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Solid bridge formation in sulphonamide-Emdex interactive systems

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Abstract

Solid bridge formation in model sulphathiazole and succinylsulphathiazole-Emdex® interactive systems was studied. Changes in adhesion during fluidized column drying were attributed to solid bridge formation caused by the deposition of dissolved material between the adhered sulphonamide particle and carrier surface reinforcing the interaction. The formation of the solid bridges was dependent on the moisture content of Emdex® in the predried mixture with adhesion reaching a maximum at a moisture content of 10.5%. The capillary interaction which occurred in the moist mixtures and preceded the solid bridge formation was generally stronger than the adhesion due to solid bridging. Consideration of the adhesion profile changes and interactive behaviour during storage provided indirect evidence of solid bridge formation. Both capillary interaction and solid bridging were influenced by the particle size of the drug with the smallest particles being most strongly adhered.

Keywords: Interactive mixing; Ordered mixing; Particle adhesion; Solid bridging; Capillary interaction

1. Introduction

The adhesion of drug particles onto pharmaceutical carriers involves several different mechanisms (Rumpf, 1961; Krupp, 1967; Zimon, 1982). Electrical interactions comprise (a) contact potential forces which are due to the difference in work function between contiguous uncharged dissimilar materials and (b) Coulombic forces due to the interaction of charged materials with surfaces. Non-electrical interactions include (a) intermolecular forces which occur between closely contacted surfaces due to Van der Waals forces, (b) capillary forces caused by the formation of liquid bridges between surfaces and (c) solid bridging between surfaces occurring by mechanisms such as melting and crystallization. At any time the total adhesion force in an interactive mixture will depend on the relative magnitude of the individual force components (Stewart, 1986).

Electrical, capillary and intermolecular interactions between particles have a considerable

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theoretical basis (Krupp, 1967; Zimon, 1982) and their application is well understood in some industries. Solid bridging has not received the same attention but offers a way to achieve strong, permanent bonding of drug particles to pharmaceutical excipients. The development of interactive systems using controlled solid bridging has the potential to overcome the variability in the degree of interaction between drug and surface due to charge decay with subsequent decrease in the electrical force component (Kulvanich and Stewart, 1987a, 1988) and due to change in capillary interaction with moisture content. The ability to form strong permanent interactions through controlled solid bridging would improve the design of these systems and would increase their potential uses. Preliminary studies in our laboratories revealed sulphonamides were strongly bound to water soluble carriers when moist interactive mixtures were dried and a hypothesis was proposed to explain the interaction through solid bridge formation (Padmadisastra et al., 1994). The purpose of this research was to investigate further solid bridge formation through the crystallization of solids in liquid films separating adhered drug particles and the surfaces of pharmaceutical carriers and to study factors which might influence the degree of interaction.

2. Materials and methods

2.1. Materials

Sulphathiazole and succinylsulphathiazole (Sigma, U.S.A.) were used as the adherent drugs. Emdex® (Mendell, U.S.A.) was the carrier. The drugs were micronized by fluid energy milling using a Chrispro Jetmill (U.K.), model 75P with filtered compressed air at 5.8 atm and 12.7 l s^{-1}. Other particle size drug fractions were classified using a Sonic Sifter, model L3P (ATM Corp., U.S.A.) and micro mesh sieves. Typical particle volume mean diameters for succinylsulphathiazole were micronized (10.9 μm), 10–30 μm (27.7 μm) and 30–45 μm (45.9 μm). Particle size fractions of the carrier were classified using a Pascal sieve shaker (Pascal, U.K.) and Endecott Test Sieves. All classification procedures were carried out at 22 ± 1°C and 50% relative humidity (RH). Drugs and carrier were stored in a desiccator containing silica gel. Other chemicals used were analytical grade and all chemicals were used as supplied from the manufacturers.

2.2. Preparation of interactive mixture

10 g of a 1% sulphonamide interactive mixture was prepared in a glass jar rotated at 25 rpm for 15 min on a friability tester (Erweka, Germany). The jar was positioned at an angle of 40° to the vertical; this position provided optimum blending conditions. The formation of an interactive system in which the drug particles were adhered to the carrier surface and in which few unattached drug particles were observed was verified by scanning electron microscopy using a Philips, model 505 SEM (Philips, U.K.). Homogeneity of the mixture was determined using 20 × 50 mg samples and the coefficient of variation of the mixes was less than ±3% indicating satisfactory mixing (Cook and Hersey, 1974; Crooks and Ho, 1976).

2.3. Adhesion measurement

A specially designed aluminium centrifuge cell consisting of a sample and collection compartment separated by a replaceable screen (150 μm) was held in position within the centrifuge rotor so that the screen was normal to the axis of rotation (Kulvanich and Stewart, 1987b). The percentage of drug retained on the carrier was determined after centrifuging in a microprocessor-controlled, high speed centrifuge (International Equipment Co., U.S.A.) with 895 rotor. The temperature in the centrifuge chamber was 20°C and the interactive mixture sample size was accurately known (40–70 mg). Adhesion profiles (i.e., % retained vs centrifuge speed) were determined by centrifuging at 2000, 5000, 10 000, 15 000 and 20 000 rpm for 30 s. For some experiments, the degree of adhesion was determined by the percent of drug retained on the carrier after centrifuging at 20 000 rpm for 30 s.

2.4. Drug analysis

The amount of sulphonamide detached after centrifugation and the sulphonamide retained on the carrier were assayed spectrophotometrically. The sulphonamides were extracted into 0.1 M NaOH and the absorbance measured at the wavelength of maximum absorption on a Pye Unicam PU8600 (U.K.) ultraviolet visible spectrophotometer (i.e., 255 nm for both sulphathiazole and succinylsulphathiazole). Spectra were determined using a double-beam Varian DMS 100S (U.S.A.) ultraviolet visible spectrophotometer. Beer's law standard curves were prepared at the wavelength of maximum absorbance using four concentrations and four replicates over the concentration range 0.25–5.0 mg%. There was no significant deviation from linearity and the sulphonamide concentrations were obtained by inverse prediction (Williams, 1959). Dissolved carriers did not interfere with the absorbance measurement of the sulphonamides. The coefficient of variation of the assay was $\pm 1.3\%$ ($n = 20$) for all drugs and extraction methods were shown to recover all of the drugs.

2.5. Moisture content

Specific moisture contents of the carriers were obtained by storage in controlled relative humidity conditions for predetermined times. Saturated salt solutions were used to maintain constant relative humidity conditions inside small desiccators incubated at 20 ± 1°C (Winston and Bates, 1960). The following saturated salt solutions were used: potassium nitrate (95%), potassium chloride (85%), sodium chloride (75%), ammonium nitrate (63%) and magnesium chloride (32%). Relative humidity was monitored within the desiccator using a Humidity Probe (Hanna Instruments, Italy) with variability being $< 3\%$. The moisture content of the carriers was determined by drying the sample at 100°C to constant weight.

2.6. Controlled drying of the interactive mixtures

The interactive mixture was dried in a vertical glass column (3 cm in diameter and 30 cm in length) possessing a sintered glass frit at the bottom to contain the mixture. Air heated using a glass coil (0.7 cm internal diameter, total length of coil 20 cm, coil diameter 5.5 cm) contained within a Selsius oven (Townson & Mercer, Australia) was passed through the glass frit causing fluidization of the mixture. The temperature of air just above the glass frit was adjusted to 60°C. The flow rate was adjusted using a valve on the air pump (Iwaki model AP, Japan) and a flow meter (Halu, Australia).

3. Results and discussion

3.1. Solid bridge formation

The hypothesis that solid bridge interaction occurred by crystal bridge formation between the adhered drug and the carrier surface and was caused by evaporation of the moisture in the liquid bridge with the subsequent crystallization of dissolved carrier was proposed (Padmadisastra et al., 1994). In that study, oven drying at 100°C produced solid bridging but was not entirely satisfactory as frequent agitation of the powder bed was necessary especially in the early stages of drying to prevent agglomeration of the interactive units and subsequent crust formation. Experiments using a microwave oven with combinations of different drying times and power settings resulted in sample degradation and severe crust formation. A laboratory fluidized bed drying column was developed to provide mild agitation to the mixture during drying to overcome agglomeration tendencies. A pump circulated heated air through a glass coil positioned in an oven at a flow rate up to 16.0 l min^{-1} causing fluidization of the drying mixture.

Care was required in the selection of the flow rate as high air velocity dislodged the drug particles from the carrier surface and transfered the particles out of the open ended column, e.g., at initial flow rates greater than 6 l min^{-1}, about 50% of succinylsulphathiazole was lost from the column. Experimentation revealed that a low initial flow rate of 1.2 l min^{-1} for 20 min followed by 10.0 l min^{-1} until dry prevented drug loss

from the column. For the moisture contents of the interactive systems used in this study, a 2 h drying time at 60°C produced equilibrium moisture contents of less than 1.0%.

3.2. Influence of moisture content

The degree of adhesion was determined for Emdex mixtures prepared at moisture contents over the range 6.1–11.4% using micronized and 45–53 μm size fractions of sulphathiazole and succinylsulphathiazole. The interactive systems were column dried and the degree of adhesion remeasured. The results of the sulphathiazole-Emdex interaction are shown in Fig. 1 and represent typical behaviour of the interactive systems studied.

The degree of adhesion of the mixtures freshly prepared using the 45–53 μm drug fraction increased from 55% drug retained at 7.5% moisture content to about 90% retained at 10.5% moisture content. The column-dried sample also showed an increase in adhesion as the moisture content of the Emdex increased ranging from about 10–20% retained at low moisture contents to about 70% retained at 10.7% moisture content. The percent retained for the dried samples was always less than that of the original mixtures. The interactive mixture produced using the micronized sulphathiazole fraction exhibited high adhesion over the range of moisture contents as expected from the decreased detachment force due to the decreased particle mass, and reached a plateau at about 10.5% moisture content, e.g., after drying the percent retained was around 87% at the lower moisture contents (< 8.6%) but increased to about 95% at 10.5% moisture.

For the freshly prepared interactive mixtures, the adhesion changes with moisture content can be explained by a consideration of the electrical and capillary force balance. Electrical forces particularly contact potential forces will be the primary interactive forces predominating in the initial interaction between drug and carrier and will be maximized in dry conditions (Zimon, 1982). These forces will decrease with time due to charge decay at a rate dependent on the moisture content of the mixture and the surrounding atmosphere (Zimon, 1982). Such behaviour has been observed for the sulphonamides used in model interactive systems (Kulvanich and Stewart, 1987a). Capillary interaction will occur due to the condensation of water in the spaces separating the contiguous surfaces forming a liquid bridge and is compounded from the surface tension and a pressure differential forces (Zimon, 1982). Capillary interaction should be more predominant as

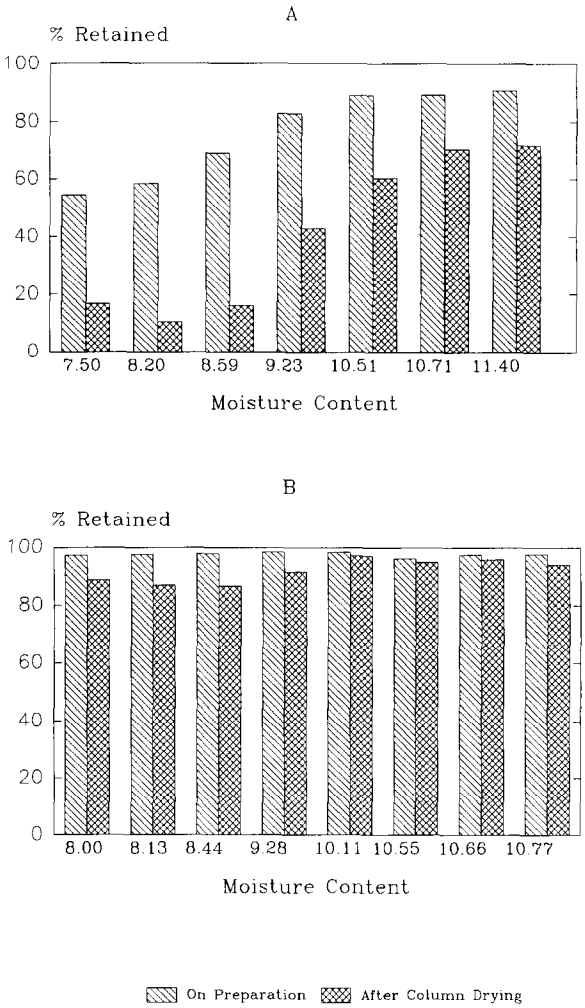

Fig. 1. Influence of moisture content on the degree of adhesion of sulphathiazole (1%)-Emdex® (250–425 μm) interactive mixtures, for sulphathiazole in 45–53 μm (A) and micronized (B) fractions determined immediately after preparation of the interactive mixture and after fluidized column drying.

the moisture content of the system increases. For the interactive system studied in Fig. 1, in spite of any decrease in interaction due to charge decay as the moisture content increased, there was a significant increase in the adhesion especially in the 45–53 μm sulphathiazole and succinylsulphathiazole interactive systems and this effect would be consistent with an increase in capillary interaction hypothesis.

The purpose of the column drying was to initiate solid bridge formation between the drug and carrier. At higher moisture contents where capillary condensation produced significant liquid bridges between the adhered drug and carrier surface, dissolution of the water soluble Emdex® can occur. During drying, evaporation of the moisture will cause the pendular ring to shrink and the dissolved Emdex® to crystallize resulting in the formation of solid bridges between the particle and surface. The increase in adhesion of the dried interactive mixtures with increased initial moisture content was attributed to solid bridging. At low moisture contents (< 10.5%) there was insufficient moisture to produce solid bridges or solid bridges which were sufficiently strong to withstand the rigours of fluidized drying or the centrifugation procedure at 20000 rpm. A critical moisture content is required for the formation of solid bridges.

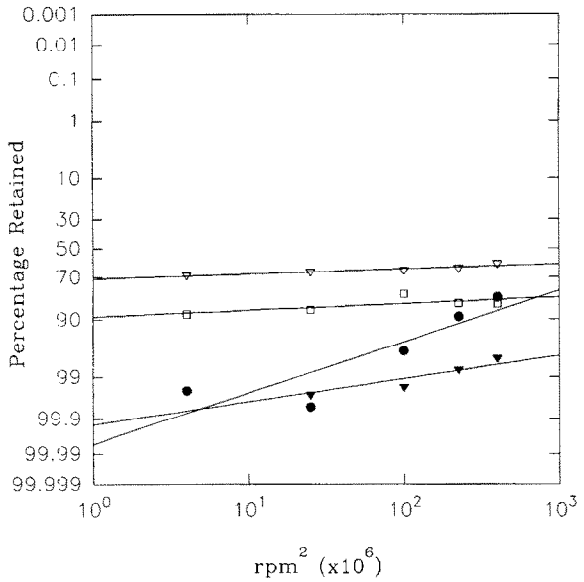

Fig. 2. Adhesion profiles of percent retained vs square of the centrifuge speed for sulphathiazole and succinylsulphathiazole (1%, 45–53 μm)-Emdex® (250–425 μm) interactive mixtures. (●) Sulphathiazole, determined immediately after preparation of the interactive mixture using Emdex® with 10% moisture content; (▽) sulphathiazole, after fluidized column drying; (▼) succinylsulphathiazole, determined immediately after preparation of the interactive mixture using Emdex® with 10% moisture content; (□) succinylsulphathiazole, after fluidized column drying.

3.3. Confirmation of solid bridge formation

Direct proof of crystal bridge formation in interactive systems was difficult to obtain. Scanning electron microscopy showed ample evidence of an interactive system with the drug particles clearly evident on the surface of the carriers. However, it was not possible to observe crystal bridge formation because of the system's geometry and orientation. Indirect evidence was therefore sought.

Adhesion profiles have been used to characterize the degree of interaction between drugs and carriers (Kulvanich and Stewart, 1987b). The adhesion profiles of sulphathiazole and succinylsulphathiazole (1%; 45–53 μm size fraction)-Emdex® interactive mixtures were determined immediately after mixing with Emdex® equilibrated to 10.5% moisture and after column drying (Fig. 2). The profiles showed some variability at high values of percent retained since the amount of drug detached from the carrier was extremely small and concentrations approached the lower quantifiable limits of the assay. In spite of this, the profiles demonstrated marked differences in the pattern of the degree of adhesion for the interactive systems before and after drying. While the sulphonamide particles were strongly adhered to the Emdex® carrier prior to drying, the shape of the profile supported a distribution of forces, i.e., for the sulphathiazole the percent retained ranged from 99.5 at 4×10^6 rpm^2 (i.e., 2000 rpm) to 81.1 at 400×10^6 rpm^2 (i.e., 20000 rpm). The capillary interactive force between a particle and a planar surface for dissimilar materials but with similar contact angles between the

surfaces and liquid can be expressed (Zimon, 1982):

$$F_c = 4\pi\gamma r \cos\Theta \quad (1)$$

where F_c is the capillary interactive force, r denotes the particle radius, γ is the surface tension and Θ represents the contact angle between the particular solids and liquids. While this is a simplistic model of capillary interaction, a force distribution can be expected for multiparticulate drug adhesion. The centrifugal detachment force (F_{det}) is:

$$F_{det} = (\pi d^3 \rho a/6) \quad (2)$$

where d is the adhered particle diameter, ρ represents the particle density and a is the centrifugal acceleration, and therefore will also be dependent on the adhered particle's size. By contrast, the adhesion profiles for the dried systems were very flat with the percent retained changing slightly over a wide speed of detachment. The shape of the profile suggested that a small fraction of weakly adhered drug was easily removed at low detachment speeds. The remainder was strongly adhered to the carrier and not removed even at the highest detachment speeds. The formation of an interactive system with crystal bridges strongly bonding the drug particles to the carrier was plausible given the profiles obtained in Fig. 2.

One possibility that must be considered during column drying was that crystal bridging did not occur and that the strong sulphonamide adhesion was effected by electrical adhesion enhanced by increased particle contact during the column fluidization. If electrical adhesion were responsible for the bonding after the drying, the flat adhesion profile obtained in Fig. 2 would be unlikely to occur. Ample evidence is available in the literature to suggest that adhesion profiles which are dominant in electrical interaction display a wide distribution of forces (Zimon, 1982; Kulvanich and Stewart, 1987b).

Further evidence of solid bridge formation is shown in Fig. 3 where the degree of adhesion of three particle size fractions of succinylsulphathiazole and sulphathiazole in an Emdex® interactive mixture was studied immediately after preparation, after column drying and during storage of the dried mixture for 7 days. For both drugs the degree of capillary interaction for the freshly prepared mixtures was high, especially for the smaller particle size fractions. The micronized drug interactive mixtures showed little adhesion change on drying or storage, reflecting strong bonding of the drug to the carrier. The 10–30 and 30–45 μm drug fractions showed a proportional decrease in adhesion on drying but remained constant during storage. This behaviour strongly supports the formation of an interactive

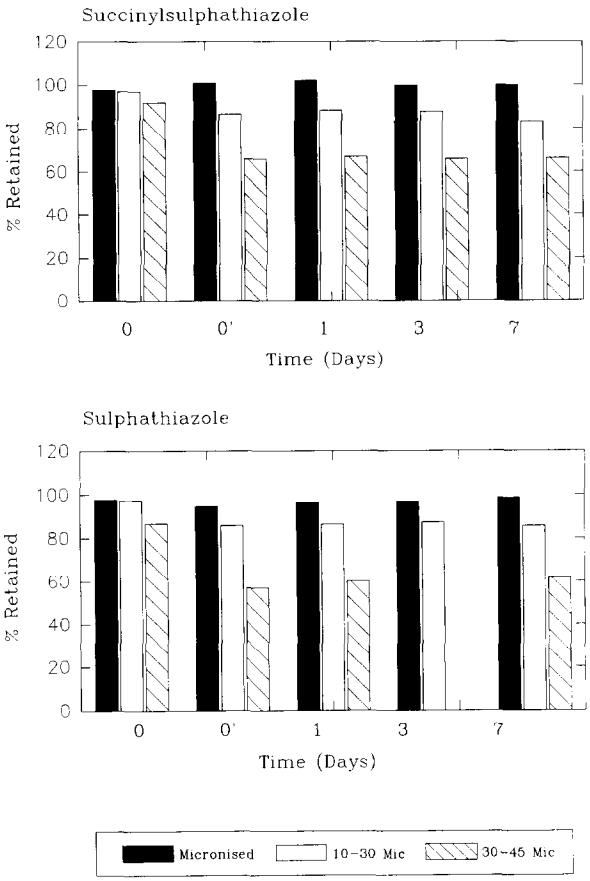

Fig. 3. Influence of storage time on the degree of adhesion of sulphathiazole and succinylsulphathiazole (1%; micronized, 10–30 and 30–45 μm)-Emdex® (250–425 μm) interactive systems. O indicates immediately after preparation using Emdex® with a 10% moisture content; O′ indicates immediately after fluidized column drying. Data for the 3 day time point for the 30–45 μm fraction were not available.

system through solid bridging as bonding through electrical interactions would have resulted in some degree of charge decay and reduction in the degree of adhesion during storage.

3.4. Influence of particle size

The effect of particle size on the adhesion of the freshly prepared and column dried interactive mixtures was seen in the previous studies and is demonstrated in Fig. 1 and 3. A further study to examine the influence of sulphonamide particle size on the adhesion of these interactive systems was undertaken (Table 1). Differences in the magnitude of the particle size effects in the various studies described in this paper were attributed to differing moisture contents of the freshly prepared mixtures. For example, for the data shown in Fig. 3, the initial moisture content of the mixture was $7.7 \pm 0.8\%$ while that of the mixture in Table 1 was $9.8 \pm 0.6\%$. These differences will affect the degree of capillary and solid bridge interaction. An analysis of variance of the data in Table 1 showed that there was no significance difference between the adhesion behaviour of the two drugs ($P = 0.12$) but that there were significant differences in adhesion between the freshly prepared and column dried mixtures ($P = 0.002$) and between the sulphonamide particle sizes ($P = 0.001$). The effect of drug particle size on the adhesion of the freshly prepared mixtures was not marked although decreased adhesion occurred with increase in sulphonamide particle size. Adhesion measurements after column drying showed a greater particle size effect especially for succinylsulphathiazole. Particle size of adhered drug will affect the magnitude of adhesion due to its contribution to the detachment and interactive forces. As the drug particle size increases, the relative detachment force will increase markedly due to the greater mass of the particle (Table 1). For the freshly prepared mixture, the drug particle interaction with the carrier surface should occur predominantly by capillary interaction. Eq. 1 would predict a linear increase in capillary interaction with particulate diameter and calculated relative values are shown in Table 1. A consideration of the force balance between detachment and capillary adhesion would indicate that larger drug particles should be extremely

Table 1
Influence of drug particle size of the adhesion of succinylsulphathiazole and sulphathiazole Emdex® interactive mixtures [a]

	Freshly prepared [b]			After column drying		
	mic [c]	10–30	30–45	mic	10–30	30–45
Succinylsulphathiazole						
Adhesion [d]	97.6 ± 3.3.	97.6 ± 2.0	92.3 ± 1.6	97.1 ± 2.1	90.5 ± 2.5	83.3 ± 4.0
Relative detachment force [e]	1.0	16.4	79.7	1.0	16.4	79.7
Relative capillary force [f]	1.0	2.5	4.3	–	–	–
Sulphathiazole						
Adhesion	97.8 ± 1.3	96.9 ± 1.2	94.7 ± 4.5	96.3 ± 1.3	91.9 ± 1.8	91.9 ± 4.1
Relative detachment force [e]	1.0	19.3	72.9	1.0	19.3	72.9
Relative capillary force [f]	1.0	2.7	4.2	–	–	–

[a] Emdex® size fraction was 250–425 μm.
[b] Moisture content of Emdex® was 10.5%.
[c] Volume mean diameters were as follows: succinylsulphathiazole – 10.9, 27.7 and 46.9 μm for mic, 10–30 and 30–45 μm fractions; sulphathiazole – 10.1, 27.1 and 42.2 μm, for mic, 10–30 and 30–45 μm fractions.
[d] Percent retained at 20000 rpm.
[e] Detachment force is defined as $\pi d^3 \rho a/6$, i.e., ma, where d is the drug particle diameter, ρ denotes the density, m is the particle mass and a represents the centrifugal acceleration. The relative detachment force was the ratio of the calculated detachment force relative to the smallest particle diameter assuming that the surface tension and contact angle remain constant.
[f] Capillary force is defined as $4\pi\gamma r \cos\Theta$, where γ is the surface tension, r the particle radius and Θ the contact angle. The relative capillary force was the ratio of the capillary forces relative to the smallest particle diameter.

prone to detachment during centrifugation (Table 1). While a trend towards decreased detachment is seen in Fig. 1 and 3 and Table 1, the reduction in the degree of adhesion was not consistent with the force balance. For example, as particle size of adhered drug increased from micronized to a 30–45 μm fraction, the relative force balance should favour detachment by about 17–18 times. This was not observed in the data with relative changes in adhesion over this particle size range being no more than a decrease to 60%. The systems were clearly more stable than the force balance indicated. The increased stability could result from contribution of other than capillary interactive forces, i.e., intermolecular. The surface rugosity of Emdex® could result in mechanical interlocking of the adhered particles producing stronger interaction. The theoretical detachment force calculations also are approximations, since the real detachment process would need to consider the interactive system's geometry with particle detachment configurations that are not normal to the carrier surface, the real multiparticulate nature of the adhered drug in the specific size fractions used and the possibility of detachment of surface agglomerates of adhered particles (Kulvanich and Stewart, 1987c). Particle size also has a significant effect on the degree of adhesion of the dried mixture and clearly influences the solid bridging process. This would be expected since the smaller particles should have closer contact with the carrier surface and have a greater surface contact with liquid bridges and subsequently with the formed solid bridges.

4. Conclusion

Solid bridge adhesion in interactive systems has been studied using a fluidized drying column to produce crystal bridges between adhered sulphonamide particles and an Emdex® carrier. The flatness of the adhesion profiles and the lack of charge decay of the dried mixtures during storage strongly support the formation of an interactive mixture through solid bridging. The adhesion studies clearly indicate that a small fraction of the sulphonamide particles are not strongly bonded to the carrier surface. Optimization of the drying process is thus necessary to achieve more stable systems. Further studies are necessary to understand the solid bridging process more fully, e.g., drug and carrier dissolution behaviour in the liquid bridge, the characteristics of the solid bridge formed during drying and factors which influence the strength of the solid bridges. An understanding of solid bridging would allow the formation of extremely stable interactive mixtures capable of effectively delivering drugs as stand alone dosage forms or after further processing.

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