Tribo-Electric Charging of Powders

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Background

- Powder handling causes electric power generation.

- Electric discharge can give rise to explosion and fire hazards.

- In pharmaceutical powder processing context tribo-electrification can cause segregation, adhesion, hence affecting powder formulation.

- Chemical, physical and electrical characteristics, and environmental conditions, i.e. temperature and humidity all affect the charging process.
## Typical Charge in Powder Processes

<table>
<thead>
<tr>
<th>process</th>
<th>specific charge (µC/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>sieving</td>
<td>$10^{-8} \sim 10^{-6}$</td>
</tr>
<tr>
<td>loading</td>
<td>$10^{-6} \sim 10^{-4}$</td>
</tr>
<tr>
<td>transportation</td>
<td>$10^{-5} \sim 10^{-3}$</td>
</tr>
<tr>
<td>polishing</td>
<td>$10^{-4} \sim 10^{-3}$</td>
</tr>
<tr>
<td>comminution</td>
<td>$10^{-4} \sim 10^{-1}$</td>
</tr>
<tr>
<td>pneumatic convey</td>
<td>$10^{-3} \sim 10^{-1}$</td>
</tr>
</tbody>
</table>

Threshold for ES hazard: $10^{-4}µC/g$
Relevance to Organic Powders

Organic powders are very prone to electrostatic interactions:

• Small particle size
   Less than 50 microns

• Usually irregularly shaped
   Rarely round

• Usually poor charge conductors
   Semiconductors or insulators

• We need to have knowledge of single particle charging in order to understand BULK behaviour.
• The polarity and level of charge transferred to the particles by tribo-electric charging need be quantified.
<table>
<thead>
<tr>
<th>MATERIALS</th>
<th>POLARITY</th>
</tr>
</thead>
<tbody>
<tr>
<td>Acetate</td>
<td>+</td>
</tr>
<tr>
<td>Glass</td>
<td>+</td>
</tr>
<tr>
<td>Human Hair</td>
<td>+</td>
</tr>
<tr>
<td>Nylon</td>
<td>+</td>
</tr>
<tr>
<td>Lead</td>
<td>+</td>
</tr>
<tr>
<td>Aluminum</td>
<td>+</td>
</tr>
<tr>
<td>Paper</td>
<td>+</td>
</tr>
<tr>
<td>Polyurethane</td>
<td>+</td>
</tr>
<tr>
<td>Cotton</td>
<td>+</td>
</tr>
<tr>
<td>Steel</td>
<td>+</td>
</tr>
<tr>
<td>Hard Rubber</td>
<td>+</td>
</tr>
<tr>
<td>Acetate Fiber</td>
<td>+</td>
</tr>
<tr>
<td>MYLAR*</td>
<td>+</td>
</tr>
<tr>
<td>Epoxy Glass</td>
<td>+</td>
</tr>
<tr>
<td>Nickel, Copper, Silver</td>
<td>+</td>
</tr>
<tr>
<td>UV Resist</td>
<td>+</td>
</tr>
<tr>
<td>Stainless Steel</td>
<td>+</td>
</tr>
<tr>
<td>Synthetic Rubber</td>
<td>+</td>
</tr>
<tr>
<td>Acrylic</td>
<td>+</td>
</tr>
<tr>
<td>Polystyrene Foam</td>
<td>+</td>
</tr>
<tr>
<td>Polyurethane Foam</td>
<td>+</td>
</tr>
<tr>
<td>Polyester</td>
<td>+</td>
</tr>
<tr>
<td>Polyethylene</td>
<td>+</td>
</tr>
<tr>
<td>Polypropylene</td>
<td>+</td>
</tr>
<tr>
<td>PVC (Vinyl)</td>
<td>+</td>
</tr>
<tr>
<td>TEFILON*</td>
<td>+</td>
</tr>
<tr>
<td>Silicone Rubber</td>
<td>+</td>
</tr>
</tbody>
</table>

* Trademark of E.I. Du Pont
Tribo-Electric Charging Techniques Developed at Leeds

- Single particle impact
- Particle shaking in a container
- Aerodynamic dispersion by bursting foils
Impact of a rubber sphere on a plate and contact area

Impact of a rubber sphere on an impact plate.

maximum contact area,

Transferred charge, $dq/dn$ (nC)

$\frac{dq}{dn} / S = 1.8 \times 10^{-5}$ C m$^{-2}$

Initial charge: $q = 0 \pm 3$ nC

$v_i = 4.8$ m s$^{-1}$

$v_i = 2.8$ m s$^{-1}$

$v_i = 2.0$ m s$^{-1}$

$v_i = 1.4$ m s$^{-1}$

$v_i = 2.8$ m s$^{-1}$

$v_i = 4.8$ m s$^{-1}$

Relationship between transferred charge and maximum contact area.

Matsusaka et al. (2000)
Impact charge with particles down to 100 µm in size can be measured even by one-by-one feeding test.

Impact charge (FC2-FC1) = -0.2pC

Impact charging test rig

With target

95-106mm a-lactose
Test Conditions

- **Target:**
  - stainless steel,
- **Impact angle:**
  - 30° and 60° (with respect to target surface)
- **Environmental conditions:**
  - ambient (RT: 20~25 °C, RH: 20 ~ 40 %)
- **Velocity:**
  - 5 to 30 m/s Particle size: 500 - 600 mm (sieve size)
Typical Results: Impact Charging Test

Sample: Sugar granules. Impacted at 9.0 m/s with impact angle 30°.
Data Scatter due to Particle Shape

Lactose (9.8 m/s): **TOMAHAWK**

Sugar granules (9.0 m/s): nearly **SPHERICAL**

Aspirin (11 m/s): **OBLONG**

Glass beads (9.7 m/s): **SPHERICAL**

[angle 30° - 4 mbar]
Impact charge vs Initial charge

- Impact charge, for example, $D_{qo}$, at zero initial charge, increases with increasing impact velocity.
- On the other hand, equilibrium charge, $Q_e$, does not depend on impact velocity.

$D_{qo}$ and $Q_e$ are essential parameters for characterising the charging tendency of the sample particles, and will be described as a function of impact velocity and impact angle.

Illustration of charging lines based on data of sugar granules (500-600mm) at impact angle 30°
Impact Charge $Dq_0$ vs. Contact Area $S$

Estimated contact area, $S [10^{-9} \text{ m}^2]$

Impact angles

$\sin q \cdot V_i$

$\cos q \cdot V_i$

# Data of SG, aLM and ASP seem to lie on one line except EC.
Equilibrium Charge: $Q_e$

$Q_e$ is independent of impact velocity and angle. Therefore, $Q_e$ is an important material characteristic.
**Triboelectric Series vs Qe**

<table>
<thead>
<tr>
<th>Positive end</th>
<th>Qe vs SS</th>
</tr>
</thead>
<tbody>
<tr>
<td>EC</td>
<td>+ 10 pC</td>
</tr>
<tr>
<td>GS, Perspex, SS, Al</td>
<td></td>
</tr>
<tr>
<td>aLM, SG</td>
<td>- 18, -16 pC</td>
</tr>
<tr>
<td>PP</td>
<td></td>
</tr>
<tr>
<td>ASP</td>
<td>- 40 pC</td>
</tr>
<tr>
<td>PTFE</td>
<td></td>
</tr>
</tbody>
</table>

**Negative end**

**Targets:** GS: glass, SS: stainless steel, PP: polypropylene, PTFE: polytetrafluoroethylene

The triboelectric series was estimated from charging tests of bulk powders (1g) presented later on in this presentation.

→ *A qualitative agreement between tribo-electric series and Qe can be seen.*
Contact Potential Difference (CPD) of the sample powders was measured by a system that has been developed at Kyoto University.

- Equilibrium charge $Q_e$ can be qualitatively correlated with the CPD.
- It should be noted that $Q_e$ is a function of particle size. Therefore, the difference in particle shape among the samples would give different $Q_e$.

$$\text{CPD of sample vs SS} = \text{CPD of sample vs Au} - \text{CPD of SS vs Au}$$

$$\text{CPD of target (SS)} = +0.12 \text{ V vs Au}$$
From Single Particle to Bulk
Charging by Aerodynamic dispersion by bursting foils

Effect of Foil Material on Tribo-electric Charging of Glass Beads

<table>
<thead>
<tr>
<th>Material</th>
<th>Thickness (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stainless Steel</td>
<td>5</td>
</tr>
<tr>
<td>Copper</td>
<td>12.7</td>
</tr>
<tr>
<td>Aluminium</td>
<td>6</td>
</tr>
</tbody>
</table>
Methodology
Q/M vs. Corresponding Work (Glass Beads)

\[ \bar{W} \propto \left( \frac{p_1}{p_2} \right)^{\frac{\gamma-1}{\gamma}} - 1 \]
Charging by Shaking

Tribo-electrification Approach:

• Development of simple test method for characterising the tribo-electrification properties of powder materials, consisting of Retsch® shaking machine, set of interchangeable shaking containers made out of common industrial materials, electrometer, Faraday cup, and isolator booth.

• The container is subjected to motions in a horizontal direction:

  (1) Multiple particle-wall interactions
  (2) Multiple particle-particle interactions
  (3) Space charge effects
Tribo-Electric Charging Device

- 1 g of sample is weighed and placed inside a container (~10 cm³).

- The sample is then vibrated at selected frequency (min. 3 Hz - Max. 30 Hz) to simulate tribo-charging effect within the container.

![Schematic diagram of a shaking container](image)

**Schematic diagram of a shaking container, (all measurements in mm).**

**1:** Shaking container attachment. **2:** Shaking container attachment with a cooling jacket (Not used in this work). **3:** Control Panel.

Stainless steel shaking container.
Charge Measurement

- The sample is poured into the inner cup, tapping the container to remove particles on the walls. The initial charge on the sample is measured.
- The charge on the sample following tribo-charging process is measured.
- The weight of sample at end is measured - adhesion.

Materials

- Two widely used materials as excipients were selected for tribo-electrification:
  - α-lactose monohydrate (α-LM)
  - Hydroxypropyl cellulose (HPC)
  - 50:50 by weight mixture of the two materials

- Container material:
  - Polytetrafluoroethylene (PTFE)

- Frequencies:
  - 10 Hz, 20 Hz, and 30 Hz
Saturated Charge Level

The amount of charge generated from particle impacts after which no further increase in charge occurs.

<table>
<thead>
<tr>
<th>Humidity (%)</th>
<th>48.2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Container</td>
<td>PTFE</td>
</tr>
<tr>
<td>Frequency</td>
<td>20 Hz</td>
</tr>
</tbody>
</table>

Charge to Mass Ratio (nC/g) vs. Time (min)

- o α-LM
- ■ HPC
- △ Binary
Frequency

<table>
<thead>
<tr>
<th>Frequency [Hz]</th>
<th>α-LM</th>
<th>HPC</th>
<th>Binary</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>24.6</td>
<td>3.3</td>
<td>11.4</td>
</tr>
<tr>
<td>20</td>
<td>24.7</td>
<td>3.2</td>
<td>11.9</td>
</tr>
<tr>
<td>30</td>
<td>22.7</td>
<td>3.8</td>
<td>12.2</td>
</tr>
</tbody>
</table>

- Electrostatic charge increases with the shaking time and reaches a maximum charge (saturated charge)
- The saturated charge is independent of the shaking frequency

Šupuk et al. (2009)
Particle Adhesion to Walls (Mass Loss)

• Particles/debris adhered to the inner wall of the shaking vessel carry the highest magnitude of charge and can have a significant effect on the generated charge.

• Important that these particles are accounted for by tapping on the outer wall of the vessel.

• Not always possible to remove all adhered particles by tapping, nor can they be scraped into the Faraday cup as this would additionally charge the powder, hence some powder is ‘lost’ in this way.

• The amount of ‘lost’ powder is a good indicator of the extent of tribo-charging that is taking place in the vessel.
Mass Loss

% Mass Lost

<table>
<thead>
<tr>
<th>Frequency (Hz)</th>
<th>% Mass Lost</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>6.0</td>
</tr>
<tr>
<td>20</td>
<td>8.5</td>
</tr>
<tr>
<td>30</td>
<td>7.5</td>
</tr>
</tbody>
</table>

- a-lactose @ 45min
- HPC @ 1min
- Binary @ 20min
Wall Adhesion

- Linear relationship:
  \[ y = 0.0022x + 0.0445 \]
  \[ R^2 = 0.9759 \]

- Graph showing:
  - Amount of Mass Adhered to Wall, \( M_a (g) \) vs. Charge to Mass Ratio (nC/g)
  - Scatter plot with fitted line
  - Data points labeled \( \alpha-LM \)
Chargeability: Excipients vs API

## Chargeability: Excipients Vs API

<table>
<thead>
<tr>
<th>Material</th>
<th>API (-) Q&lt;sub&gt;Max&lt;/sub&gt;</th>
<th>API (+) Q&lt;sub&gt;Max&lt;/sub&gt;</th>
<th>EXC (-) Q&lt;sub&gt;Max&lt;/sub&gt;</th>
<th>EXC (+) Q&lt;sub&gt;Max&lt;/sub&gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td>CB</td>
<td>-323</td>
<td>170</td>
<td>-11.3</td>
<td>3.87</td>
</tr>
<tr>
<td>KTS</td>
<td>-182</td>
<td>156</td>
<td>-8.78</td>
<td></td>
</tr>
<tr>
<td>ASP</td>
<td>-117</td>
<td>130</td>
<td>-4.37</td>
<td></td>
</tr>
<tr>
<td>SCA</td>
<td>-79.2</td>
<td>102</td>
<td>-3.92</td>
<td></td>
</tr>
<tr>
<td>AAP</td>
<td>-74.2</td>
<td>49.8</td>
<td>-3.67</td>
<td></td>
</tr>
<tr>
<td>CC</td>
<td>-48.4</td>
<td>10.7</td>
<td>-3.53</td>
<td></td>
</tr>
<tr>
<td>THP</td>
<td>-36.5</td>
<td>6.22</td>
<td>-1.92</td>
<td></td>
</tr>
<tr>
<td>CA1</td>
<td>-15.1</td>
<td></td>
<td>-1.53</td>
<td></td>
</tr>
<tr>
<td>PRS</td>
<td>-1.24</td>
<td></td>
<td>-1.16</td>
<td></td>
</tr>
</tbody>
</table>

* *Q<sub>Max</sub> - maximum charge to mass (nC/g)*

Maximum charge to mass ratio values inside a stainless steel container operated at 20 Hz, at a temperature range of 19-24 °C and a relative humidity range of 20-55 % for selection of excipients and active materials.
Four different compositions of HPC:α-LM tested:
- 20:80, 40:60, 60:40, and 80:20. Pure samples of HPC and α-LM were added for comparison.

- Frequency:
  - 20 Hz

- Container:
  - PTFE

- Humidity:
  - 38 % RH

- Shaking time:
  - correspond to saturated charge level
Charge in Binary Mixtures

Charge to Mass Ratio (nC/g) vs. HPC (% wt)

- PTFE

0 20 40 60 80 100

Charge to Mass Ratio (nC/g)
Tribo-Electrification in Binary Mixtures

• Charge-to-mass (Q/m) ratio generally follows a trend reflecting the composition of the individual components, i.e. a decrease in Q/m with an increase in the amount of HPC in a binary mixture.

• The binary mixture charged at different compositions follows roughly the formula:

\[ q_{\text{mixture}} = x_1 q_{\alpha-LM} + x_2 q_{\text{HPC}} \]

\( x_1 \) and \( x_2 \) are mass fractions of \( \alpha\text{-LM} \) and HPC respectively.
Segregation Studies

- **Background:** Charge transfer between excipient particles and the PTFE wall of the shaking container occurs readily and reaches a sufficiently high level to cause particle adhesion.

- **Aim:** investigate whether such adhesion could give rise to the segregation of components within a binary mixture.

- **Previous Work on Segregation and Electrostatics:** formation of ordered mixtures by tribo-electrification shown to minimise segregation (Staniforth and Rees, 1981)

- Ordered mixtures are formed following tribo-electrification, in which particle surface electrical properties are altered in a way that components are charged opposite to each other to encourage inter-particle adhesion.

- However, in some formulations, model drug and the associated excipients will both charge with the same polarity against a particular surface. Particle will not be inclined to attract to each other to form stable mixtures.
Segregation

- Such mixtures are prone to adhering to the oppositely charged walls, and may promote segregation.

- This issue has not yet been widely addressed and little work reported.

- Link tribo-electric charging tendencies with the segregation tendencies by comparing the composition of particles adhered to the walls with that of the original formulations.

- Ratios of binary mixture (α-LM:HPC) compositions tested:
  - 80:20
  - 60:40
  - 50:50
  - 40:60
  - 20:80

- Pure samples of α-LM and HPC of known weight used for calibration.
Segregation Wall-Adhered Mixture

- The extent of segregation of $\alpha$-LM on the wall (SW) is determined using the following equation:

$$S_W = 1 - \frac{\text{Amount of } \alpha - \text{LM on wall}}{\text{Ideal amount of } \alpha - \text{LM on wall}}$$

- As SW increases from zero the more segregated the mixture on the wall is.
Segregation Wall-Adhered Mixture

$y = -0.1876x + 0.3412$

$R^2 = 0.4623$

Extent of Segregation of $\alpha$-LM in the Wall-Adhered Mixture ($S_W$)
Summary

• A notable amount of the powder is adhered to the container wall, the amount being highest for α-LM, intermediate for the 50:50 mixture and lowest for the HPC. This trend follows the charge level on the adhered powder.

• The saturated charge is independent of the shaking frequency.

• As pure components, α-LM charges significantly higher (approx. 25 nC/g) than HPC (approx. 5 nC/g)

• Binary mixtures showed a decrease in the net charge as the HPC mass fraction increased.
Summary

• **Main mixture:**
The extent of segregation does not exceed 0.1 when the main mixture is considered.

• **Wall-adhered mixture:**
The extent of segregation is considerably higher than the main mixture with the highest value being 0.31 for the 20:80 ratio mixtures and the lowest of 0.19 for the 80:20 ratio mixtures.
Analysis of Charging by Shaking

Simulation of Charging due to Bulk Motion at 10 Hz

Strategy

1. Single Particle Experiment (2 mm particle)
2. Calibration of Model Parameters
3. Bulk Test by Varying Particle Number (1 to 230 particles)
4. Evaluation of DEM Modelling
Materials

PTFE capsule

Alumina beads
# Experimental conditions

<table>
<thead>
<tr>
<th>Particle</th>
<th>Alumina (2.01 mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Capsule</td>
<td>PTFE</td>
</tr>
<tr>
<td>Amplitude</td>
<td>10 mm</td>
</tr>
<tr>
<td>Frequency</td>
<td>30 Hz</td>
</tr>
<tr>
<td>Number of particles</td>
<td>1-230</td>
</tr>
<tr>
<td>Shaking time</td>
<td>10-60 s</td>
</tr>
</tbody>
</table>
Results

Charge (nC/particle) vs. Time (s) for 30 Hz:

- 1 particle: 0.40 nC/particle
- 10 particles: 0.30 nC/particle
- 20 particles: 0.30 nC/particle
- 50 particles: 0.30 nC/particle
- 100 particles: 0.20 nC/particle
- 150 particles: 0.20 nC/particle
- 230 particles: 0.10 nC/particle

Saturation charge (nC/particle) vs. Number of particles:

- 50 particles: 0.40 nC/particle
- 100 particles: 0.30 nC/particle
- 150 particles: 0.30 nC/particle
- 200 particles: 0.20 nC/particle
- 250 particles: 0.10 nC/particle
Distinct Element Method

LIGGGHTS (LAMMPS Improved for General Granular and Granular Heat Transfer Simulations)

LAMMPS (Large-scale Atomic/Molecular Massively Parallel Simulator)

Features
1. Open Source
2. Readability of mesh data made by CAD

Christoph Kloss, Christoph Goniva, Alice Hager, Stefan Amberger, Stefan Pirker
Models, algorithms and validation for opensource DEM and CFD-DEM
Progress in Computational Fluid Dynamics, 2012
1. Contact charging model
2. Model of space charge effect
3. Boundary conditions
4. Electrostatic force
1. Contact charging model

\[
\frac{\Delta q}{\alpha \Delta S} + \frac{q_i}{q_{max}} = 1
\]

\( \Delta q \) (C): Transferring charge
\( q_i \) (C): Charge on particle
\( \Delta S \) (m²): Maximum contact area
\( q_{max} \) (C): Maximum charge
\( \alpha \) (C/m²): Coefficient \( \left( \frac{q_{max}}{\pi d^2} \right) \)

1. Determination of Maximum charge $q_{max}$

$q_{max} = 0.6 \text{ nC}$
2. Model of space charge effect

\[ \Delta q = \alpha \Delta S \left( 1 - \frac{q_i}{q_{\text{max}}} \right) \]

\[ q_e = \pi d^2 \varepsilon_0 E \]

\[ \Delta q = \alpha \Delta S \left( 1 - \frac{q_i + q_e}{q_{\text{max}}} \right) \]

- \( \Delta q \) (C): Transferring charge
- \( q_i \) (C): Charge on particle
- \( \Delta S \) (m\(^2\)): Maximum contact area
- \( q_{\text{max}} \) (C): Maximum charge
- \( \alpha \) (C/m\(^2\)): Coefficient \( (q_{\text{max}}/\pi d^2) \)
- \( q_e \) (C): Equivalent charge
- \( E \) (V/m): Electric field at contact point
2. Model of space charge effect

\[ \Delta q = \alpha \Delta S \left(1 - \frac{q_i + q_e}{q_{\text{max}}} \right) \]

When \( q_i + q_e = q_{\text{max}} \) transferring charge, \( \Delta q \) goes to equilibrium state.
3. Boundary conditions (Simulation Charge Method)

Boundary conditions

\[ V_{\text{Boundary}} = 0 \]

\[ V_{TP_k} = \sum \frac{SC_j}{4\pi \varepsilon_0 r_{SC_jTP_k}} + \sum \frac{q_i}{4\pi \varepsilon_0 r_{q_iTP_k}} \]
3. Simulation charge method
3. Simulation Charge Method

![Charge (pC) diagram]

Charge (pC)

-1

0

1
3. Simulation Charge Method

Potential (V)

| 10 | 0 | -10 |

-10 0 10
3. Simulation charge method
4. Electrostatic force

Coulomb force \( F = \frac{q_i q_j}{4\pi r^2 \varepsilon_0} \)
Simulation

- Number of particle: 100
- Particle diameter: 2mm
- Coefficient of restitution: 0.6
- Coefficient of friction: 0.5
- qmax: 0.7nC
- Frequency: 30Hz

Time: 19.90 s
Simulations show the relationship between charge (nC/particle) and time (s) for different numbers of particles. The saturation charge (nC/particle) is also plotted against the number of particles. The data is compared to experiments by Imba et al. (2013), presented at the Powders and Grains Conference in Sydney, Australia.
Conclusions

The saturation charge level decreases with the number of particles in the experimental work.

Models of charge transfer and space charge effect have been incorporated into the DEM code LIGGGHTS.

The simulated saturation charge levels are overestimated.
Summary of DEM

- Tribo-electrification of spherical beads inside a horizontally shaken sealed capsule has been analysed experimentally and simulated using a DEM model.

- An empirical first order rate equation, based on experimental data, has been incorporated in the DEM simulations.

- It is found that the model significantly overestimates the total charge build-up for bulk shaking, as compared with the experimental results.

- However, the inclusion of space charge effects and boundary condition into the DEM model significantly improves the prediction of total charge build up.
Relevant Publications:


Acknowledgements

• **Special thanks to:**
  - Masayuki Imba
  - Tatsushi Matsuyama
  - Kendal Pitt, MSD (now GSK)
  - Hideo Watanabe
  - Enes Supuk