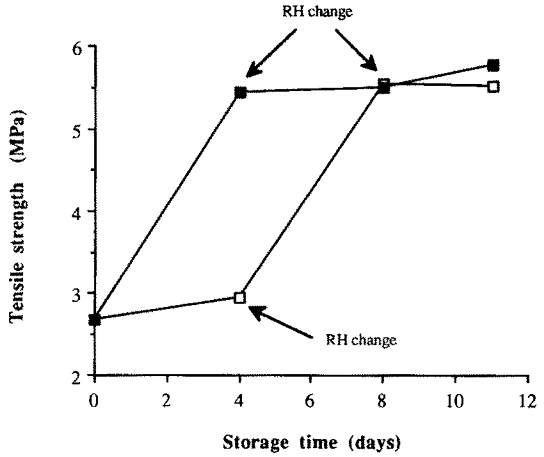

Fig. 7. Tensile strength of sodium chloride tablets as a function of storage time for tablets transferred between different storage conditions. The tablets were compacted of size fraction 20–40 μm at 150 MPa and the tablets were stored for a total of 11 days (i.e., 4+4+3). (□) Tablets transferred from 0 to 57 to 0% relative humidity; (■) tablets transferred from 57 to 0 to 57% relative humidity.

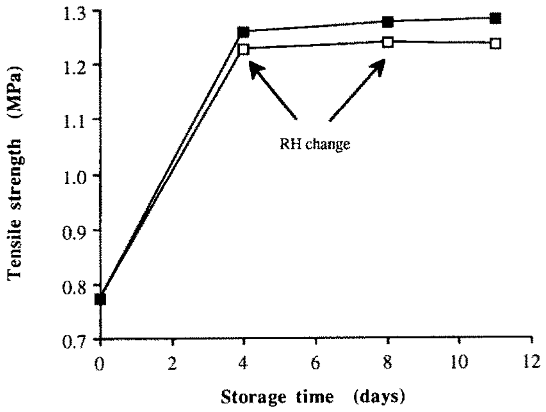

Fig. 8. Tensile strength for sodium chloride tablets as a function of storage time for tablets transferred between different storage conditions. The tablets were compacted of size fraction 425–500 μm at 150 MPa and the tablets were stored for a total of 11 days (i.e., 4+4+3). (□) Tablets transferred from 0 to 57 to 0% relative humidity; (■) tablets transferred from 57 to 0 to 57% relative humidity.

Compacts of the 425–500 μm fraction (Fig. 8) showed a different pattern. During storage at both 0 and 57% relative humidity, the compact strength increased during the first 4 days of storage after the formation of the compact and thereafter reached a plateau, i.e., it was not possible to intitate a time-delayed process by the transfer of a compact between different atmospheres.

Generally, for each individual size fraction, the relationship between compact strength, storage time and storage condition was similar for both compaction pressures. An exception, however, occurs with compacts produced from the fraction 90–125 μm. At the higher compaction pressure (Fig. 4a), the material behaves in accordance with the other coarse particulate materials (Fig. 4b–d), while at the lower compaction pressure (Fig. 5), the material conforms with the relationship for the fine particulate materials (Fig. 3). It seems thus that there exists a compaction pressure related threshold which affects the physical structure of the compact in such a way that a mechanistic change occurs in the post-compaction bond formation process.

4. Discussion

4.1. Suggested bond formation process for post-compaction strength increase: mechanism α

The interparticulate bond formation process denoted mechanism α seems to be characterised by the following features:

(i) The bond formation process is initiated by a change in the properties of the material, e.g., due to sorption of water or to a change in temperature.

(ii) Bonds are formed parallel to a small change in the physical structure of the compact, involving a change in the geometry of the pore structure.

(iii) When sorbed water initiates the bond formation process, an increased relative humidity of the environment, and thus an increased amount of water sorbed at particle surfaces, increases the rate of bond formation and perhaps also the total compact strength increase. Consequently, a relative humidity of the environment close to the critical relative humidity of the material probably results in the fastest and most marked compact strength increase.

This bond formation process has earlier been discussed in some detail in a series of papers from this laboratory (Ahlneck and Alderborn, 1989; Alderborn and Ahlneck, 1991; Elamin et al., 1994) and can be summarised as a diffusion-like transport of molecules or ions at the surface of particles, followed by a localisation of material at zones where particle surfaces are close to each other, i.e., in connection to bonds formed during compression. The consequence will be the formation of new bonds, either solid bridges or intermolecular attraction forces. A requirement for such a bond formation process is thus that the ions at the particle surfaces possess a certain mobility. The driving force for the localisation of ions at bonding sites already created during the compression process must be the release of energy as a consequence of the development of stronger bonds. It has been suggested (Elamin et al., 1994) that a disordering of the surface material in combination with water absorbed into the disordered material is necessary in order to reach a sufficient mobility of ions/molecules so that a rearrangement of material can occur. A sufficient mobility might be reached as a consequence of the transition of the disordered, amorphous material from a glassy to a rubbery state. The bond formation process will thus occur parallel to a change in the solid structure of the surface material, i.e., crystallisation. It has also been suggested (Elamin et al., 1994) that some materials might posses such a mobility in the disordered state although water is not present in the material, i.e. the material has a glass transition temperature below room temperature. Thus, a bond formation process can occur in the dry material. The compact strength increase will, in such a case, occur in direct connection to the formation of the compact and will not be initiated by exposure to a humid atmosphere.

It is suggested that compacts of the two finest size fractions of sodium chloride increase in strength entirely in accordance with mechanism α. These size fractions were milled before compaction and can thus be expected to be disordered at the particle surfaces. However, compacts of the unmilled 90–125 μm fraction prepared at 75 MPa behaved like compacts of the smaller size fractions. It is possible that surfaces of particles can be disordered locally during the actual compression phase, e.g., due to friction at interparticulate contact points. If a disordered material structure is a prerequisite for the initiation of a bond formation process by repositioning of ions at particle surfaces, it seems that the mechanical treatment during compression is sufficient to induce such changes in the solid state.

The milled particles 212–300 μm B represent coarse particles which are also milled before the compression phase. However, due to the increased compact strength measured directly after compaction, compared to compacts of the unmilled fraction 212–300 μm A, it is difficult to know whether or not the milled particles possess a more marked post-compaction strength increase when exposed to moist air.

4.2. Suggested bond formation processes for post-compaction strength increase: mechanism β

The results discussed above indicate that the interparticulate bond formation process denoted mechanism β is characterised by the following features:
(i) The bond formation process occurs in direct connection to the formation of the compact.
(ii) Bonds are formed without a change or with a minute change in the physical structure of the compact, i.e., not detectable with a permeametry technique.
(iii) An increased relative humidity of the environment, and thus an increased amount of water sorbed at particle surfaces, seems to slow down the compact strength increase. Consequently, a dry environment probably results in the fastest compact strength increase. However, the total compact strength can also be significant in relatively humid air or even independent of the relative humidity.

In addition, Karehill and Nyström (1990b) have earlier shown that the formation of a lubricant (magnesium stearate) film around coarse sodium chloride particles before compaction, reduces the total compact strength increase during post-compaction storage of tablets.

A possible underlying process for the post-compaction strength increase denoted mechanism β is the formation of new interparticulate bonds as a result of a change or adjustment in the relative positions of particle surfaces in the compact. This adjustment of surfaces can be caused by a continuing, time-dependent deformation of particles, after ejection from the die. Thus, distances between particle surfaces can decrease and interparticulate bonds can be formed. Although the distances between particle surfaces are changed, the effect on the pore structure of the compact is limited. In order to explain the effect of the magnesium stearate addition, it seems reasonable to assume that the bonds formed are to a large extent intermolecular attraction forces, i.e., interparticular bonds which act over a certain distance and can be disturbed by a change of the composition of the particle surfaces.

Possible prerequisites for a visco-elastic deformation of particles are firstly, that the particles in the compact exhibit internal stresses after compaction which can force the particles to deform and, secondly, that the particles are not fixed so rigidly to each other that a repositioning of surfaces is excluded. An increased compaction pressure, which corresponds to a reduced porosity, increased the post-compaction strength change (Tables 2 and 3). Furthermore, for the 90–125 μm particles, compaction at the higher pressure used in this study changed the behaviour of the compacts in such a way that a significant compact strength increase occurred at 0% relative humidity. A possible explanation is that an applied stress threshold during the compression process must be exceeded in order to induce such an internal stress state in the particles that a visco-elastic deformation will occur. The results indicate that a reduced particle size will increase this applied stress threshold.

An alternative process which can explain the post-compaction strength increase denoted mechanism β is a stabilisation of bonds which have been formed already during the compaction process. During compression, high temperature zones may be formed at interparticulate contact points as a result of interparticulate friction as well as bond formation (Pilpel et al., 1991). The temperature increase can soften the solid material and thus increase the mobility of the ions/molecules. It has even been suggested (Jayasinghe et al., 1969/70) that the solid material can melt as a result of increased local temperatures. Nevertheless, the consequence might be that a continuous phase of material will be formed at the interparticulate junctions, i.e., solid bridges. After compaction, the temperature of these solid bridges will decrease when the heat is conducted through the material and finally transferred to the gas phase. A rise in temperature of materials when compacted has recently been shown by means of infrared thermography of compacts as well as a decrease in compact temperature during a post-compaction storage period of some minutes (Bechard and Down, 1992; Ketolainen et al., 1993). The consequence of the transfer of heat from the hot spots will be a reduction in the temperature of the solid bridges. This reduction in temperature can have different effects on the structure of the material but it is possible that an increased strength of the solid bridge will be obtained with consequent increased tablet strength. Such a post compaction stabilisation of the bonds already formed during compression does not require any change in the physical structure of the compact. Also in this case, a lower applied stress limit might exist which must be exceeded in order to create solid bridges during compression. It cannot be excluded that both of the proposed processes discussed under this heading can occur simultaneously and affect the post-compaction compact strength increase.

The effect of the humidity of the environment on the compact strength increase, both its rate and its total amount, is not easily explained for the coarse particulate materials. The first possibility is that the adsorption of water from the environment will affect and slow down the rate of the processes discussed under this heading (i.e., bond formation or bond stabilisation), because water is initially sorbed from the humid air and thereafter desorbed during bond formation or bond stabilisation. A second possibility is that the exposure of the compact to humid air will initiate an α-type bond formation process, i.e., rearrangement of surface material. The potential for this process to occur also in compacts of unmilled materials was discussed above. Since the total compact strength increase during storage was similar under all storage conditions but the rate of increase seemed to be slower, this explanation assumes that there is a shift in the processes responsible for the compact strength increase, i.e., with an increased incidence of bond formation by mechanism α, a reduced incidence of bond formation by mechanism β will occur. It seems reasonable that if the potential for particles to deform visco-elastically exists, adsorbed water will not reduce the degree of visco-elastic deformation of the particles. The consequence must then be that the particle deformation will not result in the formation of bonds when water is present at the particle surfaces. A possible reason for this is that adsorbed moisture reduces the bonding force of intermolecular attractions. However, no compact strength increase was observed in this study when compacts were transferred from 57 to 0% relative humidity, i.e., a change in storage condition which facilitates desorption of water. Thus, a significant effect of adsorbed water on the bonding force is questionable. Furthermore, it is difficult to accept that a bond stabilisation process as discussed in this paper will cease in the presence of a humid atmosphere. It is therefore suggested that the role of water is to slow down a bond formation process by visco-elastic deformation or a bond stabilisation process.

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