Moisture Sorption Profiles and Tensile Strength of Tableted Phenobarbitone Formulations

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Abstract—The tensile strength of tablets made from phenobarbitone and sodium phenobarbitone formulations after storage at increasing ambient relative humidity has been investigated. The moisture sorption and desorption profiles of the formulations were analysed for three locations of moisture: monolayer adsorbed moisture, normally condensed moisture and absorbed moisture. Maxima in tensile strength occur at moisture distributions determined by the disintegrant used. The changes in tensile strength have been explained in terms of changes produced in the interparticle separation, the range of the interparticle forces and changes in the ratio of the binding to diffusional forces, acting on the water molecules which are on the particles' surface.

Several investigators have reported that the mechanical strength of tablets initially increases, reaches a maximum and then decreases as moisture content is increased (Shotton & Rees 1966; Armstrong & Griffiths 1970; Esezobo & Pilpel 1976; Bangudu & Pilpel 1985). In addition to crystal bridge formation by dissolution and subsequent recrystallisation of the solids in water during compression, these changes could be associated with a variety of effects caused by the moisture content, for example, reduction in the resistance to particle deformation and interparticulate friction, change in the strength of interparticulate bonds, and reduction of the amount of fragmentation occurring on compression. For materials which take up water in a non-specific manner, these changes may be related to the fact that moisture can be present in different physical states: as an adsorbed monomolecular layer, as an adsorbed multilayer, and as moisture absorbed into the interior of the particles.

For powdered biological materials (e.g. starch) the amounts of moisture present in the three different physical states can be calculated from the sorption and desorption isotherms and the Young & Nelson (1967a, b) equations which were derived on the basis of the kinetic theory of gases:

$$\theta = \frac{RH}{RH + (1 - RH)E} \qquad \text{(eqn 1)}$$

$$\psi = RH\theta \qquad \text{(eqn 2)}$$

$$\beta = -\frac{E(RH)}{E - (E-1)RH} + \frac{E^2}{E-1}\log_e\frac{E - (E-1)RH}{E} \\ -(E + 1)\log_e(1 - RH) \qquad \text{(eqn 3)}$$

$$M_s = A(\theta + \beta) + B\psi \qquad \text{(eqn 4)}$$

$$M_d = A(\theta + \beta) + B\theta RH_{max} \qquad \text{(eqn 5)}$$

where θ is the fraction of surface covered by a monomolecular layer, ψ is the fraction of surface covered by a layer of

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water two or more molecules thick, β is the total amount of adsorbed moisture in a multilayer, M_s and M_d are the moisture contents of the powder during sorption and desorption conditions and RH and RH_{max} are the relative humidity and maximum relative humidity of the environment. A, B and E are variables unique to each material. $E = \exp[(-q_1 - q_L)/kT_a]$, where $q_1 = $ heat of adsorption of water molecules bound to the surface, $q_L = $ normal heat of condensation of water molecules, $k = $ Boltzmann's constant and $T_a = $ absolute temperature. $A = (\rho_w V_m)/W_m$ and $B = (\rho_w V_a)/W_m$, where V_m and V_a are, respectively, the volumes of the adsorbed and absorbed moisture, ρ_w is the density of water and W_m is the mass of dry material. A θ is the amount of moisture present as a monolayer, $A(\theta + \beta)$ is the externally adsorbed moisture and $B\psi$ is the amount of the internally absorbed moisture during the sorption cycle.

The above equations have been used by York (1981) to analyse the moisture distribution patterns of phenobarbital/starch mixtures and by Chan & Pilpel (1983) to correlate the moisture content with the tensile and shear properties of sodium cromoglycate/lactose mixtures. However, Zografi et al (1983) suggested that the Young & Nelson model may not adequately describe the physical state of water vapour taken up by biological materials, such as starch.

The aim of the present investigation was to see whether, at various relative humidities, any correlation could be found between interparticle separation, range of interparticle forces, the tensile strength of the tablets and the moisture distribution in the three different physical states. Such correlations might provide information about the bonding mechanisms that occur during a compression process and assist formulators in the choice of excipients and manufacturing conditions.

Materials and Methods

Phenobarbitone (phenobarbital USP, Siegfried-Switzerland) and sodium phenobarbitone (sodium phenobarbital DAB, USP, Merck, FRG) were used. Maize starch (Carl Roth, W. Germany) and polyplasdone XL (PVPP, Fluka,

Table 1. Particle size parameters $\bar{d}$, $\bar{s}$ and $\bar{v}$ for the powder mixtures.

Mixture	$\bar{d}$ (μm)	$\bar{s}$ (μm)^2	$\bar{v}$ (μm)^3
M₁ Phenobarbitone/Starch	21·9	1225·5	33,807
M₂ Phenobarbitone/PVPP	22·4	1278·1	36,229
M₃ Sod. Phenobarbitone/Starch	22·4	1272·5	34,846
M₄ Sod. Phenobarbitone/PVPP	22·9	1325·1	37,286

Switzerland) were used as disintegrants. All powdered materials passed a 65 μm stainless steel sieve.

Four mixtures (Table 1) were prepared containing equal drug/disintegrant proportions (1/1 w/w) and their particle size parameters (mean effective diameter, $\bar{d}$; surface area, $\bar{s}$; and volume, $\bar{v}$) were determined by optical microscopy and using the relationships proposed by Cheng (1968).

The mixtures were wet granulated with a 6% w/v aqueous solution of gelatin (Bloom no. 250, acid treated hide gelatin, Richard Hodgson Ltd, UK) or a 6% solution of PVP k-30 in isopropanol. Eight batches of granules (Table 2) were prepared and the fractions with granule size < 450 μm were collected.

Moisture sorption and desorption isotherms

Samples of the granules were dried over phosphorus pentoxide and transferred to tightly closed desiccators at 25°C, and relative humidities of 11, 23, 33, 43, 52, 65, 75, 83 and 93% which were provided by saturated salt solutions (Callahan et al 1982). After standing for 8 days, their equilibrium moisture contents were determined in triplicate to within ±0·5% by drying to constant weight in a hot air oven at 105°C. The results yielded the sorption isotherms. A similar procedure was followed for determining the desorption isotherms, after initial equilibration over saturated potassium nitrate (RH 93% at 25°C).

Tablet preparation and testing

After the sorption cycle, the equilibrated granules, whose moisture content had been determined as above, were formed into tablets in a hydraulic press. Granules in 500 mg batches were placed in small aluminium dishes and were stored in the same desiccators until they were compressed. The granules were quickly emptied into a 13 mm diameter die with flat faced punches, which were lubricated with a suspension of magnesium stearate in chloroform. Compression pressure from 20 to 100 MPa was applied for 10 s to the upper punch by lowering the hydraulic ram at a rate of 1·5

mm s^{-1}. The resulting tablets were weighed to the nearest 0·1 mg and their thickness (th) and diameter (d) were measured to the nearest 0·01 mm. The maximum force (P) transmitted by the tablets before they broke in tension was measured using a Heberlein tester (Mod. 2E/205) immediately after preparation. The tablets' packing fraction (p$_f$ = bulk density/ true granule density) and tensile strength (T = 2P/(πdth)) were calculated according to Fell & Newton (1970). True granule density was measured by an air comparison pycnometer (Beckman Mod. 930).

Results

The granules containing sodium phenobarbitone and polyplasdone XL showed the greatest tendency to moisture uptake, 36% w/w at 93% RH; and those containing starch and phenobarbitone the least, 10% w/w at 93% RH (Table 2).

The experimentally measured sorption and desorption isotherms for all the granulations studied, exhibited hysteresis. By computer fit to both sorption and desorption data, simultaneously using three adjustable parameters, E, A and B and a combination of iteration and multiple regression techniques, the values of these parameters describing equations 4 and 5, were obtained for each granulation and are given in Table 2. The combined iteration and multiple regression technique used has been described in detail by Young & Nelson (1967a, b), York (1981) and Chan & Pilpel (1983). The highest correlation coefficients obtained in this study are given in Table 2.

The values of θ, ψ and β were obtained by inserting E, A, B, values into equations 1, 2 and 3. These values yielded the amount of moisture present as a monolayer (Aθ) the amount of the externally adsorbed moisture A(θ+β) and the amounts of the internally absorbed moisture during the sorption cycle ($\beta\psi$) and during the desorption cycle (BθRH$_{max}$). The theoretical sorption and desorption isotherms were then plotted by adding A(θ+β) plus Bψ and A(θ+β) plus BθRH$_{max}$, respectively, at different relative humidities. The theoretical plots for the granulations that exhibited the highest and lowest moisture uptake are shown in Fig. 1.

From the diametral compression results, it was found that the tensile strength, (T), for all the samples at all the relative humidities studied fitted the general equation:

$$\log T = A_1 p_f + B_1 \qquad \text{(eqn 6)}$$

Table 2. Computed values of E, A, B and correlation coefficients (r) from equations 4 and 5.

| Granulation			% Moisture uptake at 93% RH	E	A	B	(r)
Drug	Disintegration	Binder					
M₁₁ Phenobarbitone	Starch	Gelatin	10	0·085	0·015	0·047	0·99
M₂₁ Phenobarbitone	PVPP	Gelatin	25	0·133	0·020	0·156	0·99
M₁₂ Phenobarbitone	Starch	PVP	12	0·135	0·016	0·061	0·99
M₂₂ Phenobarbitone	PVPP	PVP	28	0·188	0·018	0·204	0·97
M₃₁ Sod. Phenobar.	Starch	Gelatin	15	0·124	0·020	0·073	0·99
M₄₁ Sod. Phenobar.	PVPP	Gelatin	29	0·145	0·027	0·178	0·99
M₃₂ Sod. Phenobar.	Starch	PVP	18	0·112	0·022	0·094	0·98
M₄₂ Sod. Phenobar.	PVPP	PVP	36	0·185	0·030	0·239	0·98
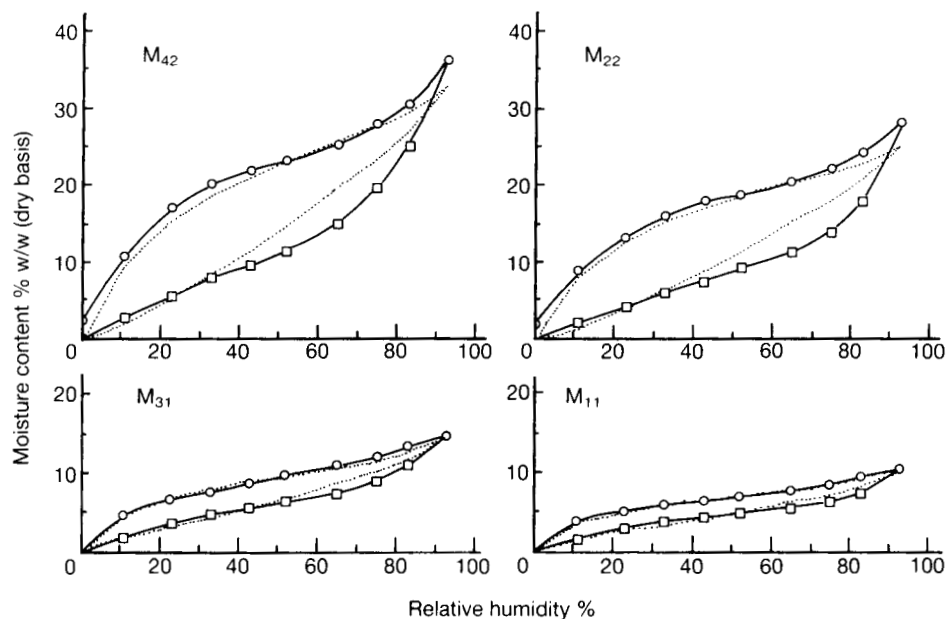
FIG. 1. Moisture sorption and desorption isotherms of granules at 25°C. (□—sorption, ○—desorption, ——experimental and · · · · computed data).

with a highly significant correlation (>0.95 for at least 7 measurements) over the range of packing fraction, (p_f), from 0·6 to 0·9. A_1 and B_1 are numerical terms which depend on the composition of the granules and the relative humidity during moisture sorption in the desiccators. The tablets which contained polyplasdone XL became very weak at relative humidities greater than 75%, making measurement of tensile strength impossible.

To determine the effect of increasing the relative humidity

on the tensile strength of the tablets, the values of T at different packing fractions were obtained from equation 6 by regression, and were plotted against the relative humidity. Fig. 2 shows the plots for the granulations that exhibited the highest and lowest tendency to moisture uptake. These plots are typical of the way in which the tensile strength varied with the relative humidity and with the packing fraction for all the granulations studied.

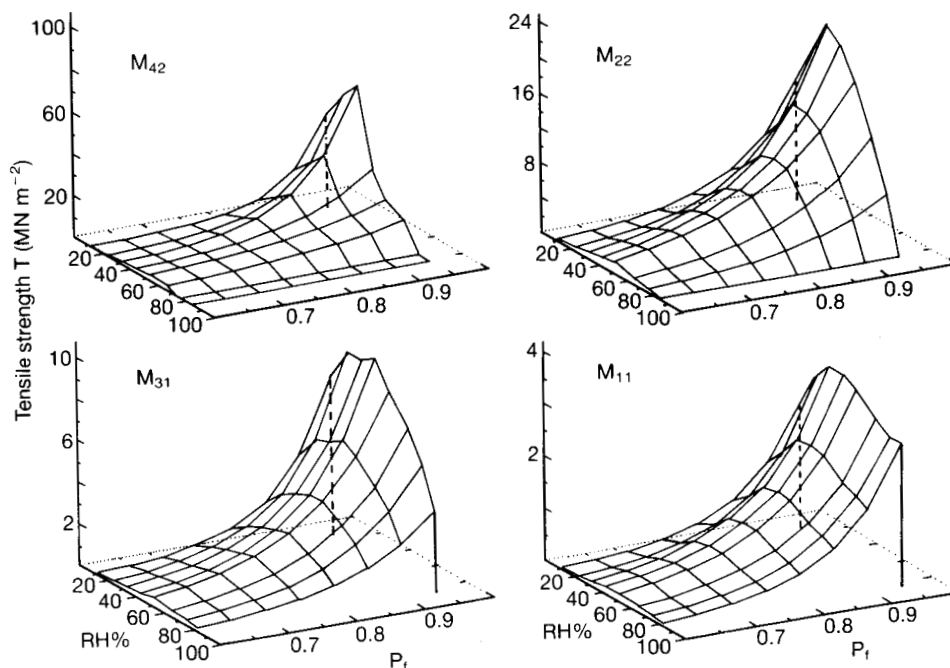At fixed packing fractions, increasing the relative humidity

FIG. 2. Tensile strength T versus relative humidity at various packing fractions for tablets.

Table 3. Relative humidity for tensile strength maxima at p_f 0·6 and 0·9 and amounts of moisture (% w/w dry basis) present in different physical states.

| Granulation | $p_f = 0·6$ | | | | | $p_f = 0·9$ | | | | |
	RH%	$A\theta$	$A(\theta+\beta)$	$B\psi$	$\frac{B\psi}{A(\theta+\beta)}$	RH%	$A\theta$	$A(\theta+\beta)$	$B\psi$	$\frac{B\psi}{A(\theta+\beta)}$
M_{11}	48	1·4	2·3	2·1	0·9	40	1·4	2·1	1·7	0·8
M_{21}	51	1·7	3·0	7·1	2·3	46	1·7	2·8	6·2	2·2
M_{12}	50	1·4	2·4	2·7	1·1	40	1·3	2·0	2·0	1·0
M_{22}	52	1·5	2·7	9·0	3·3	44	1·4	2·3	7·2	3·1
M_{31}	55	1·8	3·3	3·7	1·1	36	1·6	2·4	2·2	0·9
M_{41}	48	2·3	3·9	7·4	1·9	43	2·3	3·6	6·4	1·8
M_{32}	50	2·0	3·4	4·2	1·2	44	1·9	3·1	3·6	1·2
M_{42}	55	2·6	4·7	11·4	2·4	42	2·4	3·7	8·0	2·2

initially produced an increase in the tensile strength to a maximum, followed by a decrease. The relative humidities at which maxima occurred, decreased as the packing fraction was increased. (The relative humidities at the maxima of packing fractions of 0·6 and 0·9 obtained by multiple regression analysis, are given in Table 3.

To interpret the tensile strength results of the tablets in terms of particle size parameters ($\bar{d}$, $\bar{s}$ and $\bar{v}$), interparticle separation (t), and range of interparticle forces t_o (Cheng 1968), values of

$$\frac{1}{F} = \frac{\bar{s}p_f}{2\bar{v}T}$$ were plotted versus $\log \frac{d}{3}\left(\frac{p_f}{p_{fo}} - 1\right)$

(Malamataris & Pilpel 1983). p_{fo} is the packing fraction of granulations milled to their original particle size when their tensile strength equals zero (T=0). The p_{fo} values were obtained from measurements in a Warren Spring split cell (Ashton et al 1964). The values of t_o were obtained by extrapolation; the log scale and straight line plots avoid the abrupt changes in slope which occur when

$$\frac{1}{F}$$ is plotted against $\frac{d}{3}\left(\frac{p_f}{p_{fo}} - 1\right)$

(Esezobo & Pilpel 1977). The interparticle separation t at p_f 0·75 was obtained from t_o by substituting into the equation

$$t = t_o - \frac{d}{3}\left(\frac{p_f}{p_{fo}} - 1\right).$$

The values of t_o and t for the four typical formulations are included in Table 4.

It can be seen that with all the granulations investigated, the values of both t_o and t increased with the increase of relative humidity up to 43–52% and above this they did not alter significantly.

The amounts of moisture present in the different physical states, when the tensile strength maxima occurred, were calculated from the computed sorption-desorption isotherms. The results at packing fractions of 0·6 and 0·9 are given in Table 3.

For all the granulations containing starch the tensile strength maxima occurred when the internally absorbed moisture $B\psi$ was approximately equal to the amount present as externally adsorbed moisture A $(\theta+\beta)$. For the granulations containing polyplasdone XL, the maxima occurred when the internal moisture $(B\psi)$ was between two and three times the amount present as external moisture $A(\theta+\beta)$.

Table 4. Interparticle separation (t) and range of forces (t_o) for tableted formulations.

Granulation	RH %	p_{fo}	t_o (μm)	t at p_f 0·75 (μm)
M_{11}	11	0·46	6·1	1·5
	23	0·44	7·1	1·9
	33	0·42	7·9	2·1
	43	0·41	9·2	3·2
	52	0·40	10·0	3·6
	65	0·40	10·2	3·8
	75	0·40	10·3	3·9
	83	0·40	10·1	3·7
	93	0·40	10·2	3·8
M_{22}	11	0·37	7·9	0·3
	23	0·37	8·5	0·8
	33	0·37	9·0	1·3
	43	0·36	10·7	2·6
	52	0·35	12·4	3·9
	65	0·35	12·4	3·8
	75	0·35	12·6	4·1
	83	*	*	*
	93	*	*	*
M_{31}	11	0·35	10·4	1·8
	23	0·35	10·4	2·8
	33	0·35	11·4	2·8
	43	0·34	12·2	3·2
	52	0·33	13·6	4·0
	65	0·33	13·7	4·1
	75	0·33	13·5	3·9
	83	0·33	14·5	5·0
	93	0·33	14·7	5·2
M_{42}	11	0·40	6·7	0·1
	23	0·40	7·1	0·4
	33	0·39	8·0	0·9
	43	0·37	11·4	3·6
	52	0·36	13·8	5·5
	65	0·37	13·7	5·9
	75	*	*	*
	83	*	*	*
	93	*	*	*

* Very soft or weak tablets.

Discussion

The occurrence of the tensile strength maxima at similar moisture distributions (ratio of internal to external moisture, $(B\psi)/A(\theta+\beta)$) for the granulations containing the same disintegrant, can be attributed to the action of the disintegrant at increasing relative humidities as a sink for the water molecules. The higher ratios $(B\psi)/(A(\theta+\beta))$ for the polyplasdone XL (Granulations M_{21}, M_{22}, M_{41}, M_{42}—Table 4) can be attributed to various factors which have been proposed as

possible explanations of the mechanisms of moisture sorption hysteresis. These are the differences in advancing and decreasing contact angles between adsorbent and adsorbate, the role of surface pore shape and volume, and the phenomenon of multilayer adsorption followed by capillary condensation in cylindrical pores (Gregg 1961).

The tensile strength of tablets is mainly determined by the range and magnitude of the van der Waals' forces between the particles, and the development of additional bonds formed by plastic deformation/melting of the powder particles or the binder films developed during granulation (Malamataris & Pilpel 1983). The initially adsorbed water molecules on the surfaces may form a monomolecular layer and increase the van der Waals' forces, smoothing out the surface microirregularities and reducing the interparticle separation (Eaves & Jones 1972). As more water molecules adhere to the surface, they are subjected to both surface binding and diffusional forces, the latter tending to cause moisture transfer into the material (Young & Nelson 1967a; York 1981). The water diffusing and penetrating the micropore structure may cause softening of the particles' surfaces, and under high pressure the area of contact between the particles will increase with plastic deformation, and more solid bonds may be formed (Bangudu & Pilpel 1985). These effects could account for the observed initial increase in the tensile strength of the tablets with increase in relative humidity. However, as more water is absorbed, the particles and the bonds soften and weaken and the tensile strength decreases.

Considering the increases in the values of t_o and t when the relative humidity is increased up to the range of 43–52% (Table 4), where, incidentally, the tensile strength maxima occur, the following explanation might apply: the moisture modifies the surface characteristics of the particles, i.e. the parameters t_o and t where t_o is the least distance between the particles at zero tensile strength and t is the average interparticle separation of every microcontact at which the interparticle attractive force exists. Similar results have been reported by Esezobo & Pilpel (1977) who showed that the values of t_o increased with moisture content in an oxytetracycline formulation and they ascribed this increase to the development of additional attractive forces due to surface tension.

Another possible reason for tensile strength decreases at higher relative humidities could be that, at these values, the water is beginning to form multilayers on the surface of the materials, thereby acting as a lubricant and thus reducing the frictional forces responsible for attraction between particles (Coelho & Harnby 1978).

The occurrence of the tensile strength maxima at similar ratios of $(B\psi)/(A(\theta+\beta))$, characteristic of each disintegrant used, and generally at lower relative humidity when the packing fraction increased, may be an indication that the tensile strength maxima should be related to the ratio of binding to the diffusional forces acting on the water adhering to the powder surface (Young & Nelson 1967a, b; York 1981; Chan & Pilpel 1983). When the dry material is exposed to moisture, water molecules are first adsorbed onto the surfaces to form a monomolecular layer and then a second layer or a multilayer which prevents the evaporation of water from the monomolecular layer beneath it. The monomolecu-
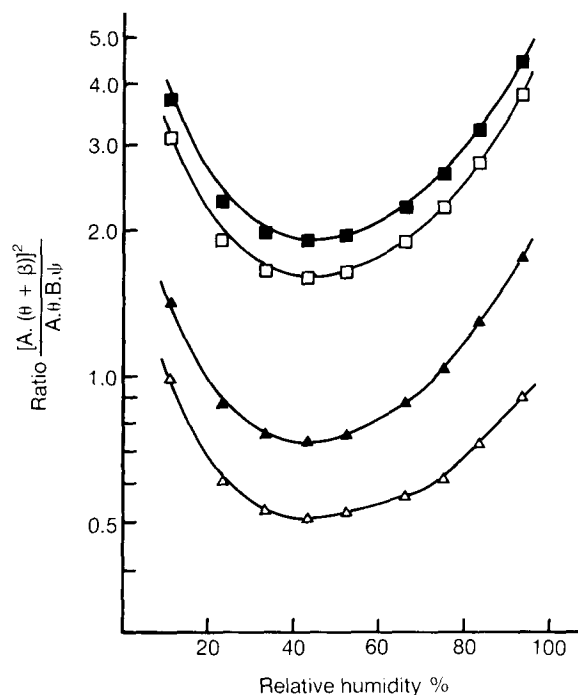

Fig. 3. Ratio $[A(\theta+\beta)]^2/A\theta B\psi$ versus relative humidity for the granulations: ■—M_{11}, △—M_{22}, □—M_{31}, ▲—M_{42}.

lar layer is subject to both surface binding and diffusional forces. The diffusional forces exceed the binding forces as more water molecules adhere to the surfaces and moisture is transferred into the material. Assuming that the surface binding forces may be represented by $(A(\theta+\beta)/A\theta)$ and the diffusional forces by $(B\psi/(A(\theta+\beta)))$ their ratio could be expressed by $[A(\theta+\beta)]^2/A\theta B\psi$. In spite of the numerous simplifying assumptions made in the derivation of the above ratios, the plots of $[A(\theta+\beta)]^2/A\theta B\psi$ versus the relative humidity (Fig. 3) do indeed show minima at the range of relative humidity between 43 and 52%, where the tensile strength maxima occur.

At higher packing fractions, the surface area of the powder in the tablets is reduced and the external moisture $A(\theta+\beta)$ probably forms thicker multilayers and diffuses more easily into the interior of the particles. This may be the reason why the tensile strength maxima occur at lower relative humidities as the packing fraction increases (Table 3).

Another index of the binding and diffusional forces may be the increment of moisture uptake in the sorption profiles (Fig. 1). At low relative humidities (adsorption phase) the binding forces which are mainly responsible for the moisture uptake are reduced with the relative humidity and cause a decrease in the increment of moisture uptake. At high relative humidities, after the adsorption phase, the presence of a second layer or multilayer causes a relative increase in the diffusional forces which tend to move the water into the interior of the particles and increase the increment of moisture uptake with the relative humidity (absorption phase). The changes in the increment of moisture uptake with respect to ambient relative humidity may be expressed by means of the differential coefficient (first derivative) of the functions computed from the experimental sorption data.
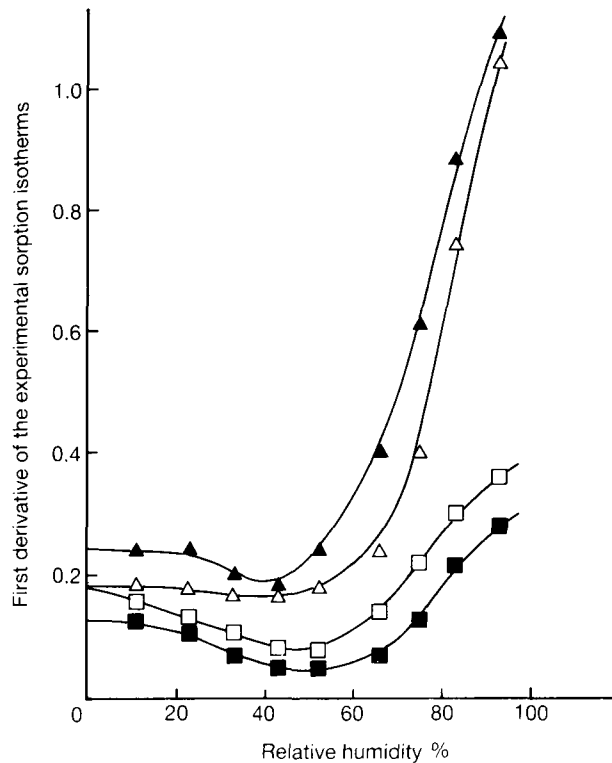
FIG. 4. First derivative of the experimental sorption isotherms versus relative humidity, for the granulations: Key as in Fig. 3.

Plots of the first derivative for the four typical formulations are given in Fig. 4. These curves also show minima close to the relative humidities where the tensile strength maxima occur.

strengths of tableted pharmaceutical formulations containing materials which absorb moisture easily (e.g. disintegrants) can be appreciably affected by the usual variation in the environmental relative humidity (30–60%). The tensile strength changes could be explained in terms of the van der Waals' forces operating between the particles and the solid bonds developed by plastic deformation. For the multicomponent systems used in this study, with moisture present as a tightly bound monomolecular layer as a less tightly bound multilayer of water, and as water absorbed into the interior of the particles in a non-specific manner, correlations have been established between the moisture distribution patterns (as derived from the moisture sorption profiles) and the changes in the tensile strength of the tablets.

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