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Influence of Relative Humidity on the Adhesive Properties of a Model Interactive System

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Abstract—The influence of relative humidity on the adhesive properties of model drug-carrier interactive systems was investigated using the centrifuge method. The degree of adhesion was quantified by determining the S50 from the adhesion profile. High ambient humidity (greater than 40% r.h.) decreased the degree of interaction of three sulphonamide powders with model hydroxypropyl methylcellulose phthalate-coated glass bead carriers during blending. All drug-carrier interactive systems displayed decreases in the degree of adhesion during storage. The extent and rate of the decreases increased with relative humidity and was interactive system-dependent. Preconditioning the powders to varying moisture contents produced contrasting results during blending, with one system showing a decrease in adhesion as the relative humidity increased, while the other's interactive capability increased. This increase was attributed to the enhancement of charging sites on the drug's powders surface by the adsorbed moisture.

Moisture in powders and the environment influences interactive behaviour during handling and operations in two ways. At low levels of humidity, the electrostatic effects markedly influence the degree of interaction, whereas at high atmospheric moisture the capillary force due to water vapour condensation at the interface of particle becomes more dominant.

Relative humidity was found to play an important role in controlling electrical charge generation and decay of powders; high humidity promoted rapid dissipation of electrical charge accumulated on a material's surface (Boschung & Glor 1980). The rapid leakage of electrical charge at a high level of atmospheric moisture was attributed to an adsorbed moisture film on the material causing an increase in the electrical conductivity of the material's surface and surrounding atmosphere (Cleves et al 1971; Boland & Geldart 1971; Hannsson & Jonassen 1978; Jonassen et al 1979). The measurement of surface electrical conductivity at various relative humidities has revealed a critical range of relative humidities at which the electrical resistance was reduced dramatically and rapid electrical charge dissipation occurred. However, materials with different water affinities exhibited different critical ranges which varied from 25 to 70% relative humidity (Boland & Geldart 1971). Triboelectrification between two dissimilar materials under high atmospheric moisture occurred but was less apparent due to simultaneous charge dissipation (Boland & Geldart 1971).

The relative humidity level at which water vapour condensation formed capillary bridging with subsequent interaction was different in different adhesive systems (Corn 1961; Zimon 1963) and was a function of the nature of adherent, ambient humidity, temperature, and atmospheric pressure (Coelho & Harnby 1978a). The humidity level at which capillary interaction occurred can be lower than the critical relative humidity at which liquid bridge was formed if the absorbed moisture film was thick enough. For a given relative humidity, the thickness of the adsorbed layer of open material would be decreased if the particle surface became more irregular (Coelho & Harnby 1978b).

Investigations of the effect of relative humidity on particle adhesion based on particle-plane or particle-fibre adhesive systems have been reported (Kordecki & Orr 1960; Corn 1961; Zimon 1963; Corn & Stein 1965; Loffler 1968). The observations generally indicated an increase in the adhesion tendency with increased relative humidity. However, some adhesive systems showed little increase in adhesion with relative humidity; some showed a sharp increase in capillary interactions when critical relative humidity levels were reached. The critical humidity levels varied from 50 to 80% depending on the nature of the adhesive systems investigated. Capillary interaction did not take place immediately after contact with the substrate but required some time to be generated (Zimon 1963). The larger particle size adherents generated stronger capillary forces than smaller sizes at the same relative humidity (Corn 1961). However, the magnitude of capillary forces determined experimentally was smaller than theoretical values (Zimon 1982).

The effect of the moisture content on the interaction properties of mixture components during mixing operations has been reported (Karra & Fuerstenau 1977; Stephenson & Thiel 1980; Thiel & Stephenson 1982). The degree of interaction was lowest at intermediate humidity levels (40–50%). Almost all the investigations of particulate interactions under the influence of humidity in pharmaceutical system have been performed in relation to the cohesive properties of the bulk powder (Shotton & Harb 1966; Pilpel 1970; Eaves & Jones 1970; 1972a, b; Fukuoka et al 1983).

Materials and Methods

Materials

Glass beads (500 μm, Selby Scientific, Australia) coated with hydroxypropyl methylcellulose phthalate (HPMCP, Type HP-55, Shin-Etsu Chemical Co., Japan) using an air suspension technique (Uni-Glatt, laboratory unit, Glatt GmbH, FRG; 5% w/v HPMCP in equal volumes of dichloromethane and methanol, 3 L kg^{-1} beads) were used as the carriers.

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The coated beads were oven-dried at 50°C for 24 h to eliminate residual solvent.

The following drugs (Sigma Chemical Co., USA) were used to form the interactive mixtures: sulphapyridine ($d_v = 27{\cdot}2$ μm, $\sigma = 1{\cdot}1$ μm), sulphamerazine ($d_v = 17{\cdot}7$ μm, $\sigma = 1{\cdot}4$ μm), succinylsulphathiazole ($d_v = 23{\cdot}4$ μm, $\sigma = 1{\cdot}6$ μm).

Particle size classification and measurement

The size fractions of drug powder were prepared using the oscillating air column method of sieving (Sonic Sifter, model L3P, ATM Corporation, USA) fitted with micromesh sieves and a horizontal pulse accessory (model L3-N8). The particle size distributions of sulphapyridine, sulphamerazine, and succinylsulphathiazole powders were determined by a laser diffraction technique (Malvern 2600/3600, Malvern Instrument, UK) using water as the suspending medium.

Preparation of interactive mixture

Carriers and drug powders were equilibrated in an Environmental Chamber (Thermoline Scientific Equipment Pty Ltd, Australia) at controlled relative humidity and temperature for 24 h. The formation of the interactive system was also performed in the chamber at the required relative humidity and temperature by blending 3 g of mixture in a glass jar at a rotation speed of 20 rev min^{-1} for 10 min. The glass jar was positioned at an angle of 42° to the vertical; this position provided optimum blending conditions. The formation of the interactive mixture was verified by observing the drug particles adhered to the carrier surface by microscopy. Samples of 100 mg were taken for adhesion measurements. The homogeneity of interactive mixes was determined using fifteen 50 mg samples; the coefficient of variation of the sample contents ranged between 1·5 and 3·9% for the mixes studied.

Storage of interactive mixes

Saturated salt solutions were used to maintain constant relative humidity levels inside small dessicators during the storage of interactive mixes and powders. The dessicator was incubated at a constant temperature of 25 ± 1°C. The following series of saturated salt solutions were used: sodium hydroxide (6%), magnesium chloride (33%), magnesium nitrate (53%), sodium chloride (75%) and potassium nitrate (94%) (Winston & Bates 1960). Relative humidity was measured within the dessicator using a Novasiha Humidity Probe (model MIK 2000) with the variability being $<3\%$.

Scanning electron microscopy

Examination of the carrier surface texture and the interactive mixes was performed by scanning electron photomicroscopy (Phillips, model 505 SEM, UK).

Adhesion measurement

A specially designed aluminium centrifuge cell consisting of a sample and collection compartment separated by a replaceable screen (250 μm) was held in position within the centrifuge rotor so that the screen was normal to the axis of rotation (Kulvanich & Stewart 1987a). Adhesion measurements were performed by means of a IEC B-20A high speed refrigerated centrifuge with a fixed rotor, type 870 (Damon/IEC Division, USA) which allowed rotation speed up to 19 000 rev min^{-1}. The temperature in the centrifuge chamber was 20–25°C. The drug particles removed were collected at the centrifugation speeds of 2000, 5000, 10 000, 15 000 and 19 000 rev min^{-1}. The rotor was accelerated to the desired speed which was maintained for 30 s before deceleration.

Analysis of drug

The amount of drug detached after each consecutive centrifugation step and the drug retained on the carrier were assayed spectrophotometrically. Complete solution of the drug was achieved in HCl (0·1 M) or NaOH (0·01 M) and the absorbance was measured at the wavelength of maximum absorbance using the Pye Unicam PU8600 spectrophotometer (Pye Unicam Ltd, UK). Beer's law calibration curves for all the drug materials over the concentration range 0·002 to 0·02 mg mL^{-1} showed no significant deviation from linearity and the drug concentrations were obtained by inverse prediction. The coating material did not interfere with the absorbance measurements during the analysis of the drugs on the carrier.

Results and Discussion

Degree of adhesion

The centrifuge method allowed the determination of the adhesion profile which was a logarithmic normal function when the percent of drug remaining on the carrier was regressed against the square of the speed of rotation (Kulvanich & Stewart 1987a). The profile could be characterized by the S50, i.e. the speed required to dislodge 50 percent of adherent particles, and σ, i.e. the geometric standard deviation of the adhesive distribution. In these experiments the total degree of adhesion of the drug particles in the interactive systems was measured by the S50 parameter. The concentration of all mixes was kept below 1% to minimize collision and layer formation effects (Kulvanich & Stewart 1987a).

Effect of ambient humidity during blending

The interactive mixes of sulphapyridine powder and polymer coated carrier were used to observe the adhesion tendency of drug particles when prepared at humidity conditions of 26, 40, 50, 65, and 80% at a constant temperature of 25°C. The carrier and drug compound were equilibrated within the environmental chamber at the required relative humidity and temperature for 24 h before blending for 10 min. The sample of mixture was immediately taken for adhesion measurement after preparation. S50 values of five replicate mixtures were determined at each humidity level.

The relationship between S50 and relative humidity at which the mixes were prepared is shown in Fig. 1. S50 values were highest at a relative humidity of 26%, showed a rapid and significant decrease (analysis of variance; $P < 0{\cdot}001$) in adhesion tendency at 50% r.h. and reached a minimum at 65–80%. The reduction of S50 with increasing relative humidity during blending can be explained by considering the forces involved in the adhesion process. Four types of forces are primarily important in the adhesion of small particles to surfaces; molecular forces, electrical forces including electrical double layer formation at contact points

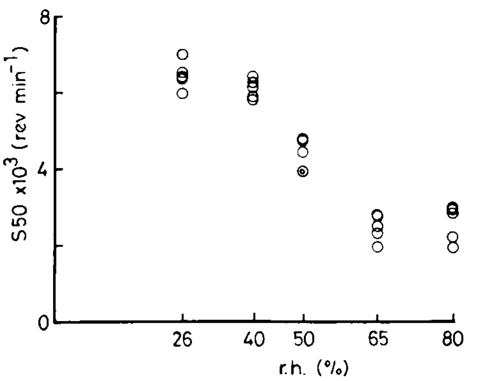

FIG. 1. Effect of relative humidity during blending on the adhesion tendency (S50) of a sulphapyridine interactive mixture.

and the Coulombic interaction of the electrical charge distributed over the particle surface due to previous electrifications, capillary forces due to the formation of liquid bridges, and interfacial forces including salt bridge formation, mechanical interlocking of particles and sintering effects (Krupp 1967; Derjaguin et al 1977).

While the molecular force is effective over the whole relative humidity range, the electrical double layer interaction contribution occurs up to intermediate humidity conditions until charge leakage begins at the interface. Coulombic interaction which will occur at low to intermediate humidity level will increase the adhesion of the interactive system, depending on the degree of static electrification of the adherents and the treatments of the materials. At high humidity levels, the capillary force should become the main component of adhesive forces. The critical humidity between 40–50% in Fig. 1 at which the rapid reduction of S50 occurred corresponded with the humidity range at which sudden increases in electrical conductivity of the materials and surrounding atmosphere have been observed. There was little evidence that the capillary interaction dominated at relative humidities between 65 and 80%. If substantial capillary interaction occurred at relative humidities between 65 and 80% an increase in the adhesion tendency would be observed following the reduction of S50 at intermediate humidities. Such a result was not unexpected since the adhesion measurements were performed immediately after interactive system preparation when the liquid bridges between surfaces were probably not well established.

Effect of storage relative humidity

Interactive systems of three drugs sulphapyridine, sulphamerazine and succinylsulphathiazole were prepared by blending for 10 min at relative humidity and temperature of 26% and 25°C, respectively. The interactive mixes then were placed in crucibles and stored in dessicators which were conditioned to various relative humidities (6, 33, 53, 75, and 94%) by saturated salt solutions at constant temperature (25°C). Samples of mixtures were then taken at time intervals for adhesion measurements. Determination of the degree of adhesion of the interactive mixes was performed over an 11 day period. In the following systems, the experimental design was not conducive to statistical analysis. The compromised design was necessary because of the long time associated with the adhesion measurements and the need to allow simultaneous decay patterns to be determined to minimize experimental time.

Sulphapyridine interactive system. Fig. 2 shows the relation between S50 of the sulphapyridine interactive mixes and time of storage at different humidity conditions. S50 values at time zero were obtained for the freshly prepared mixes before storage. The S50 of the mixes kept at 53, 75, and 94% r.h. were markedly reduced after one day's storage whereas that of the mixes kept at 6 and 33% showed a gradual decrease over the 11 days. The S50 profiles of the mixes kept at 6 and 33% decreased at about the same rate. The average drug concentration of the samples taken from each mix kept at 6 and 33% r.h. for adhesion measurement were 0·69 ± 0·03 and 0·70 ± 0·02%, respectively. A significant dislodgement of drug particles before the adhesion determinations was observed in the samples taken from the mixes stored at 53, 75, and 95% r.h., e.g. after one day the adhered powder concentrations reduced to 0·58, 0·33, and 0·36%, respectively. The loss of drug powders might have occurred during storage or while the samples were taken for the adhesion measurements due to the decreased adhesion.

Fig. 3 shows the change of adhesion tendency of the sulphapyridine mixes at similar humidity conditions when re-examined during short term storage. The mixes kept at 6 and 33% r.h. were stable and showed little decrease in adhesion tendency over 9 h. Further observations of adhesion tendency of these mixtures (kept at 6 and 33% r.h.) at

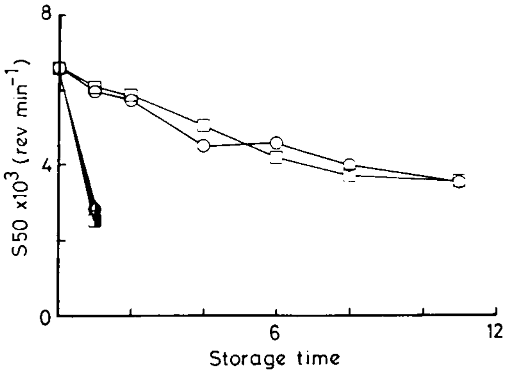

FIG. 2. Effect of relative humidity on the adhesive tendency (S50) of sulphapyridine interactive mixtures during storage over 11 days. ○ = 6% r.h.; □ = 33% r.h.; ◑ = 53% r.h.; ◨ = 75% r.h. and △ = 94% r.h.

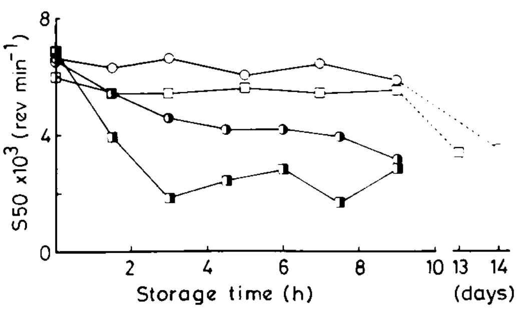

FIG. 3. Effect of relative humidity on the adhesive tendency (S50) of sulphapyridine interactive mixtures during storage over 9 h. ○ = 6% r.h.; □ = 33% r.h.; ◑ = 53% r.h. and ◨ = 75% r.h.

day 13 and 14 showed decreases in S50 of the same order as previously observed in Fig. 2.

However, the mixes stored at 53 and 75% r.h. exhibited a rapid decrease in the S50 in the short period after preparation. The drug particles were weakly bound to the carriers in the mixes kept at 75% r.h. after storage time of 3 h. Only half the amount of drug was retained in the sample mixes taken for adhesion measurement at this time interval. The average concentrations of drug powder of all samples taken for adhesion measurement form each mixes kept at 6, 33, and 53% r.h. were 0·66±0·01, 0·55±0·01 and 0·63±0·03%, respectively.

Suphamerazine interactive system. Fig. 4 shows the change in adhesion tendency of sulphamerazine interactive mixes when kept at relative humidity of 6, 33, 53, and 75% for a period of 9 h. The mix stored at 6% r.h. showed no change in adhesion tendency with the time of storage. Little decrease in S50 of the mixes stored at 33 and 53% r.h. were found over 9 h; both showed similar adhesion reduction profiles. The mix kept at 75% r.h. exhibited a marked decrease in S50 in comparison with those mixes kept at lower humidities.

Further observations of adhesion tendency of the mixes over several days are presented in Fig. 4. The mix kept at 6% r.h. was stable, with no change in the S50 up to 9 days. The reduction in the degree of adhesion of sulphamerazine powder was more obvious for the mixes stored at 33, 53, and 75% r.h. and the rate of decrease in adhesion tendency increased with increased humidity.

Succinylsulphathiazole interactive system. The effect of storage time and humidities on the stability of succinyl-sulphathiazole interaction mixes is shown in Fig. 5. Little decrease in S50 values of the mixes kept at 6% r.h. was found in period of 9 h. The sharp decrease in S50 at 53 and 75% r.h. was observed within 1·5 h and no further reduction in adhesion tendency was found up to 9 h. The mix kept at 33% r.h. showed a gradually decreased degree of adhesion with time and reduced to the same degree of the mixes stored at 53 and 75% r.h. at storage time of 5 h.

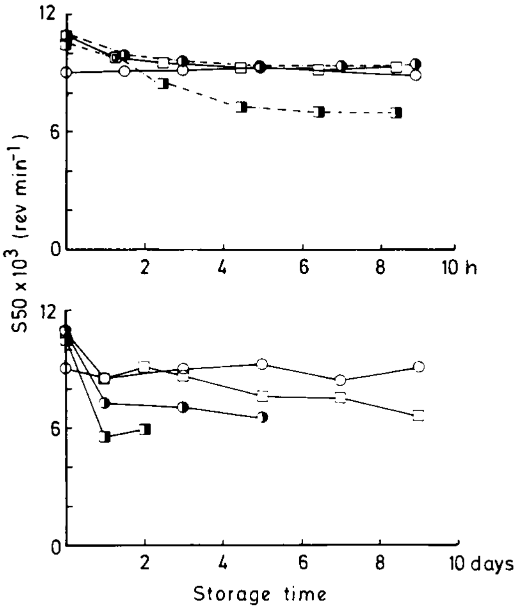

FIG. 4. Effect of relative humidity on the adhesive tendency (S50) of sulphamerazine interactive mixtures during storage. ○ = 6% r.h.; □ = 33%r.h.; ◑ = 53% r.h.and ◧ = 75% r.h.

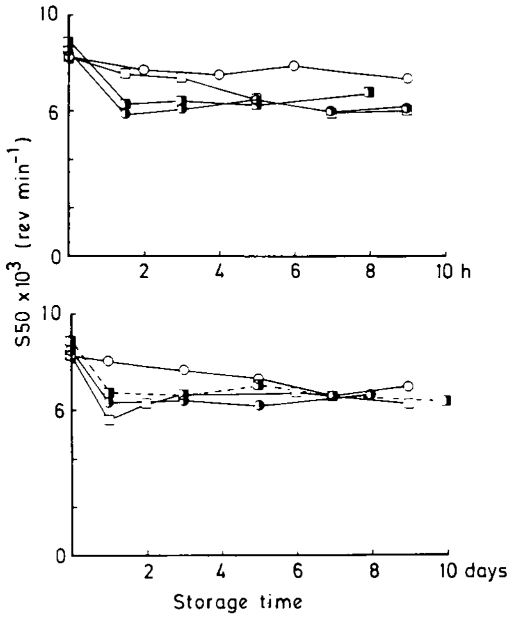

FIG. 5. Effect of relative humidity on the adhesive tendency (S50) of sulphathiazole interactive mixtures during storage. ○ = 6% r.h.; □ = 33%r.h.; ◑ = 53% r.h.and ◧ = 75% r.h.

Further investigations on the stability of those mixtures for several days showed that the mix kept at 6% r.h. exhibited further reduction of S50 with longer time of storage. However, little decrease in stability of the mixes was found for the mixes kept at 33, 53, 75% r.h. after reaching minimum S50 at 9 h of storage.

Generally, all interactive mixes exhibited reduction in adhesion tendency when the storage relative humidity increased. The degree of S50 reduction was dependent on the level of humidity and time of exposure and was probably caused by faster electrical charge dissipation at higher humidities. Charge decay was shown previously to be responsible for the decrease of adhesion tendency of interactive mixes during storage (Kulvanich & Stewart 1987b). The interactive drug systems behaved differently during storage. The S50 of the succinyl sulphathiazole system reached an asymptotic level faster than the other drug systems. Sulphamerazine powder possessed the slowest rate to reach the asymptote in these three drug materials.

The charge decay curves often exhibit a two-step decay, the first step being a fast decay, the second a slow process according to Lee & Weser (1979). But, not all our S50 reduction profiles were observed to be two-step decays. Only sulphamerazine powder showed an initial fast rate followed by a slower process at 33% r.h. and above. The S50 reduction curve always retains a certain asymptotic level as the other force components, i.e. molecular forces, capillary forces will always exist when the electrical interactions are diminished by charge decay.

Effect of moisture content of the powder

The cumulative effects of the previous history of a powder reflect its physical properties as well as its interactive characteristic (Hiestand 1966). The most likely contaminant of powder during storage is the moisture vapour of the ambient atmosphere. Since the contact electrification process is a surface phenomena, the moisture contamination of the surface of contacting bodies will alter its nature of charge generation and the rate at which it is dissipated. The investigation of the effect of relative humidity conditions during material storage on the S50 of the interactive mix was carried out.

The carrier and drug powders were stored for 24 h in a dessicator at controlled temperature (25°C) and relative humidity (26, 43, 53, 75, and 84%) using saturated salt solutions. Four replicate interactive systems were prepared at each relative humidity by blending for 10 min.

Figs 6 and 7 show the relation between the degree of adhesion of succinylsulphathiazole and sulphmerazine powders and the relative humidities at which the powders were stored before blending. The adhesion tendency of succinyl sulphathiazole powder mixes was significantly reduced when the relative humidity conditions were increased (analysis of variance; $P<0{\cdot}001$). This behaviour conformed to the general trend of the effect of humidity on electrostatic interactions. The S50 values of the mixes prepared from the powder stored at 75% r.h. were in the same order as the minimum S50 reduction of the mixes during storage (Fig. 5).

Unexpected results for the adhesion tendency of sulphamerazine interactive mixes were observed (Fig. 7). The adhesion tendency of sulphamerazine powder was significantly increased when powders were stored at 53% r.h. and above (analysis of variance; $P<0{\cdot}001$). The capillary force would be expected to be low in this adhesive system as the hydrophobic surface property of materials, in particular the sulphamerazine powder, would not form adsorbed moisture film layers thick enough to induce such capillary interaction. Adsorbed water vapour could have two effects on the surface charge (Turner & Balasubramanian 1976). Although it is well known that adsorbed water will increase surface conductance and hence cause rapid charge dissipation, the adsorbed moisture vapour may also provide more charging sites on a particle. This effect possibly occurred on sulphamerazine particle with the water vapour associated with the surface localizing at various spots and behaving as additional charging sites resulting in an increase of electrostatic interactions.

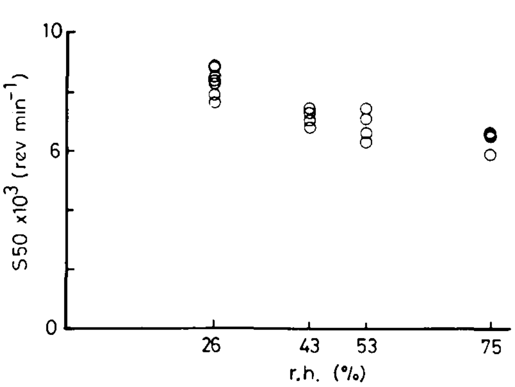

FIG. 6. Effect of relative humidity preconditioning on the adhesive tendency (S50) of succinylsulphathiazole interactive mixtures.

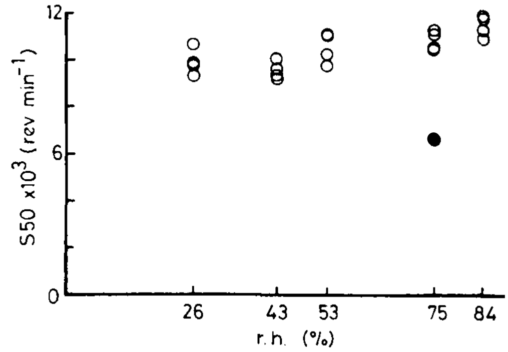

FIG. 7. Effect of relative humidity preconditioning on the adhesive tendency (S50) of sulphamerazine interactive mixtures. ● = S50 of mixture stored at 75% r.h. for 24 h.

This effect was tested by observing the adhesion tendency of a mixture which was prepared from the materials stored at 75% r.h. for 24 h and then stored at the same relative humidity. The result of adhesion measurement showed a decrease in S50 to 6672 rev min^{-1} (initially 11224 rev min^{-1}, shown by closed circle in Fig. 7). Such a decrease was in agreement with the data in Fig. 4 reflecting that charge decay had occurred.

Conclusions

Relative humidity has a marked effect on the degree of adhesion between drug powders and model polymer-coated carriers. Not only will the adhesive properties of powders be dependent on the relative humidity during blending operations and on the moisture of the powders, but also the degree of interaction will change during storage. Awareness of these effects, therefore, is needed in the design of powder systems in order to achieve optimization in processing.

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