ELSEVIER International Journal of Pharmaceutics 219 (2001) 113–126

international journal of pharmaceutics

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Drying behaviour of two sets of microcrystalline cellulose pellets

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Received 22 September 2000; received in revised form 26 February 2001; accepted 27 February 2001

Abstract

The objective was to study contraction and densification of two sets of microcrystalline cellulose pellets, prepared using water (W) or a 25/75% w/w water/ethanol (W/E) mixture, during drying. The pellets were dried on microscope slides, photographed and weighed at set times. The porosity of the dry pellets was determined by mercury pycnometry. From pellet size, weight and porosity data, contraction and densification of the pellets and the relationship of these to the liquid content of the pellets during drying were calculated. Both types of pellets contracted and densified during drying. The initial porosity was similar for both types, but the final porosity of the dry pellets was higher for the W/E pellets. Thus, the difference in final pellet porosity between the two types was caused by a difference in densification during drying rather than a different degree of densification during the pelletisation procedure. The contraction rate and the relationships between contraction and the volume of removed liquid, and contraction and the degree of liquid saturation differed between the two types of pellet. The difference in drying behaviour between the two types of pellets can be explained by a liquid related change in both contraction driving force and contraction counteracting force or by a different contraction mechanism. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Microcrystalline cellulose pellets; Granulation; Drying; Contraction; Densification; Porosity

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1. Introduction

Granules prepared from fine drug particles are used frequently in the production of pharmaceutical preparations because of their suitable processing properties, such as packing, flow, coating and tabletting properties. For some period of time, the problem of how to engineer granules with respect to their tabletting behaviour has been studied in our laboratory (Wikberg and Alderborn, 1993; Johansson et al., 1995; Nicklasson and Alderborn, 2000). In this context it has been found that irrespective of the composition of the granules, the intragranular porosity is a significant factor for their compression shear strength, compressibility and compactability. The granules used in our studies were prepared by forming granules by

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PII: S0378-5173(01)00636-6

the aid of a liquid which is subsequently removed by drying. The procedures thus used, i.e. wet granulation in convective mixers alone or in combination with a spheronisation process, can be described as the main or standard procedure for preparation of granules of various shapes in pharmaceutical production. One approach used in our laboratory to accomplish a variation in granule porosity has been to vary the ratio of ethanol to water of the granulation liquid.

The influence of different variables during the wet granulation process on granule properties, such as porosity, has been studied extensively in the literature. It appears that, during agitation in a convective mixer, the granules will densify (Kristensen et al., 1984) parallel to their growth by coalescence. The mechanical treatment during agitation, such as impeller speed (Holm et al., 1983), as well as the amount of granulation liquid added (Tapper and Lindberg, 1986), appear to affect the densification of the granules during their formation. It has also been suggested (Wells and Walker, 1983; Ritala et al., 1988) that the densification of the granules might be affected by the surface tension of the granulation liquid. This might explain the effect of the ethanol/water ratio of the granulation liquid on the porosity of the dried granules. However, the granules might densify further during the drying phase which thus can affect the final porosity of the dry granules. This is important, because discussions on granule densification during wet granulation are often based on measurements of the porosity of dried granules (Schaefer, 1996). It seems that the question of the relative importance of the granulation step and the subsequent drying step for the final granule porosity has not been addressed in the literature. This study represents a contribution in this context.

It has often been observed that the drying of porous plastic bodies, formed from a solid and water, is associated with the contraction of the body and even fracturing. It has been suggested that this contraction is driven by a compressive force developed due to, e.g. capillary and osmotic forces in the liquid in the pores of the material (Newitt and Coleman, 1952; Scherer, 1990; Hasatani et al., 1993). The consequence is that the primary particles forming the granule are pulled together and the granule shrink. It has also been proposed that most of the contraction occurs during the constant rate period of drying and, normally, the reduction in volume of the granule equates the volume of removed liquid (Scherer, 1990).

Kleinebudde (1994) has shown that also pharmaceutical pellets, containing drug and celluloses and with water as granulation liquid, contract considerably during drying in an oven or in a fluidized bed. In contrast, contraction was limited when drying took place in a freeze dryer, and the freeze-dried pellets were considerably more porous than those dried in the convective dryers. In addition to this article, it has also been reported that the drying conditions in a broad sense are of significant importance for the physical properties of pharmaceutical granular materials, such as their friability, bulk density (Remon and Schwartz, 1987), surface structure, mechanical strength (Bataille et al., 1993; Dyer et al., 1994), drug release properties (Dyer et al., 1994), porosity (Bataille et al., 1993; Kleinebudde, 1994; Habib and Shangraw, 1997; Kleinebudde and El Saleh, 1998) and compaction properties (Chatrath and Staniforth, 1990; Habib and Shangraw, 1997).

In most studies on contraction of different types of granules during drying, water has been used as agglomeration liquid. However, in pharmaceutical granulation, it is common to use other liquids as well, such as alcohol (Wells and Walker, 1983). It has been reported (Mitsyuk et al., 1964) that the final porosity of materials prepared by drying a gel was dependent on the type of alcohol in which the gels was soaked before drying. It is thus possible that differences in the porosity of granules prepared with different liquids (e.g. different mixtures of water and ethanol) (Wikberg and Alderborn, 1993; Johansson et al., 1995; Nicklasson and Alderborn, 2000) also can be explained, at least in part, by differences in contraction and densification during drying.

In conclusion, the phenomenon of contraction of granules during drying has seldom been discussed in the pharmaceutical literature as an explanation for differences in the physical and biopharmaceutical properties of granules. In this study, nearly spherical granules, hereafter referred to as pellets, of microcrystalline cellulose were prepared by wet granulation with two different liquids, water alone and a 25/75% w/w mixture of water and ethanol. These two liquids give dry pellets with markedly different porosity (Johansson et al., 1995). After agglomeration, the pellets were dried in air and the contraction and densification that occurred were determined. The aim of this study was thus to investigate the relative importance of the wet granulation and the drying phases on the porosity of microcrystalline cellulose pellets and to propose an explanation for the reported differences (Johansson et al., 1995) in porosity between these types of pellets.

2. Materials and methods

2.1. Preparation of pellets

Two sets of pellets were prepared by extrusion-spheronisation from microcrystalline cellulose (Avicel PH101, FMC, Ireland, with an apparent particle density of 1.58 g/cm^3). The preparation process was designed to produce pellets of acceptable sphericity. Water was used as agglomeration liquid for one set of pellets (W) and a 25/75% w/w mixture of water and ethanol (Finsprit 95%, Solveco Chemicals AB, Sweden) was used as agglomeration liquid for the other (W/E).

The powder (400 g) was dry mixed for 1 min at 500 rpm in a high shear mixer (model QII, Donsmark Process Technology, Denmark). The agglomeration liquid (in a proportion of 1.1 times the weight of the dry powder) was added by atomisation at a flow rate of 100 g/min followed by wet mixing for 1 min at 500 rpm.

The wet powder mass was extruded in a radial screen extruder (model E140, Nica System, Sweden) equipped with a 1.2 mm thick screen with circular openings of a diameter of 1.0 mm. The extrudate was spheronised for 3 min at 875 rpm on a radial plate spheroniser (model S320, Nica System, Sweden). The pellets were then immediately placed in closed containers and stored at room temperature.

2.2. Drying of pellets

In order to follow parallel changes in both volume and liquid content of the pellets during drying, some pellets were placed on microscope slides and dried by leaving the slides in the open atmosphere in the laboratory (about 20°C temperature and 40% relative humidity) without any forced convection.

Nine of the W pellets were placed in wells (diameter approximately 0.6 mm) in a specially constructed microscope slide (Fig. 1a). This preparation procedure took about 3 min. This slide was constructed so as to maintain a constant distance between individual pellets, which could be reproduced for each trial ($n = 3$). In addition, the microscope slide could be moved without changing the position of the pellets. At set intervals, the pellets were photographed in an optical light microscope (model Vanox, Olympus, Japan) at 1.3 times magnification and weighed on an analytical balance (model Micro, Sartorius, Germany) immediately after each photograph. Between each photographing/weighing session, the pellets were dried in the open atmosphere. Drying was continued until the weight of the pellets was constant with time. The pellet weight at this equilibrium point was considered to represent the solid mass.

The W/E pellets dried much more rapidly than the W pellets. In fact, substantial drying occurred during the first few minutes after pellet preparation which meant that the preparation procedure involving the slide with the wells could not be used. Instead, several hundred W/E pellets (approximately 600) were randomly placed on an ordinary microscope slide (Fig. 1c) and some of them (approximately 20) were photographed in the optical light microscope at 1.3 times magnification at set intervals ($n = 2$). Drying profiles were obtained by drying an approximately equal number of the W/E pellets on a microscope slide on an analytical balance (model AG245, Mettler Toledo, Switzerland) ($n = 3$).

2.3. Assessment of pellet volume and liquid content during drying

The volume of the pellets and their liquid content were calculated from the microscopy and weight data. The pellet volume (V_p) was calculated from the projected area diameter of the pellets, i.e. the diameter of a circle of equal projected area as the pellet (D_p), by assuming a perfect spherical shape, i.e. $V_p = \pi D_p^3/6$. The pro-

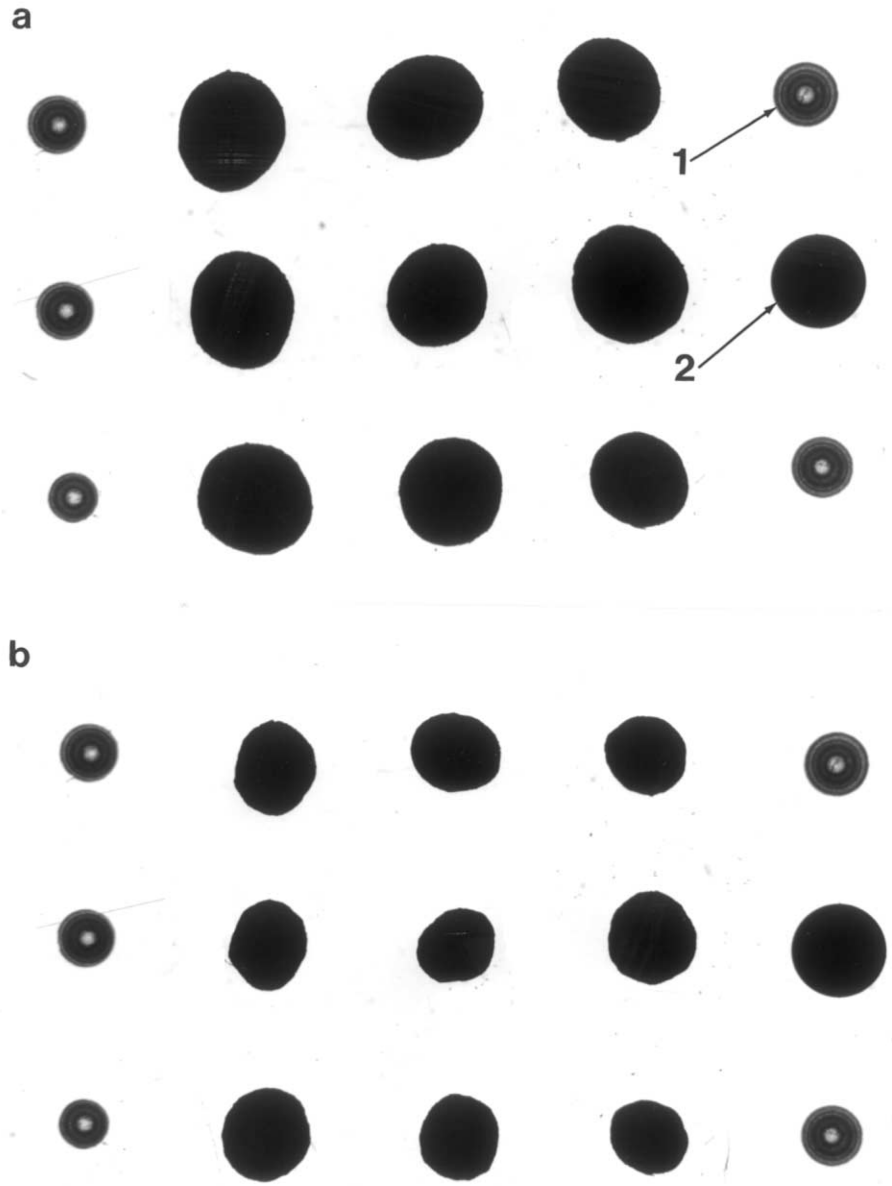

Fig. 1. Optical light photomicrographs at 1.3 times magnification of microcrystalline cellulose pellets before and after drying. (a) Pellets prepared with water, before drying. (b) Pellets prepared with water, after drying. (c) Pellets prepared with 25/75% w/w water/ethanol, before drying. (d) Pellets prepared with 25/75% w/w water/ethanol, after drying. (1) Well with a diameter of 0.6 mm. (2) Ball bearing ball of stainless steel (SKF, Sweden) with a diameter of 1000 μm.

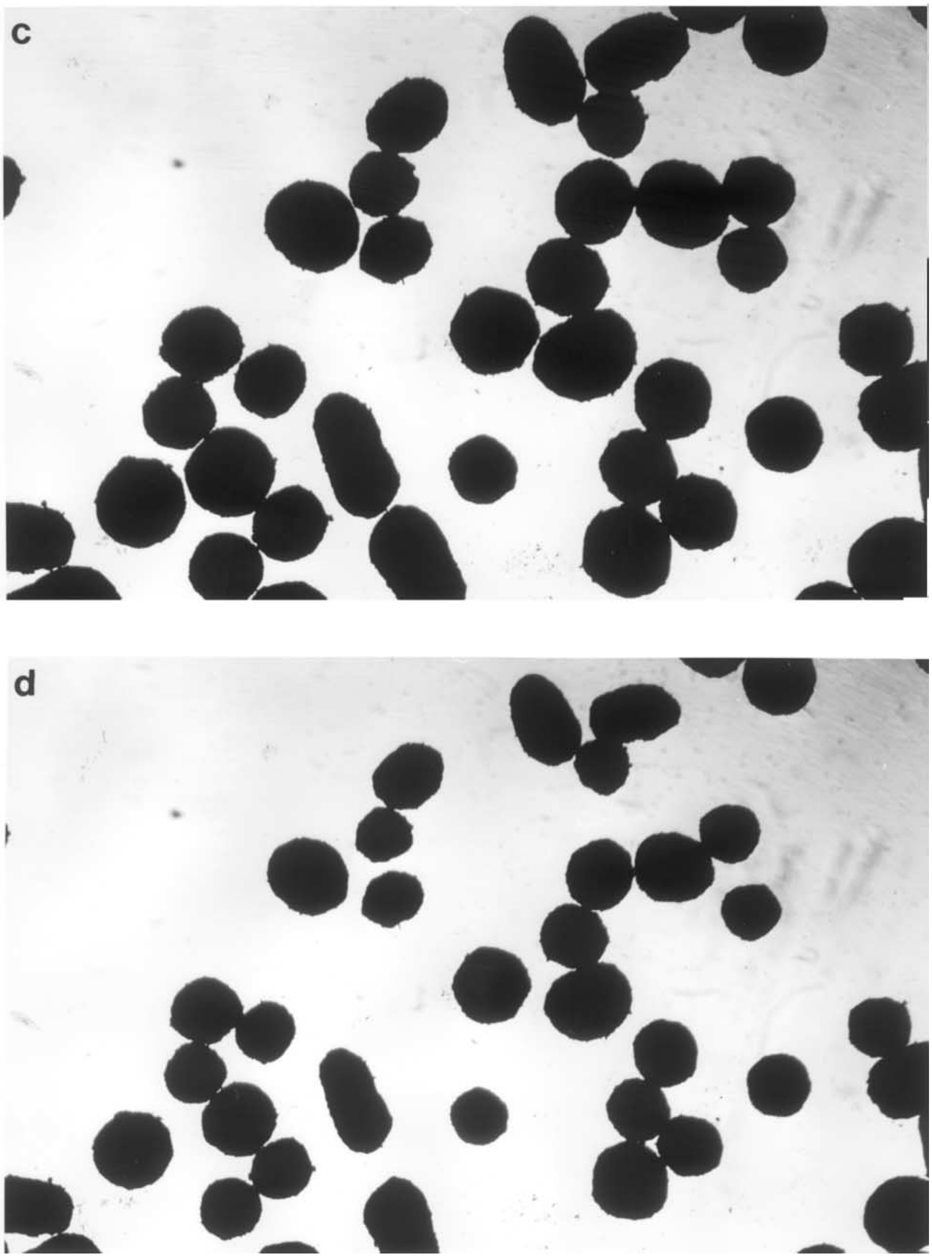

Fig. 1. (*Continued*)

jected area of the pellets was determined from digitalised microscopy photographs of the pellets (NIH Image 1.61, USA, available on the Internet at http://rsb.info.nih.gov/nih-image/). The liquid content of the pellets was calculated as the mass of liquid divided by the mass of solid and as the volume of liquid divided by the mass of solid (H_t).

2.4. Assessment of pellet porosity and pore size distribution after drying

After drying, all pellets were stored in a desiccator over a saturated solution of sodium iodide (corresponding to 40% relative humidity at 20°C (Nyqvist, 1983)) for at least 10 days before determination of the final pellet porosity. The pellet porosity was calculated from the apparent particle density of the microcrystalline cellulose powder and the effective particle density of the pellets. The apparent particle density (British Standard 2955: 1958) of the microcrystalline cellulose particles was determined using a helium pycnometer (AccuPyc 1330 Pycnometer, Micromeritics, USA) ($n = 3$). The effective particle density (British Standard 2955: 1958) of the pellets was determined by mercury pycnometry at 101 kPa (equivalent to 760 mm Hg) using a mercury porosimeter (Autopore III, Micromeritics, USA). As a plateau was observed around this pressure in the cumulative volume of mercury intruded versus pressure graph it was concluded that the intergranular pores were filled at this point. In the mercury intrusion experiments a pressure range of 0.101–413 MPa was used. The pore openings were assumed to be circular and a surface tension and a contact angle of mercury of 480 mN/m and 130°, respectively, were used in the calculations ($n = 2$).

2.5. Assessment of porosity, contraction, densification and degree of liquid saturation of pellets during drying

The initial porosity and the porosity at different times during the drying process of each individual pellet (E_t) was calculated from the following equation:

$$\mathrm{E_t}\,(\%) = \left(1 - \left(\frac{V_{\mathrm{dry}} \times \rho_{\mathrm{e}}^{\mathrm{dry}}}{V_{\mathrm{t}} \times \rho_{\mathrm{a}}^{\mathrm{dry}}}\right)\right) \times 100 \tag{1}$$

where V_{dry} and V_{t} are the volumes of the dry pellet and the wet pellet (at time t), respectively (calculated from the projected area diameter). $\rho_{\mathrm{e}}^{\mathrm{dry}}$ and $\rho_{\mathrm{a}}^{\mathrm{dry}}$ are the effective particle density (from mercury pycnometry) of the dry pellet and the apparent particle density, respectively.

The contraction and densification of each pellet during the drying process were calculated as follows:

$$\text{Contraction}\,(\%) = \left(1 - \left(\frac{V_{\mathrm{t}}}{V_0}\right)\right) \times 100 \tag{2}$$

$$\text{Densification}\,(\%) = \left(1 - \left(\frac{E_{\mathrm{t}}}{E_0}\right)\right) \times 100 \tag{3}$$

where V_0 is the initial pellet volume and V_{t} is the pellet volume at time t. E_0 and E_{t} are the initial pellet porosity and the pellet porosity at time t, respectively (from Eq. (1)).

The ratio of the pore volume occupied by agglomeration liquid to the total volume of the pores, i.e. the degree of liquid saturation (Kristensen et al., 1984) S_{t}, was calculated for each individual pellet at set times during the drying process from the following equation:

$$\mathrm{S_t}\,(\%) = \left(H_{\mathrm{t}} \times \rho_{\mathrm{a}} \times \frac{1 - E_{\mathrm{t}}}{E_{\mathrm{t}}}\right) \times 100 \tag{4}$$

where H_{t} is the liquid content at time t (volume liquid/mass of solid), E_{t} is the porosity at time t (from Eq. (1)) and ρ_{a} is the apparent particle density. A density of 0.85 g/cm^3 was used for the ethanol/water mixture and 1.0 g/cm^3 for water.

3. Results

The preparation procedure gave nearly spherical pellets. The pellet shape did not appear to change during drying, as judged visually from the microscopy photographs (Fig. 1), which is consistent with earlier reported results (Kleinebudde, 1994). In no case, formation of cracks during the coarse of the drying was observed. The formation of cracks can be a problem in drying some agglomerated materials such as ceramics (Scherer, 1990). The surfaces of the W pellets were generally smoother than those of the W/E pellets.

Drying profiles for both types of pellet are shown in Fig. 2a. Drying was nearly complete within 30 min for both types of pellet. The liquid content (both initial and final) was similar for both types of pellet, but the W pellets had a higher initial and a greater final pellet volume than the W/E pellets

(Fig. 2b). Thus, the absolute reduction in pellet volume and the relative change in pellet volume (i.e. contraction) (Fig. 3a) were considerably higher for the W pellets.

The drying profiles (Fig. 2a) were nonlinear for both types of pellets. The rate of drying fell throughout the drying process, but with a gradually reduced falling rate with decreased liquid

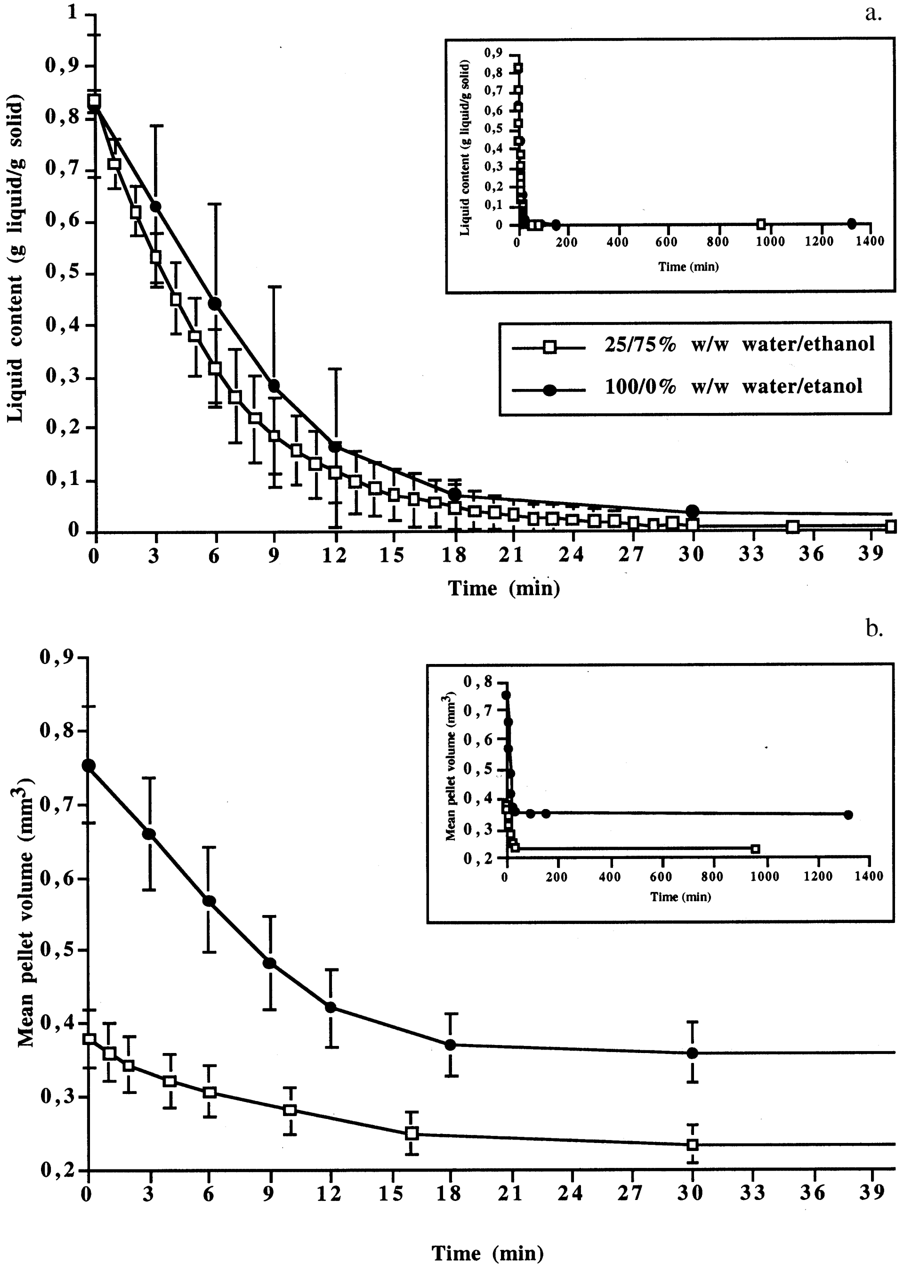

Fig. 2. (a) Drying profiles of monolayers of microcrystalline cellulose pellets prepared with water and with 25/75% w/w water/ethanol ($n = 3$). (b) Mean pellet volume versus time during drying ($n = 22$ for pellets with water and $n = 39$ for pellets with 25/75% w/w water/ethanol). Symbols are defined in the graph. Error bars show 95% confidence limits of arithmetic mean values.

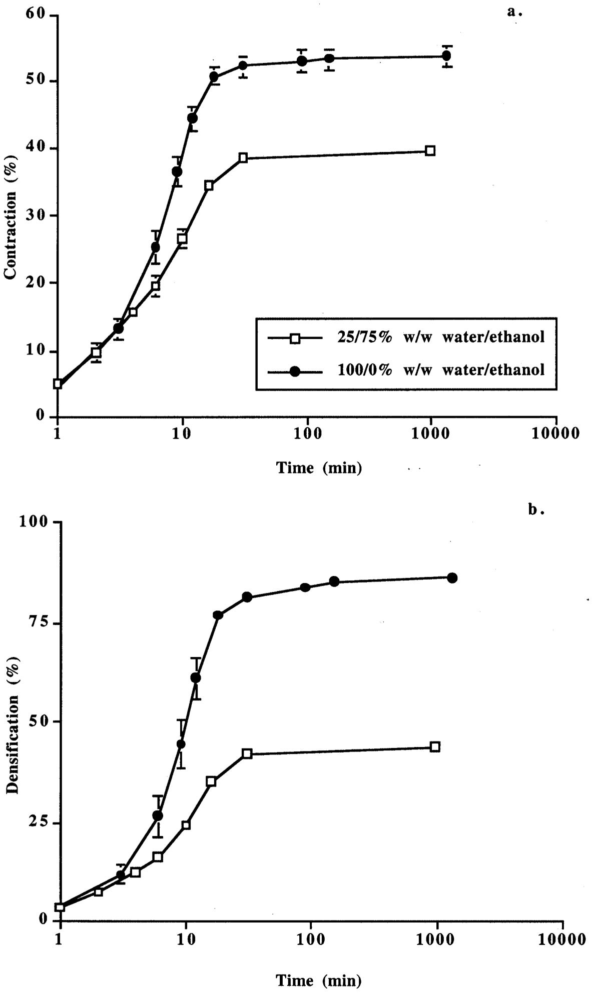

Fig. 3. (a) Contraction and (b) densification of the two pellet types during drying ($n = 22$ for pellets with water and $n = 39$ for pellets with 25/75% w/w water/ethanol). Error bars show 95% confidence limits of arithmetic mean values. Symbols are defined in the graph.

content. Similar type of curves (Fig. 2b) were also obtained for the relationship between pellet volume and time. The absence of a period when the rate of drying was constant (i.e. when the rate of drying per unit area of surface is independent of time) was possibly the result of reduction in the area from which liquid was evaporated during drying, as the pellets contracted (Fig. 3). Moreover, the drying profile is obtained from a number of pellets. It is possible that, while individual pellets may dry at a constant rate (which probably vary between the pellets), the sum of these individual relationships might be a curved profile.

Both pellet types contracted by a significant amount: 54 and 40% for the W and W/E pellets, respectively (Fig. 3a). As a consequence, the densification of the W and W/E pellets was 86 and 44%, respectively (Fig. 3b). Both the contraction and densification profiles as a function of time were sigmoidal in shape. While the initial porosity of the wet pellets was similar for both types (57.3 and 60.0%, respectively, for the W and W/E pellets), the final porosity was markedly different (7.85 and 33.8%, respectively; Table 1). The volume of intruded mercury into the pores of the W/E pellets was greater (Fig. 4) and the size of the pores were generally larger than of the W pellets.

The contraction of the cellulose pellets as a function of the moisture content is shown in Fig. 5. The slope of this graph (dC/dH) is the rate of contraction. The curves were non-linear for both pellet types during the drying process, but the rate of contraction was higher for the W pellets than for the W/E pellets between liquid content of 0.9 and 0.3.

For both pellet types, the cumulative decrease in pellet pore volume related linearly to the cumulative volume of liquid removed by drying (Fig. 6). The gradient of this relationship was close to unity for the W pellets (1.08, $r^2 = 0.99$) but significantly below unity for the W/E pellets (0.69, $r^2 = 0.98$). Consequently, the degree of liquid saturation was high (about 100%) during most of the drying process for the W pellets (Fig. 7) but

Table 1
Pellet porosity before and after drying[a]

	Pellets prepared with water	Pellets prepared with 25/75% w/w water/ethanol
Initial pellet porosity[b] (%)	57.3[c] (11)	60.0[d] (8.7)
Final pellet porosity[e] (%)	7.85 (0.39)	33.8 (0.83)

[a] Mean values. Range in parentheses.
[b] Calculated for each individual pellet from Eq. (1).
[c] $n = 22$.
[d] $n = 39$.
[e] Determined with mercury pycnometry ($n = 2$).

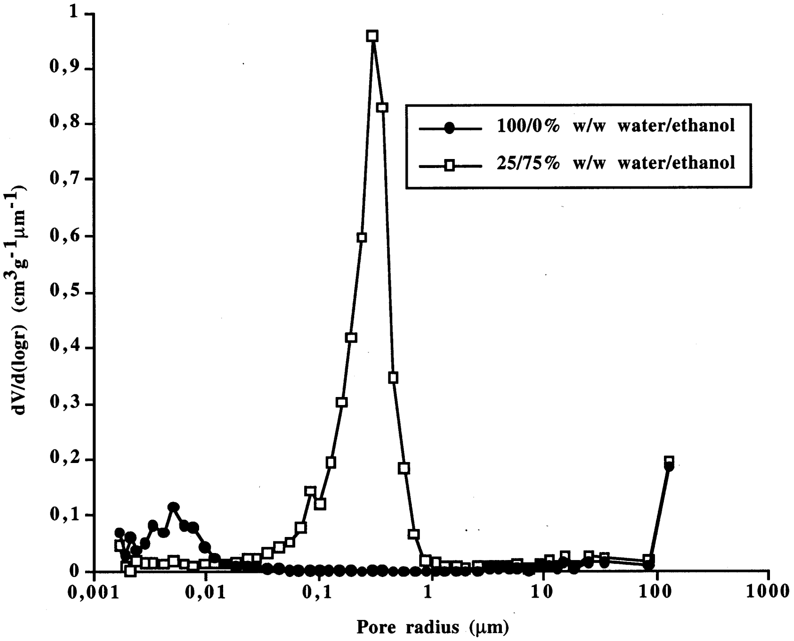

Fig. 4. Pore size distributions of the two pellet types ($n = 1$). Symbols are defined in the graph.

decreased continuously throughout the process for the W/E pellets. The pores in the granules, of both pellet types, were completely filled with liquid at the beginning of the drying process.

4. Discussion

As discussed above, the granulation of particles with a liquid involves parallel to the formation of clusters often also a densification of the granules which is affected by variables such as the intensity of the mechanical treatment and the properties of the powder and the granulation liquid. The mechanical treatment to which the pellets in this study was subjected during their preparation, densified both sets of pellets to a maximum, i.e. to a degree of liquid saturation of approximately 100% (Fig. 7). This is consistent with earlier findings on the behaviour of wet microcrystalline cellulose masses during ram extrusion (Jerwanska et al., 1995). The porosity of the wet pellets directly after preparation was therefore similar (Table 1). However, there was a marked difference in porosity between wet and dry pellets for both types and, as reported earlier (Johansson et al., 1995), the porosity of the dry pellets was highly dependent on the type of granulation liquid used (Table 1). It can thus be concluded that pellets of microcrystalline cellulose, prepared by wet granulation with water and a 25/75% w/w mixture of water and ethanol, contract and densify considerably during nonagitated convective drying. Furthermore, the difference in the final dry porosity between the two types is the result of a difference in densification (caused by contraction) during drying rather than a difference in the degree of densification during the granulation procedure. The drying behaviour of these pellets will hence be of decisive importance for their functional tabletting behaviour. A closer investigation of the effect of the type of granulation liquid used on the drying behaviour of the pellets was thus performed. Contraction occurred throughout the whole drying phase for both sets of pellets (Fig. 5), but was particularly apparent while the pellets retained a significant degree of liquid saturation (Fig. 7). However, the drying behaviour differed between the two sets of pellets in terms of the contraction rate (Fig. 5), the relationship between contraction and the volume of removed liquid (Fig. 6) and the degree of liquid saturation during drying (Fig. 7).

For the pellets prepared with water, the volume of contraction was approximately equal to the volume of removed liquid, meaning that the degree of liquid saturation of the pellets remained at nearly 100% during a considerable fraction of the drying time. Thus, the drying front was located at the exterior of the granules for a significant fraction of the drying period and the system strives to keep the solid pore surfaces wetted. For the pellets prepared with water and ethanol, in contrast, the volume of contraction was less than the volume of removed liquid, so the degree of liquid saturation fell during the drying phase. It would be reasonable to assume, therefore, that the drying front moved progressively towards the interior of the granules throughout the drying period. Also the rate of contraction differed between the two sets of pellets, i.e. the contraction rate was higher for the water system during a significant part of the drying phase. It thus seems that the difference in drying behaviour is established in the beginning of the drying phase, and is sustained throughout the drying process.

In a liquid filled granule, each solid particle can in a simplified way be described to be surrounded by a film of liquid which separates the particles from each other. Removal of this liquid film during drying allows the particles to approach each other, and the granule contracts, i.e. when the liquid film is gradually reduced in thickness, the interparticulate separation distance and granule volume will be reduced. The process of contraction can thus be described as a repositioning of particles in a similar way as the deformation of liquid filled agglomerates (Kristensen et al., 1985), i.e. granule contraction can thus be described as a flow of particles within a stagnant fluid. However, in order for this particle flow to occur, it is normally considered that the process must be driven by a contraction or compressive force which pull the particles together (Scherer, 1990). This driving force is considered to originate from pressure gradients in the liquid in the pores, such as capillary pressure and osmotic pressure (Newitt and Coleman, 1952; Scherer, 1990; Hasatani et

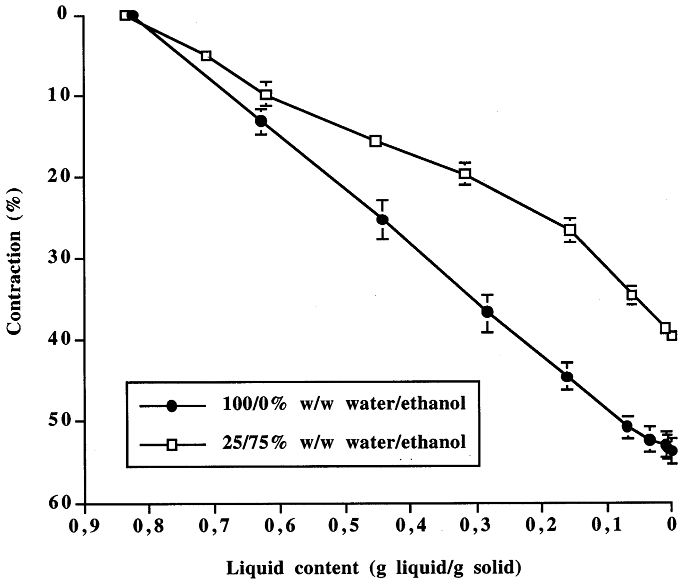

Fig. 5. Contraction as a function of liquid content of the two pellet types (for contraction; $n = 22$ for pellets with water and $n = 39$ for pellets with 25/75% w/w water/ethanol, and $n = 3$ for liquid content). Error bars show 95% confidence limits of arithmetic mean values. Symbols are defined in the graph.

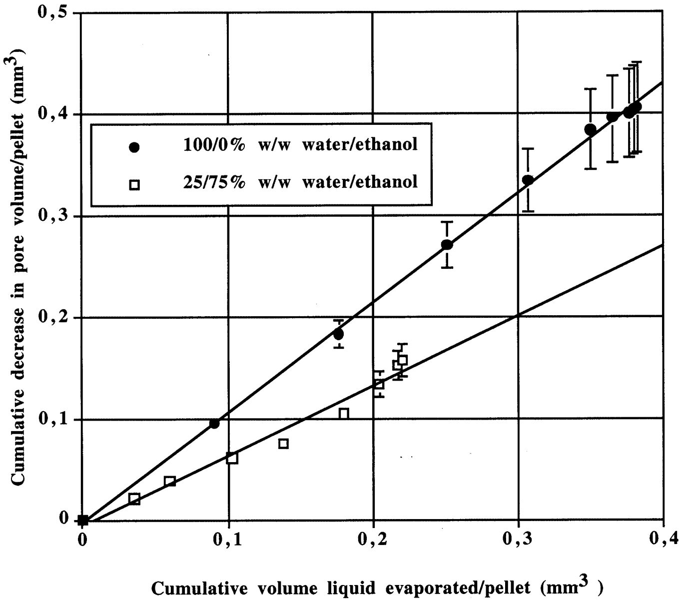

Fig. 6. Mean cumulative decrease in pore volume as a function of mean cumulative volume of liquid evaporated during drying for the two pellet types ($n = 22$ for pellets with water and $n = 39$ for pellets with 25/75% w/w water/ethanol). Error bars show 95% confidence limits of arithmetic mean values. Symbols are defined in the graph.

al., 1993). However, the flow of particles in a fluid can be counteracted by friction forces between the particles, i.e. the movement of particles and progressive reduction in interparticulate separation distance will be counteracted by the bonding between the particles. The drying behaviour, in terms of the localisation of the drying front and the contraction of the granules, will thus depend on the balance between the driving and the counteracting forces. Both these factors will probably differ between the two sets of pellets used in this study and can thus explain the different contraction behaviour during drying.

Since the pellets contained no water-soluble components, it is reasonable to assume that capillary pressure was the driving force behind the contraction in this study. It has been suggested (Scherer, 1990) that the maximum capillary pressure (P_R) of a liquid in a pore with a hydraulic radius of r_H can be assessed as follows:

$$P_R = \frac{\gamma_{SV} - \gamma_{SL}}{r_H}$$

where γ_{SV} and γ_{SL} are the solid/vapour and solid/liquid interfacial energies respectively. From the equation, often referred to as the Young's equation (Becher, 1965), $\gamma_{SV} - \gamma_{SL}$ can be expressed as γ_{LV} $\cos(\theta)$. Thus, θ is the contact angle between the liquid and the solid and γ_{LV} is the liquid/vapour interfacial energy (or surface tension). The change in granulation liquid from water to the water–alcohol mixture will change the interfacial energies of the system (Wells and Walker, 1983) and capillary forces of different magnitude can thus explain the differences in contraction between the two sets of pellets. However, the two liquids may also have different lubrication properties, which will affect the bonding and friction between the particles. It has been shown previously (Karehill and Nyström, 1990; Olsson et al., 1996) that the tensile strength of microcrystalline cellulose compacts is markedly affected by the presence of liquids in the pores of the compacts. In addition, the dielectric constant of the liquid affected the relative strength reduction caused by the presence of the liquid, i.e. the bonds between the primary particles are affected by the medium surrounding the particles. It is suggested that this effect can be extended to other agglomerates and the type of liquid present in the interparticulate pores will thus affect the strength of the agglomerate and the repositioning propensity of the primary particles.

Alternatively, the difference in contraction behaviour between the two types of pellet could be interpreted in terms of differences in the dominant contraction mechanism. It has been suggested (Caramella et al., 1984; Wan and Prasad, 1990) that microcrystalline cellulose particles swell during absorption of water. Consequently, the primary cellulose particles shrink during desorption. The whole granule might then contract as a result of this shrinkage of the cellulose particles. Desorption of absorbed water might occur when the water which surrounds the particles in the granule has evaporated and the particle surfaces become dry. This situation would only have developed during the latter part of the drying phase for the

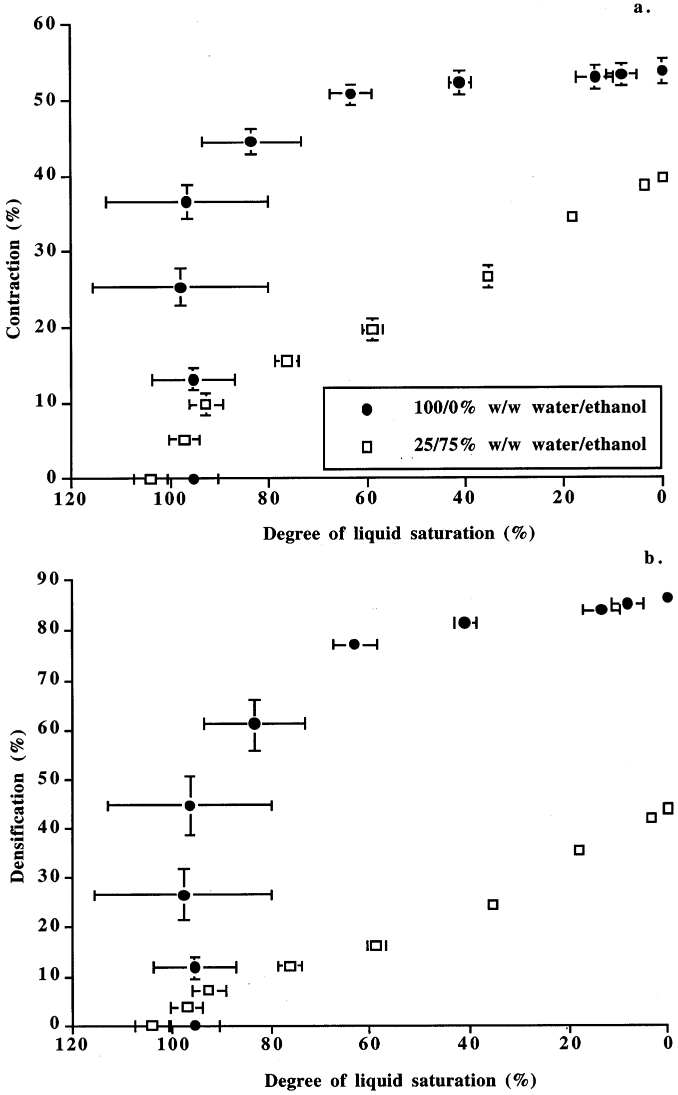

Fig. 7. (a) Contraction and (b) densification of the two pellet types as a function of degree of liquid saturation during drying ($n = 22$ for pellets with water and $n = 39$ for pellets with 25/75% w/w water/ethanol). Error bars show 95% confidence limits of arithmetic mean values. Symbols are defined in the graph.

W pellets, when the drying front moved towards the interior of the granules. Thus, contraction of the W pellets was probably only partly affected by the shrinkage of the primary cellulose particles. In contrast, the degree of liquid saturation was reduced early in the drying phase for the W/E pellets, which can be interpreted as a drying of cellulose particles surfaces and a movement of the drying front towards the interior of the pellets. Conditions enabling the shrinkage of the cellulose particles therefore existed during nearly the whole drying phase. The contraction of W/E pellets might thus be explained by an alternative mechanism, i.e. pellet contraction due to shrinkage of the cellulose primary particles.

5. Conclusions

From the experiments performed in this study, it is concluded that:

1. Pellets formed from microcrystalline cellulose and water and a 25/75% w/w mixture of water and ethanol contract (by 54 and 40%, respectively) and densify (by 86 and 44%, respectively) during static drying in air.
2. The initial porosity of the two types of pellet was similar, suggesting that the difference in final porosity was the result of a difference in densification (due to contraction) during drying rather than a difference in densification during the granulation procedure.
3. The drying behaviour differed considerably between the two types of pellets in terms of the contraction rate and the relationships between volume of contraction and the volume of removed liquid and contraction and the degree of liquid saturation of the pellets. The difference in contraction rate between the pellet types was sustained during a considerable fraction of the drying process.
4. The difference in contraction during drying and in drying behaviour between the two types of pellets can be explained by either liquid related changes in the contraction driving force (caused by pressure gradients in the liquid) and the contraction counteracting force (caused by friction between particles) or a different contraction mechanism (particle rearrangement due to pressure gradients in the liquid or primary particle shrinkage).

Acknowledgements

This study is part of a project financed by Pharmacia AB, AstraZeneca AB and NUTEK (Swedish National Board for Industrial and Technical Development).

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