International Journal of Pharmaceutics, 60 (1990) 197–202
Elsevier

IJP 02044

Bonding mechanisms in tabletting

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(Received 12 May 1989)
(Modified version received 1 October 1989)
(Accepted 1 November 1989)

Key words: Tablets; Bonding mechanism; Van der Waals force; Surface free energy

Summary

An investigation has been undertaken to ascertain the role of Van der Waals forces in the formation of tablets. Hamaker constants have been estimated from surface free energy measurements on polymers and materials of pharmaceutical interest. The relationship betwen the Hamaker constants in media of different dielectric constant and the tensile strength of tablets determined by diametral compression in these media has been examined. It was found that the change in Hamaker constant in the different media paralleled the change in tablet tensile strength. It is considered that the bonding of these tablets can be largely accounted for by Van der Waals attractive forces.

Introduction

There have been limited studies carried out on the bonding mechanisms involved in tabletting, due, in part, to difficulties in making appropriate measurements on powders. Rumpf (1962) has proposed five mechanisms by which powders may be bonded together of which, solid bridges caused by melting and subsequent cooling, mechanical interlocking of irregular particles and attractive forces are likely to apply to compressed powders. Attractive forces include coulombic forces between charged species, covalent bonds, hydrogen bonds and Van der Waals forces. Recent work by Luangtana-Anan and Fell (1987, 1988a,b) has shown that the surface energy characteristics of powders can be successfully determined, hence enabling the calculation of Hamaker constants for Van der Waals interactions.

This paper presents work designed to interpret the tensile strength of tablets in terms of calculated Hamaker constants. As the Hamaker constant, which is a measure of the Van der Waals forces of attraction, varies predictably in media of different dielectric constant, tensile strengths are measured in different liquid media and theoretical and experimental results compared. In this respect the work is similar to that of Rowe (1988a,b,c), who used solubility parameters as a measure of interactions, in that it endeavours to use fundamental information to help explain the practical technique of tabletting.

Theoretical Background

The magnitude of Van der Waals forces between two flat surfaces can be estimated by vari-

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ous methods (Hamaker, 1937; Lifshitz, 1956). It can be expressed by:

$$\Delta F = -\frac{A}{12\Pi d_0^2} \tag{1}$$

where A is the Hamaker constant, d_0 is the distance of separation and ΔF is Van der Waals energy.

A can be obtained from surface free energy measurements. Since Van der Waals energy is the reversible work required to separate two parallel plates from a distance d to infinity, producing two new surfaces, the energy can be given by:

$$\Delta F_{ii} = -2\gamma_{iv} \tag{2}$$

where subscripts i and v represent the solid surface and vapour phase, respectively. Combining Eqns 1 and 2 gives:

$$A = 24\Pi d_0^2 \gamma_{iv} \tag{3}$$

If the surfaces are in liquid media, L, rather than air, then Eqn 3 becomes:

$$A = 24\pi d_0^2 \gamma_{iL} \tag{4}$$

The interfacial force is given by the equation of Wu (1971) and can be determined from contact angle measurements (Luangtana-Anan and Fell, 1987, 1988a).

$$\gamma_{iL} = \gamma_i + \gamma_L - 2\left(\gamma_i^d \gamma_L^d\right)^{1/2} - 2\left(\gamma_i^p \gamma_L^p\right)^{1/2} \tag{5}$$

where the superscripts d and p represent the dispersion and polar components of the surface energy, respectively.

Materials and Methods

Materials

The materials chosen were those that could be readily characterised by contact angle measurements and would give minimal problems due to solubility. They were polytetrafluoroethylene (PTFE, BDH Chemicals, Poole, U.K.), polymethylmethacrylate (PMMA, BDH), polyvinylchloride (PVC, BDH), vinyl chloride vinyl acetate copolymer (Corvic 47/488, ICI), Emcompress (Cambrian Chemicals), magnesium carbonate (BDH) and methylcellulose (BDH). These materials were all sieved to give a 180–250 μm size fraction except for Corvic (90–125 μm). The liquids used, which had dielectric constants varying from 5.82 to 32.7, were methanol, ethanol, 2-propanol, 1-butanol and 1-hexanol (Fisons), 1-octanol and 2-methyl-2-butanol (BDH).

Methods

Preparation of tablets. Preparation of 1.27 cm diameter flat-faced tablets was by direct compression using a single-punch tablet machine (F3 Manesty, Liverpool). The upper punch of the machine was instrumented using strain gauges in order to detect the force applied to the powder. The output of the strain gauges was recorded on a UV recorder (SE 3006 North Feltham Trading Estate, Middlesex, U.K.). Different weights of powder were used and pressures applied to give acceptable tablets (Table 1). The speed of compression was about 300 cm/min.

Diametral compression test. The tensile strength of the tablets was determined following the procedure of Fell and Newton (1968). The test specimens were studied in air and in different liquid media. Speciments in liquid media were obtained by immersion of the compacts in the appropriate liquid for periods of time sufficient to ensure complete saturation. The time periods were

TABLE 1

The compressed materials, weights, applied pressures and method of preparation

Materials	Weight (mg)	Pressure (MN/m^2)	Method
PTFE	530	135.5	compressed continously
PVC	530	135.5	compressed continously
Corvic	530	226	compressed continously
Encompress	570	226	compressed individually
Magnesium carbonate	570	226	compressed individually
Methyl cellulose	565	108.4	compressed individually

TABLE 2

Surface free energies and their dispersion and polar forces

Solids	γ_s^p	γ_s^d	γ_s
PTFE	2.0	16.0	18.0
PVC	26.0	20.0	46.0
Corvic	24.0	12.0	36.0
PMMA	17.7	18.2	35.9
Encompress	27.2	21.2	48.4
Magnesium carbonate	19.5	24.6	44.1
Methyl cellulose	31.8	18.1	49.9

TABLE 3

Properties of immersion liquids

Liquids	Dielectric constants [a]	θ on paraffin	Surface tension (mN/m)		
			γ_s^d	γ_s^p	γ_s
Methanol	32.7	35.1	18.2	5.5	23.7 [b]
Ethanol	24.5	29.5	19.3	4.4	23.7 [b]
Propanol	19.9	16.9	19.5	3.5	23.0 [b]
Butanol	16.1	21.0	22.2	2.4	24.6
Hexanol	13.3	25.2	22.8	2.5	25.3
Octanol	10.3	27.0	26.6	0.9	27.5
2-Methyl-2-butanol	5.8	19.4	22.9	1.7	24.6

[a] Data from Weast (1987).

[b] Data from Ohm and Lippold (1985).

determined experimentally from observation of broken compacts and varied from 5 min to 2 h.

These wet and dry test compacts were then placed between the two flat plattens of an Instron instrument (Instron, floor model, High Wycombe, U.K.). The load was applied to the tablet diametrically at a cross-head speed of 0.1 cm/min. The maximum load at failure was obtained from the chart recording of applied load and cross-head

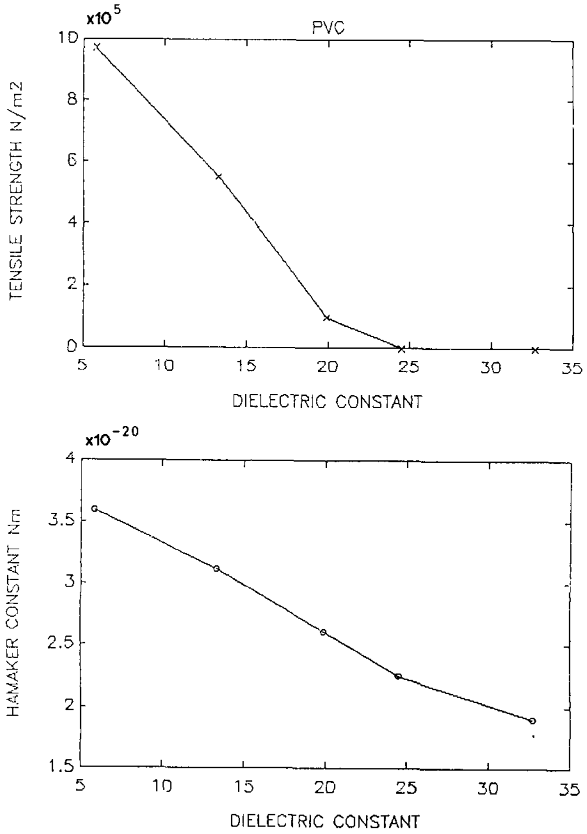

Fig. 1. Relationships between changes in tensile strength and Hamaker constant to those in dielectric constant for PVC.

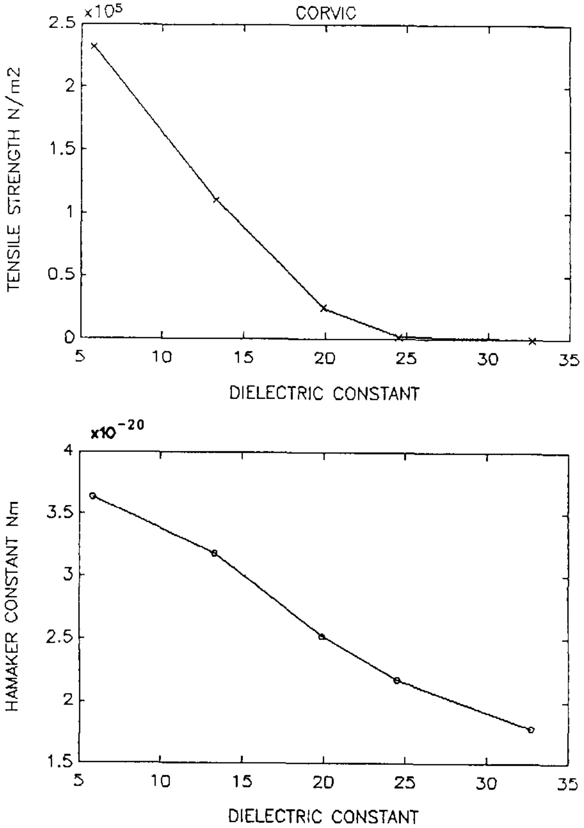

Fig. 2. Relationships between changes in tensile strength and Hamaker constant to those in dielectric constant for Corvic.

movement. The tensile strength was calculated by the application of the following equation:

$$\sigma = \frac{2P}{\Pi d T} \tag{6}$$

where σ is the maximum tensile stress, P is the applied load, d is the specimen diameter, and T is thickness of the specimen. The tablets were examined after breaking to ensure their saturation and to check that failure occurred in normal tension. Only results from tablets breaking clearly in tension were accepted. The mean tensile strength of at least five tablets was calculated.

Surface free energy of test compacts. The surface free energies were determined from contact angle measurements as described by Luangtana-Anan and Fell (1987). The surface free energies and their components were calculated using a pair of liquids with known dispersion and polar forces. The results are listed in Table 2.

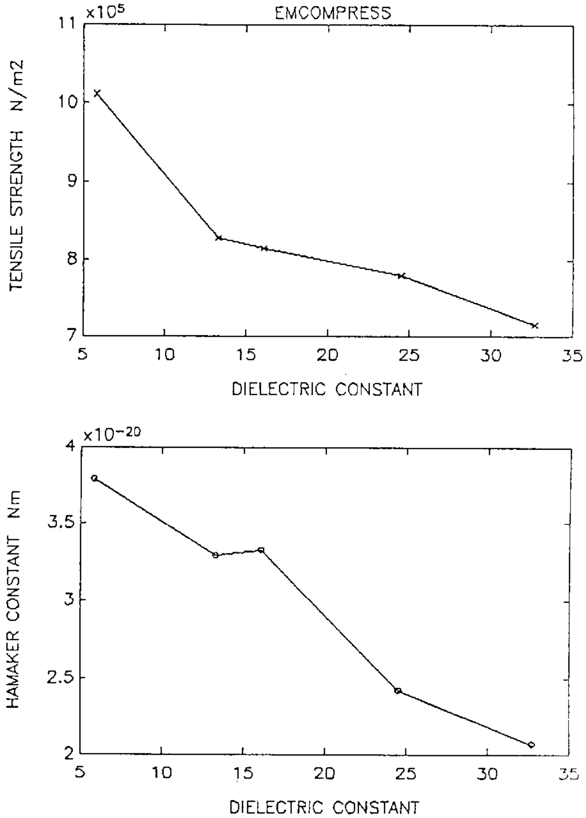

Fig. 3. Relationships between changes in tensile strength and Hamaker constant to those in dielectric constant for Emcompress.

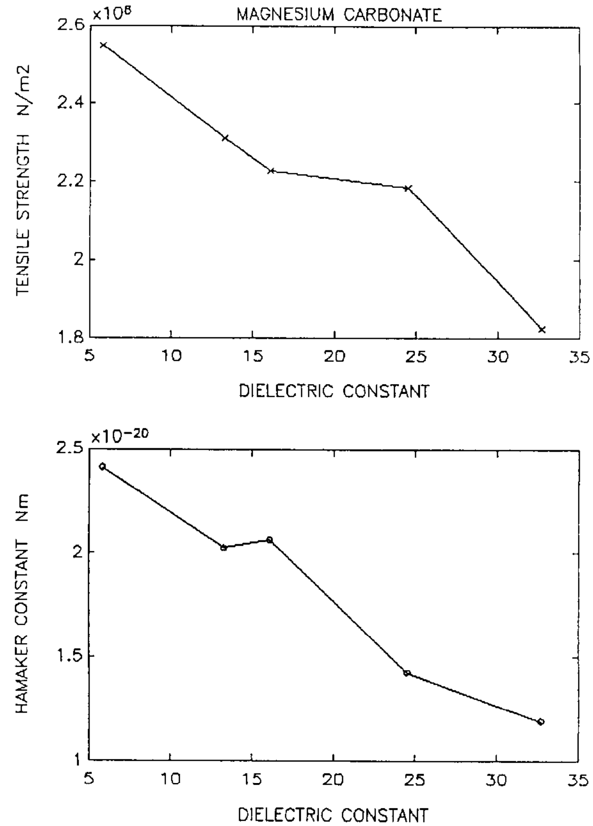

Fig. 4. Relationships between changes in tensile strength and Hamaker constant to those in dielectric constant for magnesium carbonate.

The dispersion and polar forces of the series of alcohols were determined from contact angles formed on paraffin compacts. These results and the dielectric constant values of the alcohols are shown in Table 3.

Results and Discussion

The tensile strengths and Hamaker constant values of the materials (A) in different liquids, calculated according to Eqns 3 and 4, are shown in Figs 1–5. The values in air are given in Table 4. The distance of separation in Eqns 3 and 4 is taken to be 0.2 nm, being the distance which gives

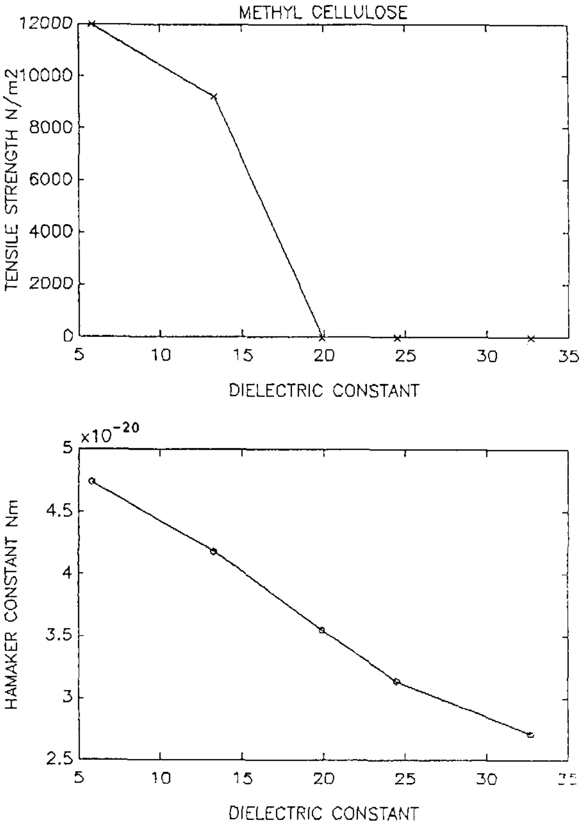

Fig. 5. Relationships between changes in tensile strength and Hamaker constant to those in dielectric constant for methyl cellulose.

the maximum attractive force (Israelachvili, 1985). The use of this value is justified on the grounds that comparative values are required in this study and not absolute values. The Hamaker constants of all the materials change with alterations in dielectric constant. The change can be divided into two groups: (1) all powders (except PTFE) which have a significant polar component of the total surface free energy and PTFE whose surface free energy is almost entirely composed of dispersion forces (Table 2). For the powders excluding PTFE the Hamaker constants increase as the dielectric constants decrease. The Hamaker constants for materials separated by an intervening medium were calculated based on the magnitude of the interfacial forces (Eqn 4). In other words, the A value depends on the balance between the liquid and solid phases (equation reported by Wu (1971)). The changes in tensile strength on variation in the dielectric constant are in agreement with the A values. The higher dielectric constant gives the lower tensile strengths. Similar results

TABLE 4

Tensile strength ($\sigma \times 10^5$ N/m^2) and Hamaker constants ($A \times 10^{-2}$ N m) for the materials in air

Material	Tensile strength (σ)	Hamaker constant (A)
PTFE	6.66	4.50
PVC	12.07	11.49
Corvic	4.19	8.99
Emcompress	8.54	12.09
Magnesium carbonate	23.64	11.02
Methyl cellulose	4.37	12.46

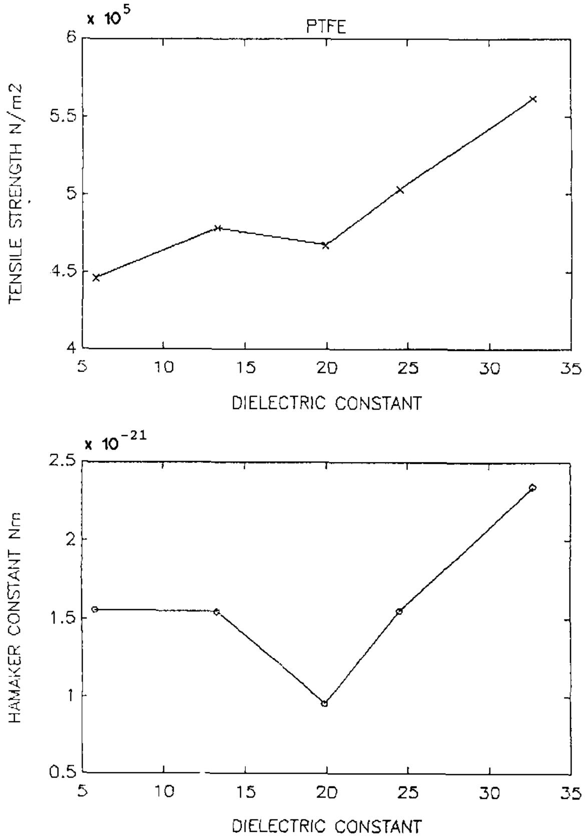

Fig. 6. Relationships between changes in tensile strength and Hamaker constant to those in dielectric constant for PTFE.

were obtained by Fraser (1973) using lactose tablets. The tensile strength of PVC and Corvic in methanol could not be tested as all the compacts disintegrated into particles. The relationships between the changes in Hamaker constant and tensile strength to the changes in dielectric constant are shown in Figs 1–5. All the curves show similar trends. This indicates that Van der Waals forces contribute significantly to the strength of the compacts and the quality of the bonds can be estimated by the Hamaker constant values.

For PTFE, however, the opposite effect was observed. The higher is the dielectric constant, the higher the tensile strength and Hamaker constant. The only forces operating in this system are dispersion forces, therefore the magnitude of these forces depends on the degree of interaction of the dispersion forces between the solid and liquid phases. Since methanol has the lowest dispersion force value it gives the lowest degree of interaction, and hence, the highest tensile strength and also the highest Hamaker constant. Fig. 6 shows the relationships between the tensile strengths, Hamaker constants and dielectric constants. As with the other powders, there is a relationship between the tensile strength and the Hamaker constant.

The results indicate that the change in the tensile strength of tablets saturated with different liquid can be predicted on the basis of Van der Waals forces and that these forces must therefore contribute significantly to the tensile strength of the tablets. An approach such as that described can only predict trends, the gulf between the theory of intermoleclar attractive forces and the complex heterogeneous nature of a tablet being too wide to expect predictions of absolute values. Nonetheless, the correspondence between the theoretical and practical results and the prediction of the opposite behaviour of PTFE to that of the other powders lends credence to the approach. It is considered that not only bonding, but also deeper understanding of the disintegration of tablets may be obtained using this type of evaluation.

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