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J. Pharm. Pharmacol. 1983, 35: 404–405
Communicated November 24, 1982

Influence of porosity on the contact angle of non-wettable solids

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The wettability of pharmaceutical powders is one of the controlling factors in the dissolution of dosage forms (Zografi & Stamley 1976; Fell & Efentakis 1978; Lerk et al 1978), in the design of suspensions (Parfitt 1973) and in some technological processes (Aulton et al 1977). Wettability is assessed by measurement of contact angles, and the usual methods are the direct method (Harder et al 1970; Ehrhardt 1973; Fell & Efentakis 1979) and the h-ε method (Lerk et al 1976; Fell & Efentakis 1979).

In the direct method a small drop is placed on the surface of the solid compact and its contact angle measured. In the h–ε method the maximum height of a large drop is measured and an equation that allows for the porosity of the compacts used. Thus in comparing contact angles derived by the two methods some discrepancy may arise owing to the fact that in the h–ε method the porosity is taken into account, whereas in the direct method it is neglected. Limited work (Fell & Efentakis 1979) has been done to compare the results obtained by the two different methods.

For surfaces in which the pores cannot be penetrated (contact angles higher than 90°), porosity can be taken into account by applying the following equation (Johnson & Dettre 1969; Adamson 1976):

$$\cos \upsilon_a = f_1 \cos \upsilon_t - f_2 \qquad (1)$$

where υ_a = apparent contact angle; υ_t = true contact angle; f_1 = solid surface fraction of compact; f_2 = void surface fraction of compact.

By assuming that surface fraction can be substituted with volume fraction (porosity), as already suggested by other authors (Lerk et al 1976), we can write equation 1 as:

$$\cos \upsilon_a = (1 - \varepsilon_v) \cos \upsilon_t - \varepsilon_v \qquad (2)$$

where ε_v = compact volume porosity. Equation (2) can be rearranged into:

$$\frac{\cos \upsilon_a}{1 - \varepsilon_v} = \cos \upsilon_t - \frac{\varepsilon_v}{1 - \varepsilon_v} \qquad (3)$$

If equation (3) holds, then a plot of

$$\frac{\cos \upsilon_a}{1 - \varepsilon_v} \text{ versus } \frac{\varepsilon_v}{1 - \varepsilon_v} \text{ should be}$$

linear and the intercept equal to the cosine of the true contact angle. Furthermore, at each compact porosity, the apparent contact angle could be transformed into the true value by the following equation, derived by (3):

$$\cos \upsilon_t = \frac{\cos \upsilon_a + \varepsilon_v}{1 - \varepsilon_v} \qquad (4)$$

Methods

Compacts of magnesium stearate (Farmitalia Carlo Erba, Italy) and Eudragit RL (Rohm Pharma, Germany) were prepared by compressing an appropriate weight of powder in a single 1·128 cm diameter flat punch press (Nassovia, Germany), instrumented with piezoelectric load washers (Kistler, Switzerland).

Solid/water contact angles were measured with a Wettability Tester (Lorentzen-Wettre, Sweden). Small drops of demineralized water were placed on the surface of the sample compacts by means of a microsyringe and, after stabilization, the magnified images of drops were projected onto a screen. The contact angle was derived, via a trigonometric relationship, from the height and length of the base of the drop image. At least 5–10 replicates were carried out.

The porosity of compacts was determined from the apparent tablet density (derived from the tablet weight and dimensions) and the powder density (measured with an air-comparison pycnometer, Beckman, USA).

Results and discussion

The apparent water contact angles measured on magnesium stearate and Eudragit RL compacts prepared at different pressures were plotted according to equation

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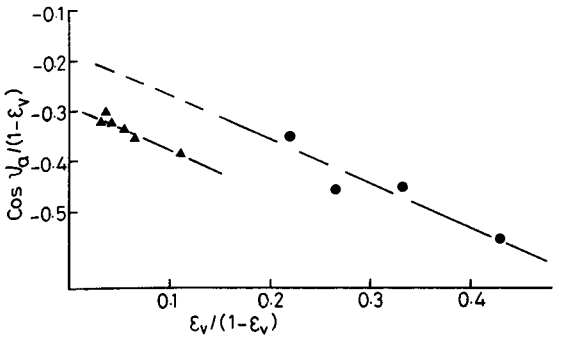

FIG. 1. Plot of apparent water/magnesium stearate (–▲–) and water/Eudragit RL (–●–) contact angles versus porosity, according to equation 3. Abscissa: $\varepsilon_v/(1-\varepsilon_v)$ dimensionless. Ordinate: cos $\upsilon_a/(1-\varepsilon_v)$ (dimensionless). Each point is the mean of 5–10 replicates (c.v. ≤0·05 for magnesium stearate and ≤0·12 for Eudragit RL).

(3) (see Fig. 1): the resulting curves were linear, confirming the validity of the proposed equation. The correlation coefficients r were 0·9576 (d.f. = 24, significant at $P < 0{\cdot}01$) and 0·82308 (d.f. = 17, significant at $P < 0{\cdot}01$) respectively. Another confirmation is given by the value of the slope of the straight lines, which proved to be not significantly different from −1 for both the samples. The true contact angles values, derived from the intercepts, were respectively 106° 37′ for magnesium stearate and 101° for Eudragit RL; both the values were in fair agreement with the true contact angles derived at each porosity via equation (4), as shown in Table 1, further confirming the validity of the proposed approach.

It remains to be stressed that equations (3) and (4) can be applied only to solids with contact angles higher than 90° and with pores on the compact surface that cannot be penetrated by the drop liquid: these solid surfaces are called 'composite' (Johnson & Dettre 1969). This means that for any non-wettable powder sample under examination, the compaction pressure range, i.e. the porosity range within which good linearity parameters are found for equation 3, must be carefully checked. For example, in the case of Eudragit RL, all the compacts prepared at pressures higher than 137·2 MNm^{-2} gave apparent contact angles which did not obey equation 3, due to the fact that only below this pressure does the Eudragit RL compact surface became 'composite'.

Table 1. Influence of porosity on water/magnesium stearate and water/Eudragit RL contact angles.

	Compaction pressure MNm^{-2}	Compact porosity	Apparent contact angle	True contact angle[a]
Magnesium Stearate	27·9	0·100	110° 18′	105° 55′
	54·8	0·060	109° 30′	106° 56′
	83·6	0·051	108° 39′	106° 27′
	138·7	0·039	108° 7′	106° 26′
	193·6	0·035	107° 9′	105° 37′
	243·1	0·031	108° 14′	106° 55′
Eudragit RL	54·3	0·30	112° 45′	97° 8′
	74·9	0·25	109° 52′	96° 53′
	99·2	0·21	111° 12′	101° 4′
	137·2	0·18	106° 42′	97° 32′

[a] derived from the apparent contact angles via equation 4.

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