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Quantifying electrostatic interactions in pharmaceutical solid systems

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Abstract

Triboelectrification of pharmaceutical powders with stainless steel and polymer contact surfaces was investigated. α-Lactose monohydrate, from 90 to 125 up to 355–500 µm, was used to quantify electrostatic interactions with negligible powder adhesion to the contact surface. Size fractions down to 53–75 µm alone and in binary mixtures with < 10 µm lactose or micronized salbutamol were used to investigate triboelectrification with powder adhered to the contact surface. Triboelectrification was performed in a cyclone charger fitted with interchangeable contact surfaces of steel and polymers, representing the surfaces of pharmaceutical processing and manufacturing equipment, packaging materials and components of dry powder inhaler devices. The results for single component powders showed charge acquisition was inversely related to particle size, where contact surface contamination was negligible. However, with particulate contamination, triboelectrification was more complex due to particle collisions with clean and contaminated contact surfaces. Analysis of adhered and non-adhered powder provided information about changes in composition of two component powders during triboelectrification. Particle size and chemical analyses showed that composition changes of mixtures may be related to powder/contact surface affinity and interparticulate forces for separation of components in a cohesive mix during triboelectrification.

Keywords: Electrostatic charge; Triboelectrification; Contact surface materials; Particle size; Adhesion

1. Introduction

The European Agency for the Evaluation of Medicinal Products has published 'notes for guidance' through the Committee for Veterinary Medicinal Products (CVMP, 1997) and Committee for Proprietary Medicinal Products (CPMP, 1998). Each specifies the need to include consideration of electrostatic properties of powders in relation to: (a) homogeneity in medicated premixes for oral administration in animal feedingstuffs (CVMP); and (b) particle and device surface charge, as well as development of charge during filling and use of dry powder inhalers (CPMP).

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Measurement of charge propensity and decay for pharmaceutical powders after contact with materials, e.g. steel, glass and polymers used in processing, manufacture, packaging and drug delivery, provides essential information relating to the behaviour of the powder and quality of the finished medicine.

Electrostatic charging in solid systems occurs by contact electrification. When two surfaces make contact, transfer of electrons can occur such that on separation, the two surfaces have opposite charges. The charge will distribute over the surface at a rate (relaxation time) which is the product of permittivity and surface resistivity of the materials. Relaxation time for conductors is very short and can be considered to be instantaneous, whereas that for insulators will be much longer, perhaps minutes or hours (Bailey, 1984). In processes where contact involves frictional effects due to sliding, rolling and impact, then the term triboelectrification is used. It is envisaged that triboelectrification events will be influenced by particle properties and processing and they should be considered at all stages, from raw material preparation through to administration of the medicine.

Electrostatic charging in pharmaceutical systems is generally by triboelectrification which can be shown to be a complex process. The complexities of the contact event between two solids arise because the contact pressure, area, time and frequency, are usually unknown and difficult to quantify. In some cases, the event may be further complicated by material transfer between the two surfaces. Despite these difficulties, the principal factors that affect electrostatic charge propensity and charge decay can be identified as contact surface properties, particle properties, the contact event and the atmospheric conditions.

The particle size, shape (Carter et al., 1998), surface nature, purity and roughness (Eilbeck et al., 1999) and the electrical and mechanical properties of the powder and the contact surface (Elsdon and Mitchell, 1976; Bailey and Smedley, 1991), all affect triboelectrification. The atmospheric conditions, e.g. relative humidity (rh) are known to affect charge generation and dissipation (Nguyen and Nieh, 1989; Mackin et al., 1993), however these effects are often unpredictable. Temperature effects are also uncertain, however it had little effect on triboelectrification of selected polymers on impact with metal (Bailey and Smedley, 1991).

Laboratory measurements with pharmaceutical solids can be made using two main approaches, one for the bulk powder and another for individual samples or particles. Measurements on bulk powder samples can be made using Faraday pail techniques (Fasso et al., 1982; Carter et al., 1992a), whereas more complex apparatus based on a capacitive probe technique (Singh and Hearn, 1985; Carter et al., 1992b) and atomic force microscopy (Pollock et al., 1996) are used for individual particles or samples. The net charge acquired by a powder sample through triboelectrification can be measured using a Faraday pail connected to an electrometer. The net charge may be either positive or negative and its magnitude for pharmaceutical powders is usually expressed as nC g^{-1}, where C = coulombs. It is possible that the net charge may result from bi-polar charging within a powder sample, particularly where there is a wide particle size distribution (Bailey, 1984)

The aim of this paper is to investigate a cyclone charge apparatus for quantifying electrostatic interactions between pharmaceutical solids during triboelectrification in cases where powder adhesion to the contact surface is either negligible or appreciable.

2. Materials and methods

2.1. Materials

The powders for triboelectrification experiments were α-lactose monohydrate (Lactochem), used to prepare size fractions and salbutamol sulphate (micronized, Glaxo-Wellcome). The contact surfaces used as inserts in the cyclone charger were: stainless steel grade 316 (Tubesales, Southampton, UK); acetal, polypropylene, polyvinylchloride (pvc) all from Norplast, Newcastle upon Tyne, UK and polyamide (Brisbay Plastics, Newcastle upon Tyne, UK).

2.2. Particle size fractions

The particle size fractions in the range from 53–75 μm to 355–500 μm were prepared by sieving using BS test sieves, followed by air-jet sieving (Alpine) to remove unwanted fine particles from the size fraction. Qualitative microscopic observation confirmed that air-jet sieving was effective for this purpose. The lactose size fraction <10 μm was prepared by air classification using a zig-zag classifier (Multi-Plex 100 MZR).

2.3. Particle size analysis

The size analysis of the binary lactose carriers containing 5% w/w <10 μm with (a) 53–75 μm and (b) 75–90 μm before and after triboelectrification, was performed by laser light scattering (Mastersizer MS1000, Malvern). The lactose samples were dispersed in a solution of 0.05% w/w lecithin in butan-1-ol before analysis.

2.4. Analysis of salbutamol in DPI formulations

The composition of the salbutamol/lactose mix deposited in the Faraday pail after triboelectrification was determined as follows. The powder sample was dissolved in a methanol/water 70:30 solution, diluted and analysed by UV spectrophotometry at 226 nm.

2.5. Triboelectrification with cyclone charger

Controlled triboelectrification of a powder sample is caused by collision of particles with the inner surface of the cyclone charger device (Fig. 1). The real time charge to mass ratio is continuously monitored by a Faraday pail/electrometer and load cell unit with computer interface. The cyclone charger is designed to enable the inner contact surface to be easily changed to undertake triboelectrification measurements against the most appropriate contact surface, e.g. steel or polymer. In addition to the effects of contact surface type, surfaces of different topography may be investigated.

The essential components of the system include an oil free compressor/drier unit to provide carrier gas with rh down to 2% and controlled gas velocities. The powder sample (typically 1 g) is fed into the gas stream from a variable control vibratory feeder via a venturi, housed in a unit purged with carrier gas at the same rh as that in the cyclone charger. A radioactive charge neutralizer, ^{210}Polonium (α source) is built in the feeder system to reduce the charge of the powder sample to a low base line value. The particles are transported pneumatically, via a horizontal pipe leading tangentially into the cyclone charger, where triboelectrification occurs by contact with the inner wall. The powder feeder system and the cyclone are easily dismantled for cleaning. Re-assembly of these components and where necessary, change of contact surface can be done rapidly.

3. Results and discussion

The charge acquisition data is presented as the net charge (Q) to mass (m) ratio at the end of each triboelectrification experiment, however analysis of data from the Q versus time and m versus time data is not presented in this paper. The results presented in the figures and tables are the mean specific charge (nC g^{-1}) for five replicate powder samples from a batch, using specified gas velocity, rh and contact surface. The relative humidity was $<10\%$ during the triboelectrification experiments, unless otherwise stated.

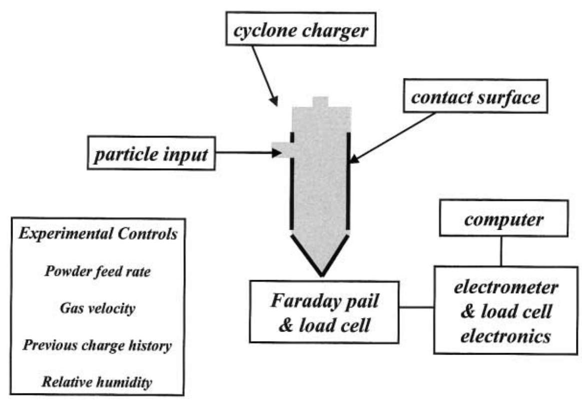

Fig. 1. Cyclone charger for quantifying electrostatic interactions.

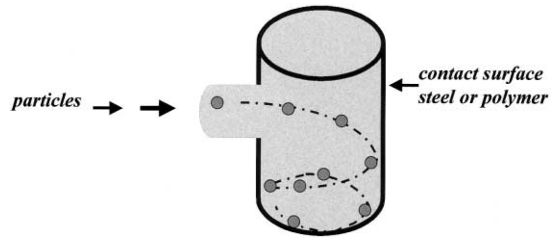

Fig. 2. Particle/contact surface interaction during triboelectrification in a cyclone charger.

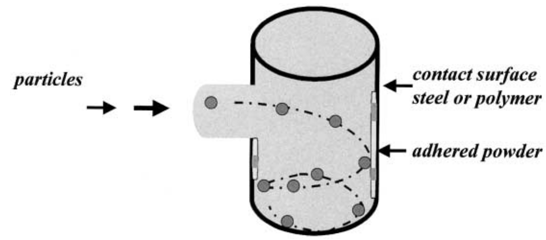

Fig. 4. Particle interaction with contact surface and adhered powder.

3.1. Effect of contact surface

A powder formulation can make contact with a variety of solid surfaces during pharmaceutical processes, product manufacture and during use of the product by the patient. It is therefore important to investigate charge acquisition of a powder during triboelectrification with the most appropriate contact surface, as shown in Fig. 2 where charge acquisition is assumed to be through interaction of the powder with a clean contact surface. Electrostatic charge propensity of active substances, excipients and formulations in contact with a range of pharmaceutical solids, can be investigated by this triboelectrification experiment. For this purpose, it is essential that the contact surface is decontaminated by thorough cleaning before each experimental run.

The results in Fig. 3 show the effect of particulate contamination of a stainless steel contact surface after triboelectrification of α-lactose monohydrate (125–150 μm). The specific charge of this lactose size fraction was -84 nC g^{-1} after triboelectrification against a clean steel surface, when using a carrier gas velocity of 9.8 m s^{-1}. The experiment was repeated without cleaning the contact surface and particulate contamination reduced the charge acquisition to -52 nC g^{-1} under the same triboelectrification conditions. Particulate contamination reduced the contact area available for charge transfer from the surface and the net charge was acquired by particle/metal and particle/particle interactions, as illustrated in Fig. 4.

Cylindrical inserts and the base cone of the cyclone charger were manufactured from steel and a range of polymers used in the fabrication of dry powder inhaler devices. Triboelectrification of α-lactose monohydrate (125–150 μm) with steel, acetal, polypropylene and pvc, showed considerable effects of contact surface type (Fig. 5). The lactose sample acquired a net negative charge with steel and acetal, whereas a net positive charge was acquired with polypropylene and pvc. The magni-

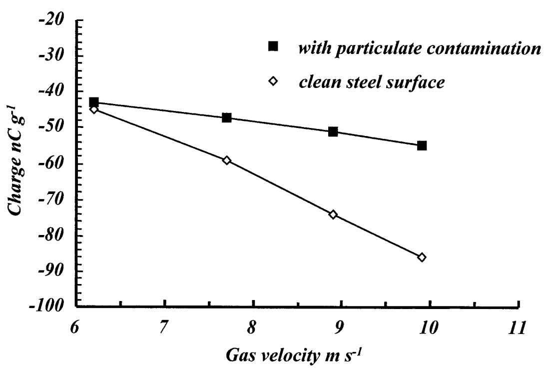

Fig. 3. Effect of steel contact surface contamination on charging of α-lactose monohydrate (125–150 μm) at different gas velocities.

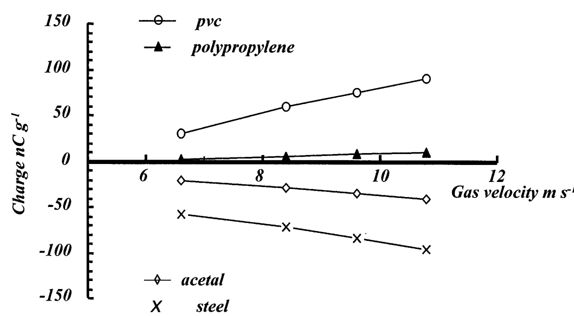

Fig. 5. Effect of contact surface type on charging of α-lactose monohydrate (125–150 μm) at different gas velocities.

tude of charge was also dependent upon the contact surface type, for example at a carrier gas velocity of 10 m s^{-1} the mean specific charge values were -80, -30, $+10$ and $+70$ nC g^{-1} after triboelectrification with steel, acetal, polypropylene and pvc, respectively.

The mechanism of charge acquisition by lactose particles is assumed to be by electron transfer from a contact surface of lower work function. In this case, the negative charge acquired by triboelectrification with steel is assumed to be by donation of electrons from steel, a conductor. The three polymer contact surfaces are insulators and would not normally be involved with electron transfer, however, in practice it is envisaged that the polymer surface will have contaminants and defects which act as electron acceptor and/or donor sites. The results in Fig. 5 provide evidence for electron donor sites on the surface of acetal and electron acceptor sites on the surface of pvc, since the lactose sample charged negatively with acetal and positively with pvc. Triboelectrification of lactose with polypropylene resulted in a very low net positive charge and thus minimal charge transfer occurred between these two solids. The work function of a material is defined as the minimum energy required to move an electron from the solid to infinity. The evidence from these triboelectrification experiments would rank steel with the lowest work function, then increasing order of work function, acetal, lactose, polypropylene and pvc. It would also appear from these results that α-lactose monohydrate and polypropylene have similar charge transfer propensity that resulted in a very low net positive charge acquired by lactose during triboelectrification between the surfaces. The polarity of the polymer can also have a role in the charge transfer process (Elsdon and Mitchell, 1976) and this could contribute in part to the differences shown for the charging of lactose in contact with polymers. The complexity of the triboelectrification event means that the effects of lactose charging with polymers cannot be explained by work function alone. As particles charge at the contact surface, a space-charge field is generated and this will influence charge transfer. This effect was investigated for polymer particles impacting on a metal target in the presence of a variable space-charge field (Bailey and Smedley, 1991) that had a considerable effect on charge transfer. It is therefore possible that a space-charge field will also contribute to triboelectrification in pharmaceutical systems.

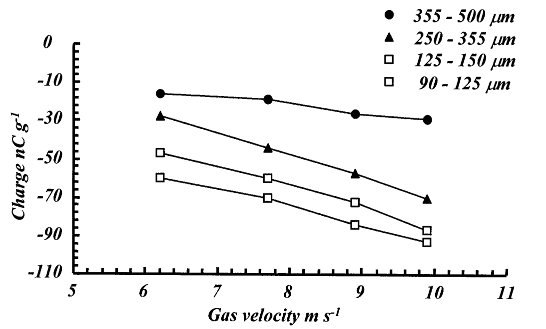

Fig. 6. Effect of particle size on charging of α-lactose monohydrate against stainless steel contact surface at different gas velocities.

Triboelectrification experiments to investigate the interaction between powders and a range of contact surfaces provide information that would facilitate the choice of materials for storage containers, packaging materials and the component parts of dry powder inhaler delivery devices.

3.2. Effects of particle size

The results in Fig. 6 show the effect of particle size on the charge propensity of α-lactose monohydrate during triboelectrification against stainless steel. The specific charge values are inversely related to particle size for sieve size fractions in the range from 90–125 μm to 355–500 μm. Particle adhesion to the contact surface was negligible and therefore triboelectrification was predominantly through particle and metal interaction. The smaller size fractions gain a higher specific charge due to the greater number of possible particle/metal collisions. Thus, where interactions are predominantly between particle and contact surface with minimal particle adhesion to the contact surface, then it can be predicted that specific charge will be inversely related to particle size. This effect is corroborated by the data obtained for triboelectrification of similar size fractions of α-lactose monohydrate with a pvc contact surface, shown in Fig. 7. In this case, the lactose acquires a net positive charge with specific charge values of +25, +55 and +75 nC g^{-1} for size fractions 125–150, 250–355 and 355–500 μm, respectively, using a carrier gas velocity of 9.5 m s^{-1}.

The relationship between particle size and specific charge becomes more complex as the size decreases below 90 μm and the potential for particle adhesion to the contact surface increases, as shown in Fig. 4. The triboelectrification process now comprises particle collisions with the contact surface and particles adhered to the contact surface. Sieve size fractions that are in the size range that may be used for carriers in dry powder inhalers were investigated. Three size fractions, 53–75, 75–90 and 90–125 μm, were investigated by triboelectrification against a steel contact surface. The specific charge results obtained at a gas velocity of 10 m s^{-1} were −20, −37 and −55 nC g^{-1} for 53–75, 75–90 and 90–125 μm, respectively. In this set of results, the smallest size fraction has the lowest specific charge and is comparable to that acquired by the size fraction 250–355 μm (Fig. 8), under the same experimental conditions. The 250–355 μm fraction was charged predominantly through interaction with the steel contact surface, whereas the 53–75 μm fraction undergoes complex triboelectrification due to particle contact with the contact surface and also with powder adhered to the contact surface.

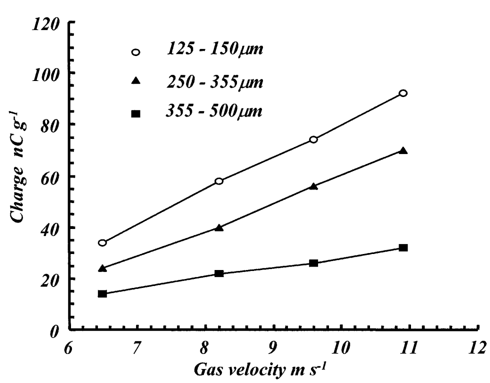

Fig. 7. Effect of particle size on charging of α-lactose monohydrate against a pvc contact surface at different gas velocities.

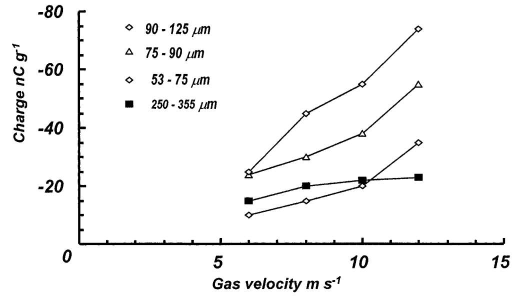

Fig. 8. Charge for carrier lactose size fractions with a steel contact surface.

In cases where adhesion of powder to the contact surface occurs, it is possible to investigate changes in composition of the powder that may occur during triboelectrification. This can be achieved by analysis of the adhered and non-adhered powder after the triboelectrification experiment. Thus, particle size analysis or chemical analysis of the powder adhered to the contact surface and that deposited in the Faraday pail, can provide data to show changes in composition of the powder during triboelectrification. There is evidence (Podczeck, 1999) to indicate that drug delivery from a dry powder inhaler may be improved by the addition of small carrier particles, e.g. <10 μm to larger carrier particles.

The effect of <10 μm α-lactose monohydrate in a binary mix with larger carrier particles on electrostatic charge, was therefore investigated. Binary carriers containing 5% w/w <10 μm lactose with (a) 53–75 μm and (b) 75–90 μm were subjected to triboelectrification against a steel contact surface at 10 m s^{-1}. The results presented in Fig. 9 show a considerable reduction in charge for the binary carrier systems, when compared to carriers without the addition of <10 μm lactose particles.

The charge of the single carriers was −64 and −36 nC g^{-1} for 53–75 and 75–90 μm, respectively. The charge for the binary system was reduced considerably by the presence of <10 μm lactose particles to −18 and −14 nC g^{-1} for the 53–75 and 75–90 μm carriers, respectively. The reduction in charge can be attributed to increased powder adhesion to the steel contact surface and hence, a reduction in the particle–steel interaction

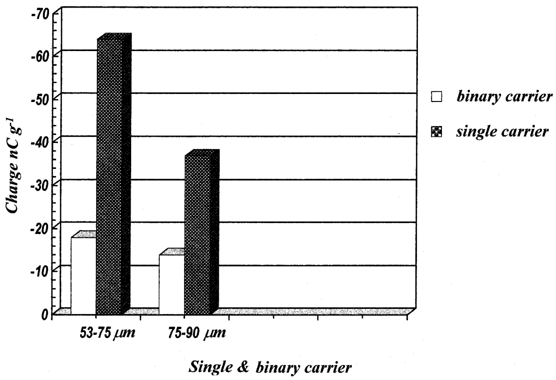

Fig. 9. Charge of single and binary lactose carriers containing 5% w/w < 10 μm lactose with a steel contact surface.

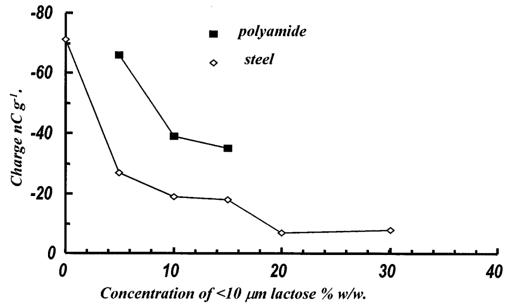

Fig. 10. Effect of < 10 μm lactose concentration on the charge of 63–90 μm lactose against steel and polyamide contact surfaces.

during triboelectrification. The electric charge neutralization of polystyrene spheres by the addition of fine particles has been reported for triboelectrification in a fluidised bed (Wolny and Opalinski, 1983). This effect could account partly for the reduction in charge in a binary lactose system, in addition to the effect of particle/particle interactions at the contact surface. In addition, where a powder comprises course and fine particles, it is possible for bipolar charging to occur with a reduction in net charge of the powder sample.

Particle size analysis of the powder deposited in the Faraday pail was compared to the size distribution of the powder sample before triboelectrification. The results of these analyses are summarized in Table 1 and show a shift in the distribution to larger sizes after triboelectrification. The difference in the 10th percentile sizes are pronounced, with a large increase in the value after charging, e.g. for the 53–75 μm binary carrier the values were 11 and 21 μm before and after triboelectrification. In the case of the 75–90 μm binary system, the 10th percentile value was 15 μm before and 53 μm after triboelectrification. The size distribution data provides evidence for separation of the < 10 μm particles from the binary carrier system by adhesion to the contact surface during triboelectrification. This analysis of a binary mixture after triboelectrification could be used in the investigation of drug/carrier particle attraction and separation during manufacture of and drug delivery from dry powder inhalers.

The effect of concentration of fine particles on the charging of a carrier size fraction was investigated in more detail with a binary system of < 10 μm and 63–90 μm α-lactose monohydrate. A comparison of contact surface effects was made using steel and polyamide inserts in the cyclone charger. Polyamide was selected as a material used in the fabrication of dry powder inhaler devices. The effect of increasing the concentration of < 10 μm lactose particles on specific charge at

Table 1
Particle size results for binary lactose carriers, before and after triboelectrification

Binary lactose carrier	Median (μm)	10th percentile (μm)	90th percentile (μm)
< 10/53–75 μm			
Before	58	11	99
After	63	21	102
< 10/75–90 μm			
Before	85	15	136
After	89	53	140

8 m s^{-1} shown in Fig. 10, is similar for each contact surface, although the decrease of charge values is greater after triboelectrification with steel.

Similar experiments were undertaken to compare the affinities of fine particles in a binary mix, for different contact surfaces, In this case, dry powder inhaler formulations of salbutamol (0.5, 1.0 and 5.0% w/w) in α-lactose monohydrate 63–90 μm, were investigated by triboelectrification experiments with steel and polyamide contact surfaces. The salbutamol (micronized) had a similar effect on the specific charge of the binary mix as that obtained by increasing the concentration of <10 μm lactose in the binary mix, Fig. 10. The specific charge results in Table 2 show that as the % w/w salbutamol increased from 0.5 to 5.0% w/w, there were decreases in charge from −91 to −38 nC g^{-1} with steel and from −42 to −10 nC g^{-1} with polyamide as the contact surface. However, in this case the polyamide produced a lower charge than the steel for the salbutamol binary mix when compared with the lactose/lactose binary mix.

The salbutamol mixtures deposited in the Faraday pail after triboelectrification were analysed for salbutamol and the results are presented in Table 3. These results demonstrate the difference in affinity of salbutamol for steel and polyamide during triboelectrification. The original blends of salbutamol and lactose contained 0.5, 1.0 and 5.0% w/w of active, whereas after triboelectrification with steel, the salbutamol concentration was 0.84, 1.05 and 7.30% w/w. In contrast, the salbutamol concentration decreased to 0.40, 0.90 and 3.90% w/w during triboelectrification with the polyamide surface. These results show that the composition of the DPI formulation deposited in the Faraday pail was dependent upon the contact surface and that contact with steel produced an increase, whereas contact with polyamide caused a decrease in salbutamol concentration. It can be seen from this experiment that salbutamol has a greater affinity for the polyamide than for the steel contact surface. In addition, the results provide evidence for separation of drug and carrier during triboelectrification and this is dependent on the relative affinities for the active and the carrier for the contact surface.

Table 2
Triboelectric charge (nC g^{-1}) of salbutamol/lactose DPI formulations

Salbutamol in DPI (% w/w)	Contact surface	
	Steel	Polyamide
0.5	−91	−42
1.0	−87	−35
5.0	−38	−10

Table 3
Change in salbutamol concentration in DPI formulations after triboelectrification with steel and polyamide contact surfaces

Salbutamol in DPI (% w/w)	Salbutamol in Faraday pail (% w/w)	
	Steel	Polyamide
0.5	0.84	0.40
1.0	1.05	0.90
5.0	7.30	3.90

4. Conclusions

The cyclone charger can be used to quantify electrostatic charge interactions during triboelectrification experiments with pharmaceutical powders and appropriate pharmaceutical solid contact surfaces.

The cyclone charger is useful for investigation of the behaviour of bulk powders during triboelectrification and this is relevant to many pharmaceutical operations.

The interactions between powder and contact surface have been discussed in two principal cases, where there is no powder adhesion to the contact surface and where adhesion is appreciable. Each of these experimental circumstances can provide information about the behaviour of pharmaceutical powders.

Where powder adhesion is negligible, then the charge propensity of active and excipients with appropriate contact surfaces will provide preformulation data which is essential to formulation and process engineering in the design and manufacture of pharmaceutical delivery systems.

When the triboelectrification event is through a combination of powder interactions with the contact surface and also powder adhered to the contact surface, then behaviour of trial formulations during mixing, filling and drug delivery can be investigated.

Acknowledgements

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