J. Pharm. Pharmac., 1973, **25**, 729–735 *Received April* 24, 1973

The packing properties of moist bulk solids‡

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The specific volumes of three bulk solids at moisture contents up to 50% *w/w* (dry basis) under various consolidating stresses have been shown to be dependent on the nature of the bulk solid. The values obtained can be related to the intraparticulate porosity and inherent cohesiveness of the dry materials. The influence of moisture on the packing properties and tensile strength of the three bulk solids is discussed in terms of the granulation process.

Knowledge of the granulation of pharmaceutical and other bulk solids has largely been gained from a close examination of the processes in use (e.g. Fonner, Banker & Swarbrick, 1966; Ahmad & Pilpel, 1967; Harwood & Pilpel, 1968; Goodhart, Draper & Ninger, 1970; Woodruff & Nuessle, 1972). Many aspects of the massing and screening process have been considered including the influence of particle size of the ingredients (e.g. Hunter & Ganderton, 1972), moisture content of the moist granules (e.g. Ganderton & Hunter, 1971) and quantity and nature of the binder (e.g. Healey, Humphreys-Jones & Walters, 1972).

Bowl, fluidized bed and spray granulation methods have been treated in a similar manner and are reviewed by Pilpel (1969).

In addition to these process studies, information relevant to granulation is available from investigations based on the tensile strength of moist agglomerates. Rumpf (1958) and Pietsch, Hoffman & Rumpf (1969) provided equations for deriving the tensile strength of moist granules with wide ranges of moisture contents.

In previous work we have reported the effects on tensile strength of particle size (Eaves & Jones, 1972a), inherent cohesiveness of raw material (Eaves & Jones, 1972b) and surface tension of wetting fluid (Eaves & Jones, 1972c). This communication describes changes in the packing properties of various particulate materials with increasing moisture contents and discusses their implications, as well as those of concurrent tensile strength changes, in granulation processes.

MATERIALS AND METHODS

Materials

Three bulk solids with different physical characteristics have been used. Physical properties of the materials are given in Table 1 and Fig. 1. The materials are from the same batches as previously reported for tensile strength studies (Eaves & Jones, 1972b). The classification of materials in terms of porosity and cohesiveness (Table 1) is dependent on the magnitude of the intraparticulate porosity and tensile strength of the dry materials, respectively (Eaves & Jones 1972c).

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‡ Presented, in part, at the British Pharmaceutical Conference, Keele University, U.K., September 11–15, 1972.

Procedure

Samples of each material were dried in layers less than 5 mm thick at 120° for 40 h. They were then transferred hot to a temperature and humidity controlled cabinet (Eaves & Jones, 1970) and allowed to equilibrate at 28° and less than 25% relative humidity. Each material was then dredged into a cylindrical cell (approx. 5 cm diameter, 1 cm high, volume 21·0 ml) with a "consolidating ring" (height 2·2 cm) located on the cell rim. In this way the cell could be overfilled and the material consolidated by means of a weighted plunger inserted vertically into the consolidating ring. After applying a fixed normal load, the plunger was twisted backwards and forwards five times through about 10° and then turned through 90°. The twisting and turning was repeated four more times making a total of 25 twists and served the purpose of reducing inhomogeneities of bulk density within the bed. The plunger and consolidating ring were removed, each with a twisting action to prevent disruption of the bed in the cell which was then scraped level with the rim by means of a flat, sharp-edged, metal spatula.

By using various normal loads, the effect on packing properties of applying stresses over a range of 0–202 g cm^{-2} was investigated. The packing properties were determined after various periods of exposure to relative humidities up to 75% and after the addition of various percentages (by weight) of distilled water (to calcium phosphate and glass) or saturated potassium chloride solution at 28° (to potassium chloride).

From a knowledge of the weight, volume and loss on drying of the cell contents, the specific volume of the sample was calculated as follows:—

$$\text{Specific Volume, } S = \frac{V}{Wu}$$

where V = volume of cell; Wu = weight of dry, undissolved solid in the cell.
The calcium phosphate and glass powder were assumed to be completely insoluble

$$\text{Thus } Wu = \frac{100Ww}{100 + b}$$

Where Ww = Weight of moist sample; b = % w/w loss on drying (dry basis).

The moisture in the potassium chloride was assumed to exist as a saturated solution (concentration at 28°, 37·0 g per 100 g water, Handbook of Chemistry and Physics 1947).

$$\text{Thus } Wu = Ww \frac{100 - 0{\cdot}37b}{100 + b}$$

RESULTS

In all cases it was found that the relation between applied stress and specific volume followed a curvilinear form as predicted by Wroth & Basset (1965).

Analysis of the changes in packing properties of the three materials at different moisture contents was made, therefore, by plotting the regression lines of log consolidating stress against specific volume by the method of least squares. A summary of the data is given in Table 2.

Each correlation coefficient exceeds the value for 95% significance. From these regression lines the changes in specific volume with moisture content at fixed

Table 1. *Physical characteristics of materials used.*

Material	Particle shape: Elongation ratio	Particle shape: Description according to B.S. 2955:1958	Method of preparation	Particle Density gml^{-1}	Specific surface (m^2g^{-1}): From Nitrogen adsorption	Specific surface (m^2g^{-1}): By calculation from data in Fig. 1	Type
Powdered Glass (B.D.H. Ltd.)	1·85	acicular and angular	as delivered	2·45	0·71	0·19	non-porous non-cohesive
Potassium chloride B.P. (McCarthy's Ltd.)	1·32	irregular	end runner milled sieved <32μm	1·96	0·74	0·31	non-porous cohesive
Calcium phosphate B.P.C. (Boots Ltd.)..	1·51	granular	as delivered	2·96	23·2	0·33	porous cohesive

Wetting fluids:

Distilled water: $\gamma = 71{\cdot}0$ dyne cm^{-1} (mNm^{-1}) at 23°; absolute viscosity over rates of shear 0–900 $s^{-1} = 0{\cdot}95$ at 27·5° (Newtonian).

Saturated KCl soln: $\gamma = 77{\cdot}0$ dyne cm^{-1} (mNm^{-1}) at 27°; absolute viscosity over rates of shear 0–900 $s^{-1} = 1{\cdot}00$ at 28·0° (Newtonian).

Table 2. *Summary of changes in packing properties with moisture content for the three materials investigated.*

Material	Method of introducing water	Moisture content (% w/w dry basis): range	Moisture content (% w/w dry basis): average	Number of loads applied	Log consolidating stress *vs* specific volume: Correlation coefficient	Slope	Intercept
Powdered glass	Exposed to RH less than 25% and tested immediately	0·2	0·2	6	0·987	−10·0	11·5
	Distilled water added	1·0–1·1	1·1	7	0·972	−2·78	5·22
	Exposed to RH 76–78%	1·5–1·6	1·5	7	0·953	−1·84	4·01
	Distilled water added	1·9–2·2	2·0	6	0·971	−1·96	4·36
	,,	3·3–3·4	3·3	7	0·963	−1·78	4·06
	,,	6·2–6·5	6·3	6	0·995	−1·72	4·07
	,,	14·4–15·4	15·0	6	0·864	−1·48	3·60
Potassium chloride	Exposed to RH less than 29% and tested immediately	0·1–0·2	0·1	9	0·862	−1·79	4·16
	Exposed to RH 60–65% overnight	0·1–0·2	0·2	5	0·983	−1·39	4·08
	Saturated potassium chloride solution added	0·4–0·6	0·5	5	0·994	−1·85	4·54
	,,	2·8–3·2	3·0	5	0·993	−1·72	4·19
	,,	6·2–6·5	6·3	5	0·995	−1·82	4·33
	,,	12·1–12·7	12·5	6	0·965	−2·13	4·55
Calcium phosphate	Exposed to RH less than 27% and tested immediately	0·4–0·6	0·5		0·998	−1·84	4·93
	Exposed to RH less than 31% for 18 h	0·6–0·7	0·7	5	0·992	−1·56	4·39
	Untreated (tested under laboratory conditions)	1·0–1·1	1·0	7	0·996	−1.37	4·17
	Exposed to RH 82–84% for 28 h	2·2–2·7	2·4	6	0·991	−1·48	4·31
	Distilled water added	4·7–6·0	5·0	6	0·983	−1·50	4·36
	,,	8·9–10·9	10·0	5	0·989	−1.35	4·18
	,,	18·1–19·5	18·9	5	0·994	−1·91	5·14
	,,	47·4–49·8	48·2	5	0·974	−2·33	5·92

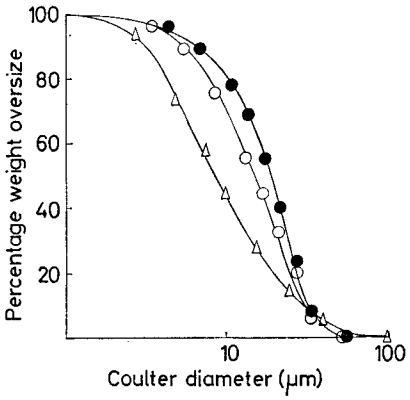

FIG. 1. Cumulative weight percentage oversize curves for materials used. △ Glass powder, ○ potassium chloride, ● calcium phosphate.

consolidating stresses were obtained and are shown in Figs 2 and 3. It can be seen from these figures that different effects are obtained with each of the materials studied.

The glass powder at increasing moisture contents up to approximately 5% w/w (dry basis) subjected to the same consolidating stress, shows increasing values of specific volume. Above 5% w/w moisture content the specific volume reaches a plateau value.

Potassium chloride shows an initial increase in specific volume with the first increase in moisture content and thereafter a decrease. Calcium phosphate, however, shows specific volumes which are more or less independent of the moisture content, even up to 50%w/w (dry basis).

The three different effects are unlikely to arise from differences in the properties of the wetting fluids since these are similar (Table 1) in those properties which are likely to affect the packing properties (i.e. viscosity and surface tension).

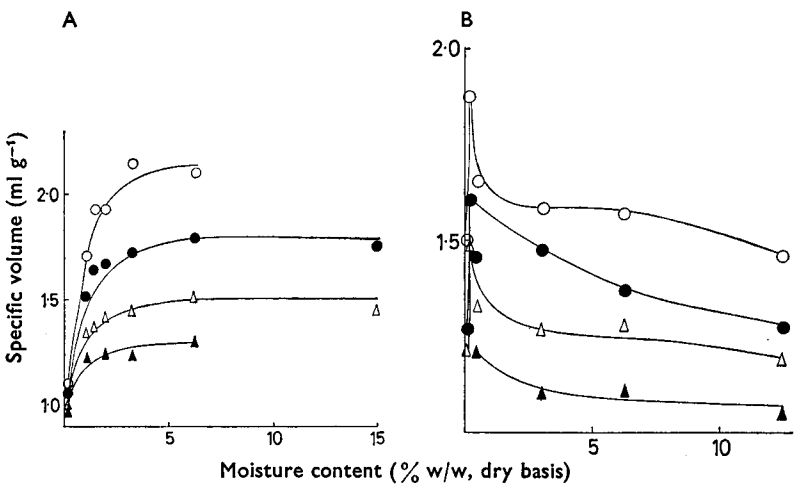

FIG. 2 (A). Effect of moisture content on the specific volume of glass powder produced by the following consolidating stresses (g cm^{-2}). ○ 3·0, ● 10·0, △ 30·0, ▲ 70·0.

(B). Effect of moisture content on the specific volume of potassium chloride produced by the following consolidating stresses (g cm^{-2}). ○ 30·0, ● 70·0, △100·0, ▲ 200·0.

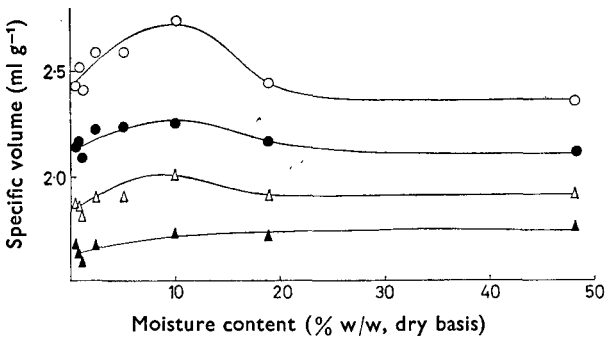

FIG. 3. Effect of moisture content on the specific volume of calcium phosphate produced by the following consolidating stresses (g cm^{-2}). ○ 3·0, ● 10·0, △ 30·0, ▲ 70·0

A striking similarity exists between Figs 2 and 3 and the corresponding curves depicting tensile strength changes with moisture content for various packing densities, Fig. 4 (Eaves & Jones, 1972b). This seems to indicate that the interparticle forces which are involved in changes in tensile strength are also responsible for changes in the specific volume.

The inherent cohesive forces in fine potassium chloride (probably van der Waals' and mechanical interactions) are initially enhanced on the addition of small quantities of moisture giving rise to an increase in strength and specific volume. These forces are subsequently disrupted by the presence of higher concentrations of moisture leading to a decrease in both tensile strength and specific volume. The other non-porous material (glass powder) possesses no such inherent cohesiveness and consequently addition of moisture builds in cohesion giving rise to an increase in tensile strength and specific volume.

The calcium phosphate, being composed of porous particles, is able to accommodate moisture in the pores so that particle interactions and, therefore, packing properties are unaffected by the addition of even large quantities of moisture.

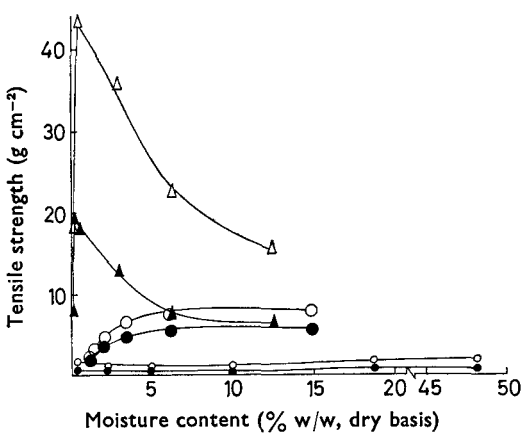

FIG. 4. Effect of moisture content on tensile strength at the following packing densities. Potassium chloride △ 0·398, ▲ 0·331; glass powder ○ 0·251, ● 0·229; calcium phosphate ○ 0·145, ● 0·120.

DISCUSSION

It is clear that the inherent cohesiveness of the bulk solid plays an important part in determining the effect of moisture on both the packing properties and tensile strength of moist beds. Differences in particle size may give rise to variation in inherent cohesiveness and the effect of such variation on packing properties and tensile strength has been demonstrated for coarse and fine sieve fractions of sodium chloride (Eaves & Jones, 1971). In this work, however, the glass powder and potassium chloride powder had similar particle size distributions but showed differences in packing properties thus indicating that inherent cohesiveness rather than particle size is the more important.

During granulation, individual particles and particle agglomerates are subject to consolidating forces by the action of machine parts, fluid velocity or interparticle abrasion. Thus the information obtained from experiments on the consolidation of moist beds is of relevance to the processes of agglomeration and granulation.

Two properties of the starting materials modified during granulation are agglomerate size and agglomerate density.

Agglomerate size is dependent, in almost every granulation process, on the tensile strength of the moist granule. For instance, in wet massing and screening the moist granules will disintegrate before or during drying if they are weak and in fluidized bed granulation, granule growth will cease if the tensile strength of the granule is insufficient to withstand the abrasive action of the fluidized bed.

The final *agglomerate density* is also dependent to a large extent on the density of the moist agglomerate, though some density changes may be expected on drying. Consequently the effects of moisture on both the densification and the strength of moist granules is worthy of further discussion.

Consider firstly the non-cohesive material; inspection of Fig. 2 (a) shows that if densification of the moist glass powder is desired, the application of consolidating stress is advantageous but an increase in moisture content is not since the specific volume produced by a given consolidating stress is increased. In processes where the applied stress cannot be varied significantly, an increase in moisture content will give rise to an increase in tensile strength (Fig. 4) which is required for granule strength. Thus a wet mass of increased strength but high porosity may be formed. (In practice it would obviously be necessary to use a binding agent where insoluble materials were to be granulated).

For cohesive, non-porous materials such as potassium chloride (Fig. 2b) an increase in consolidating stress is advantageous in achieving agglomerate densification and unlike the non-cohesive glass, an increase in moisture content is also advantageous provided it is in excess of approx. 0·5% w/w. At high moisture contents, however, the tensile strength of the wet mass is not significantly greater than the tensile strength of the dry bulk solid (Fig. 4). Consequently for optimal agglomerate densification *and* strength it would be necessary to use a small amount of wetting fluid and a maximum amount of stress. This could involve prolonged massing times to ensure homogeneity of the mix.

For both cohesive and non-cohesive materials, increasing the consolidating stress at a fixed moisture content produces a decrease in specific volume and an increase in tensile strength. Granules manufactured by processes which involve a consolidation step (e.g. massing and screening, extrusion) would, therefore, be expected to be denser than others (e.g. granules produced by fluidized bed granulation or pan

granulation). This is in agreement with work reported by Ganderton & Hunter (1971) on the intragranular porosities of calcium phosphate granules prepared by massing and screening and by pan granulation.

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