INSTITUTE OF PARTICLE SCIENCE & ENGINEERING

UNIVERSITY OF LEEDS

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

process	specific charge (μC/g)
sieving	10^{-8} ~ 10^{-6}
loading	10^{-6} ~ 10^{-4}
transportation	10^{-5} ~ 10^{-3}
polishing	10^{-4} ~ 10^{-3}
comminution	10^{-4} ~ 10^{-1}
pneumatic convey	10^{-3} ~ 10^{-1}

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.*

TRIBOELECTRIC SERIES

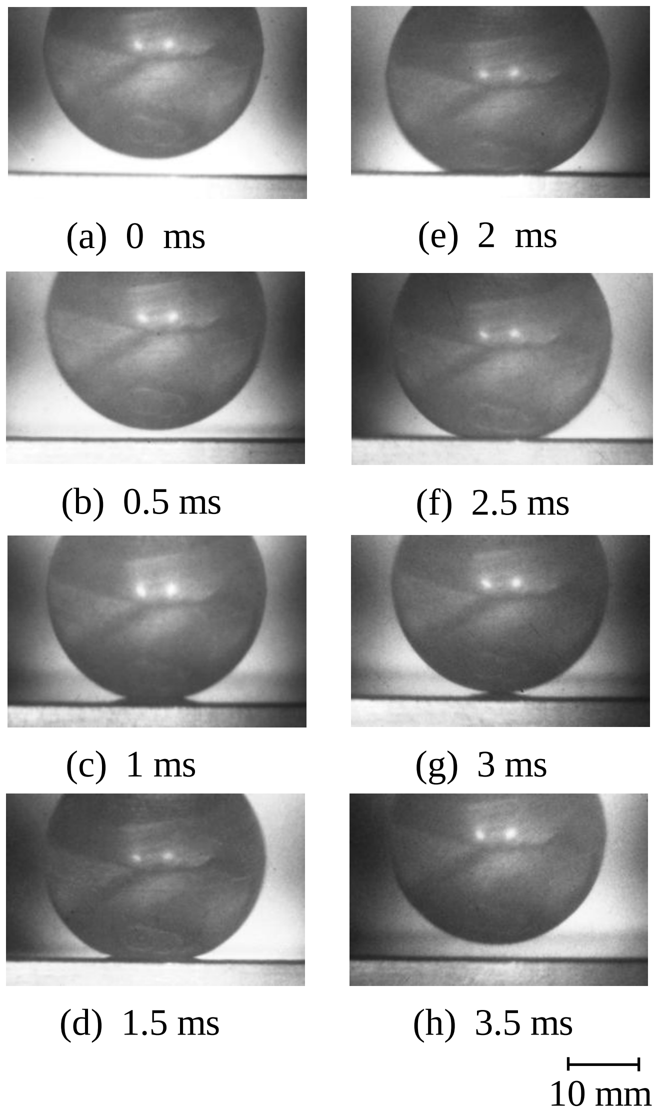
MATERIALS	POLARITY	
Acetate	+	—
Glass	+	—
Human Hair	+	—
Nylon	+	—
Lead	+	—
Aluminum	+	—
Paper	+	—
Polyurethane	+	—
Cotton	+	—
Steel	+	—
Hard Rubber	+	—
Acetate Fiber	+	—
MYLAR*	+	—
Epoxy Glass	+	—
Nickel, Copper, Silver	+	—
UV Resist	+	—
Stainless Steel	+	—
Synthetic Rubber	+	—
Acrylic	+	—
Polystyrene Foam	+	—
Polyurethane Foam	+	—
Polyester	+	—
Polyethylene	+	—
Polypropylene	+	—
PVC (Vinyl)	+	—
TEFLON*	+	—
Silicone Rubber	+	—

* Trademark of E.I. Du Pont

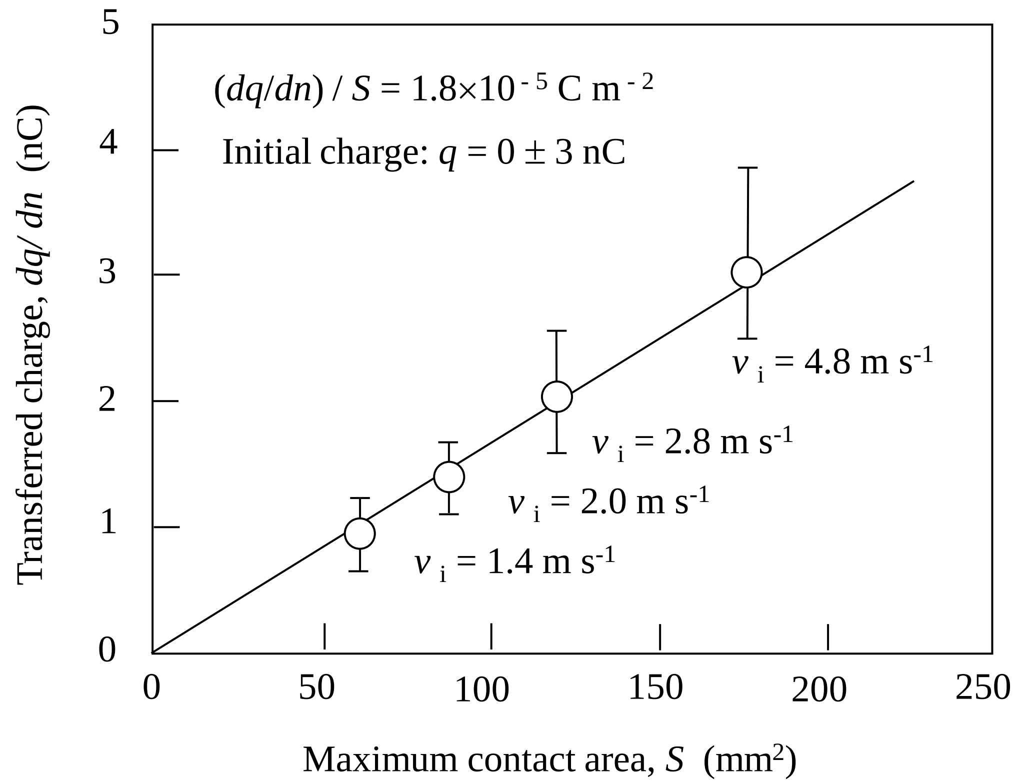
Tribo-Electric Charging Techniques Developed at Leeds

- Single particle impact
- Particle shaking in a container
- Aerodynamic dispersion by bursting foils

Impact of a rubber sphere on a plate and contact area



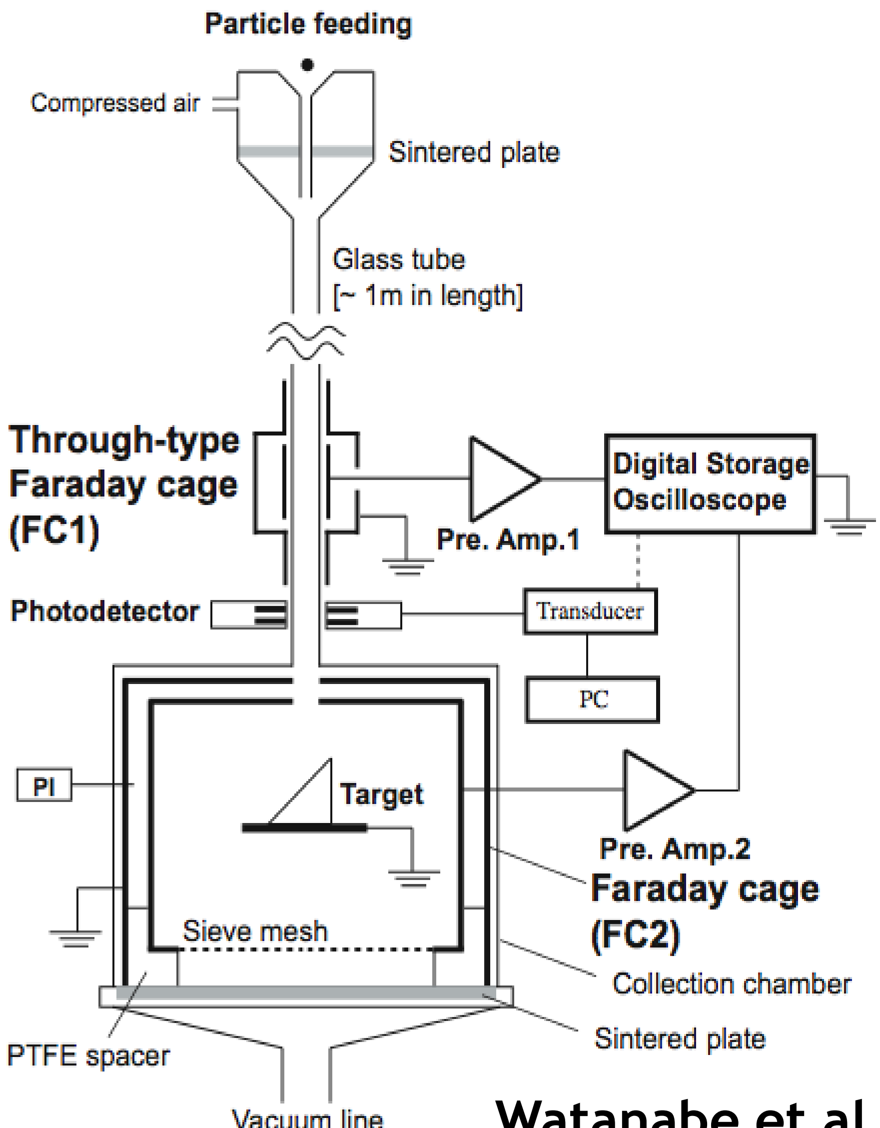
Impact of a rubber sphere on an impact plate.



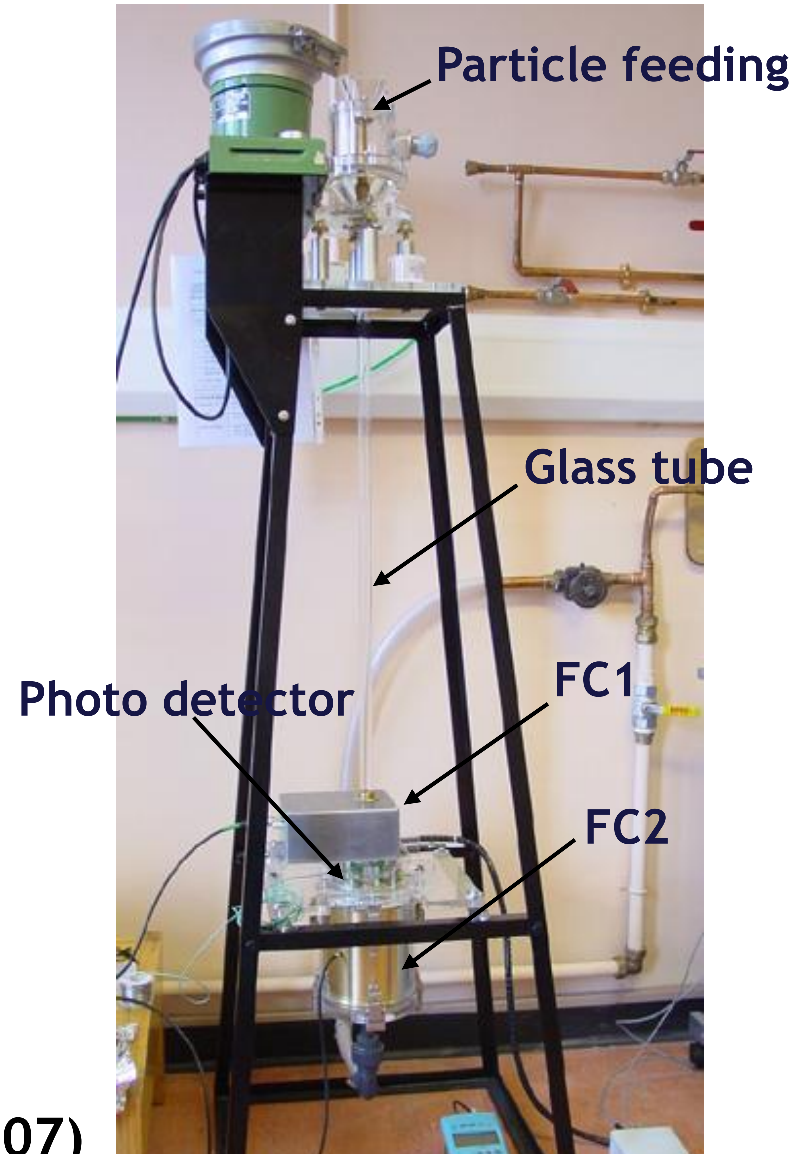
Relationship between transferred charge and maximum contact area.

Matsusaka et al.(2000)

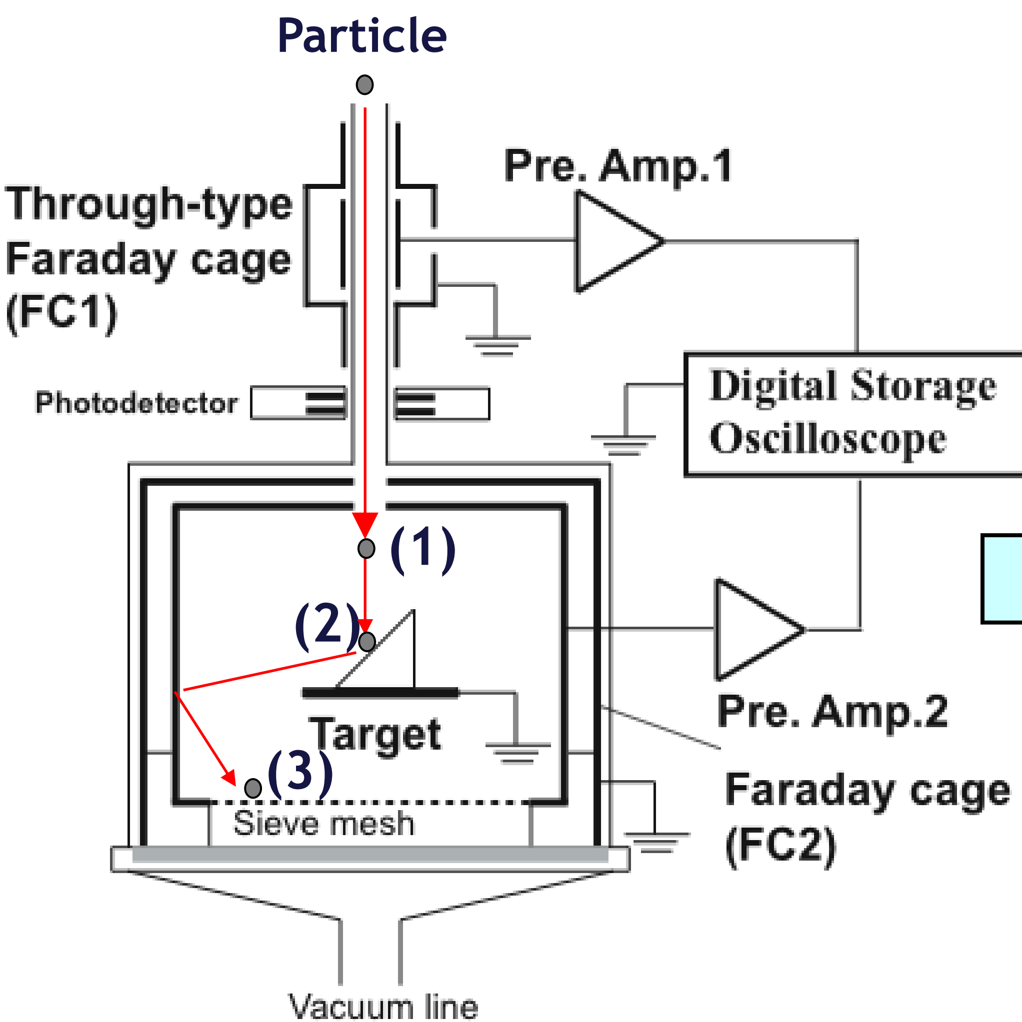
Impact Charging Test Rig



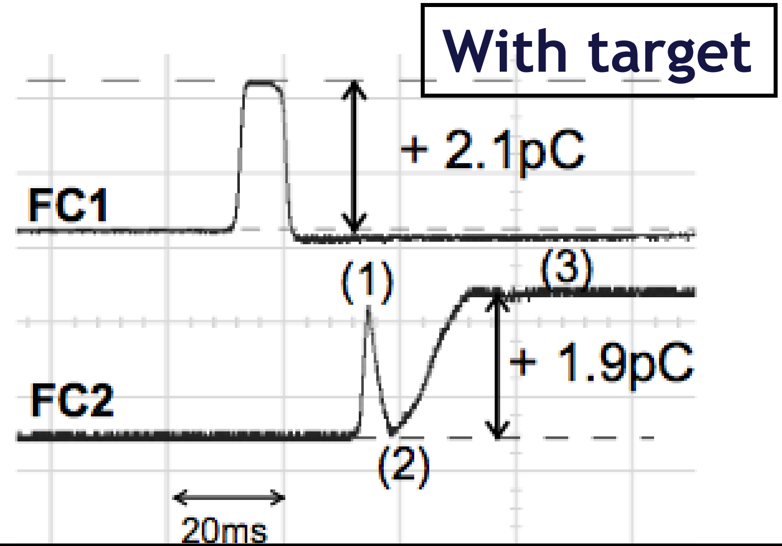
Watanabe et al.(2007)



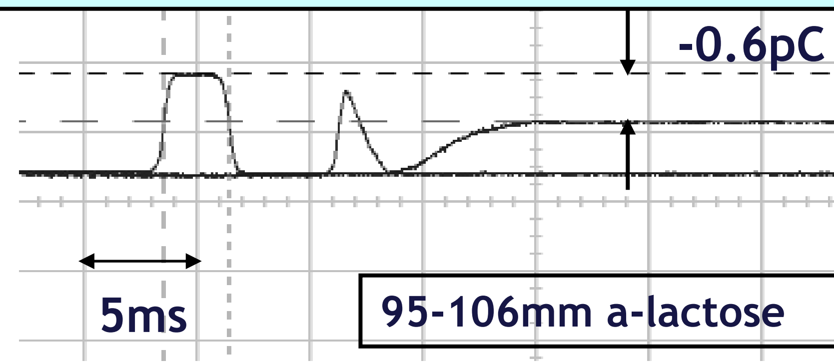
Charge Measurement



Impact charging test rig



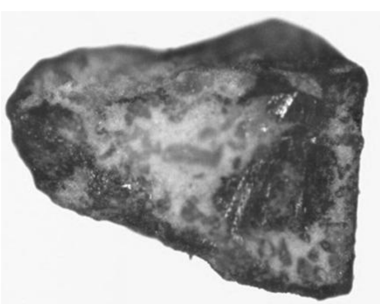
Impact charge (FC2-FC1) = **-0.2pC**



Impact charge with particles down to **100 μm** in size can be measured even by one-by-one feeding test.

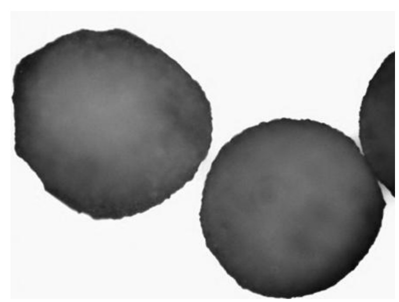
Test Conditions

- **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)

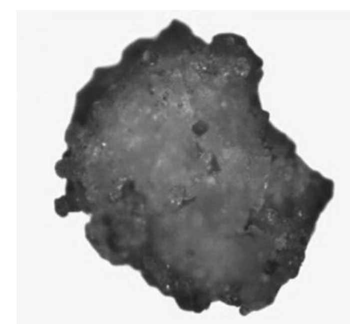


α-lactose monohydrate (α-LM)

Aspirin (acetylsalicylic acid) (ASP)

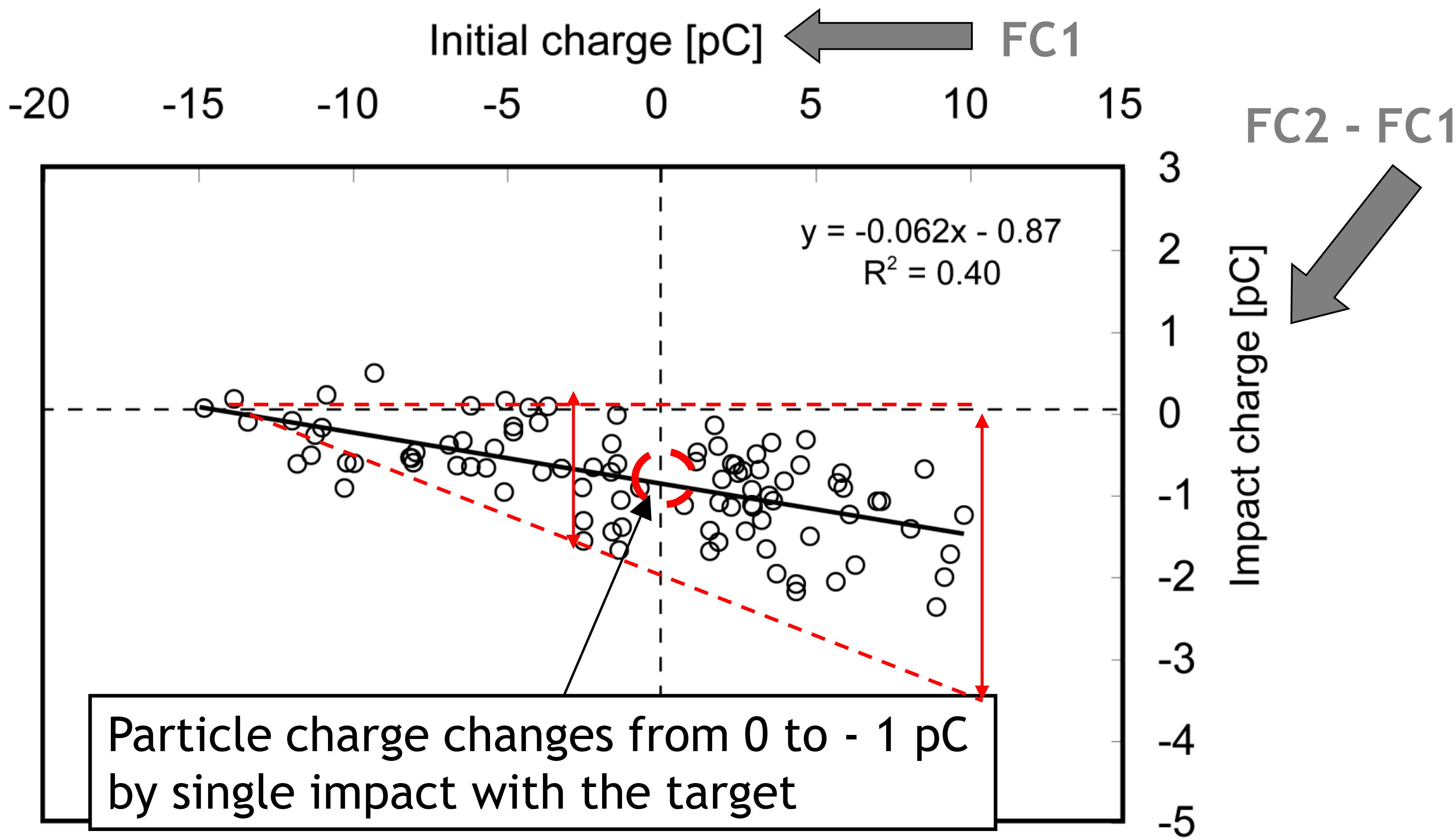


Sugar granules (mainly sucrose) (SG)



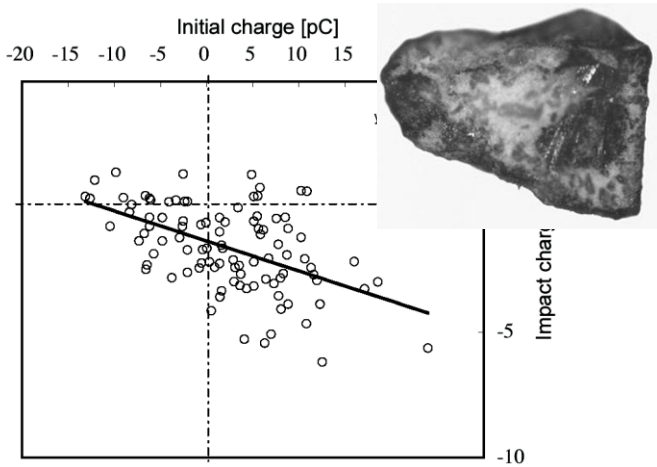
Ethylcellulose (granules) (EC)

Typical Results: Impact Charging Test

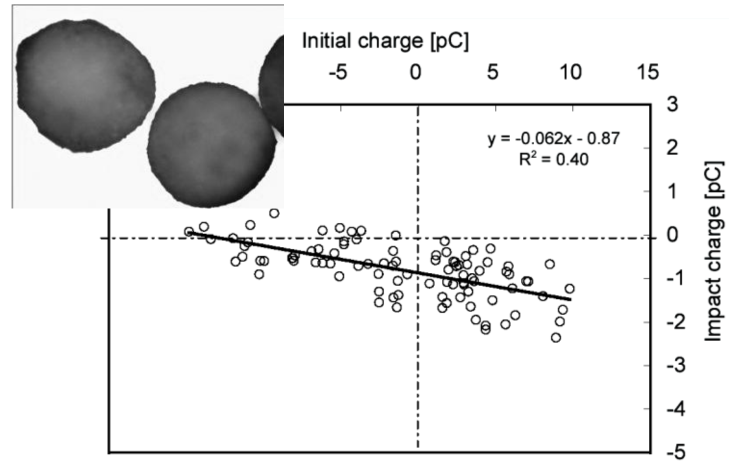


\# Sample: **Sugar granules**. Impacted at **9.0 m/s** with impact angle **30º**.

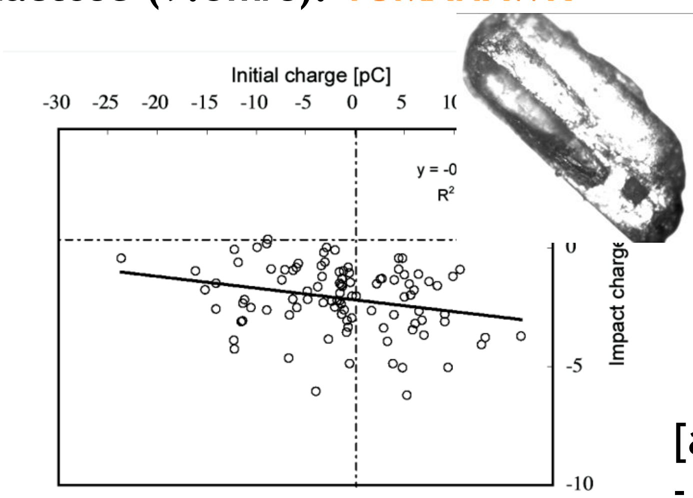
Data Scatter due to Particle Shape



Lactose (9.8m/s): TOMAHAWK

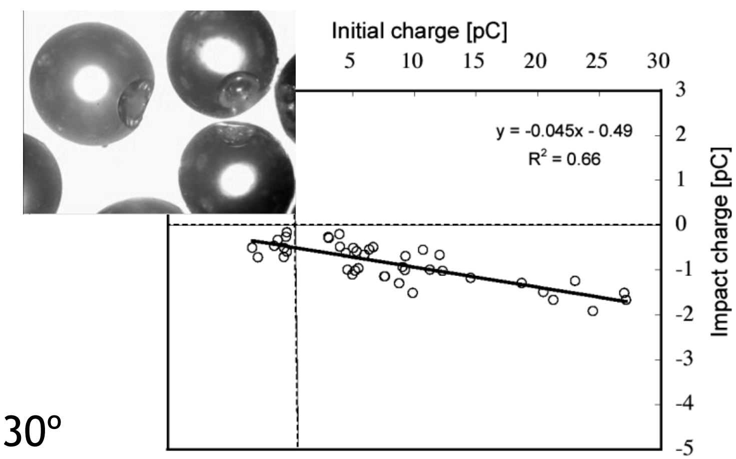


Sugar granules (9.0m/s): nearly SPHERICAL



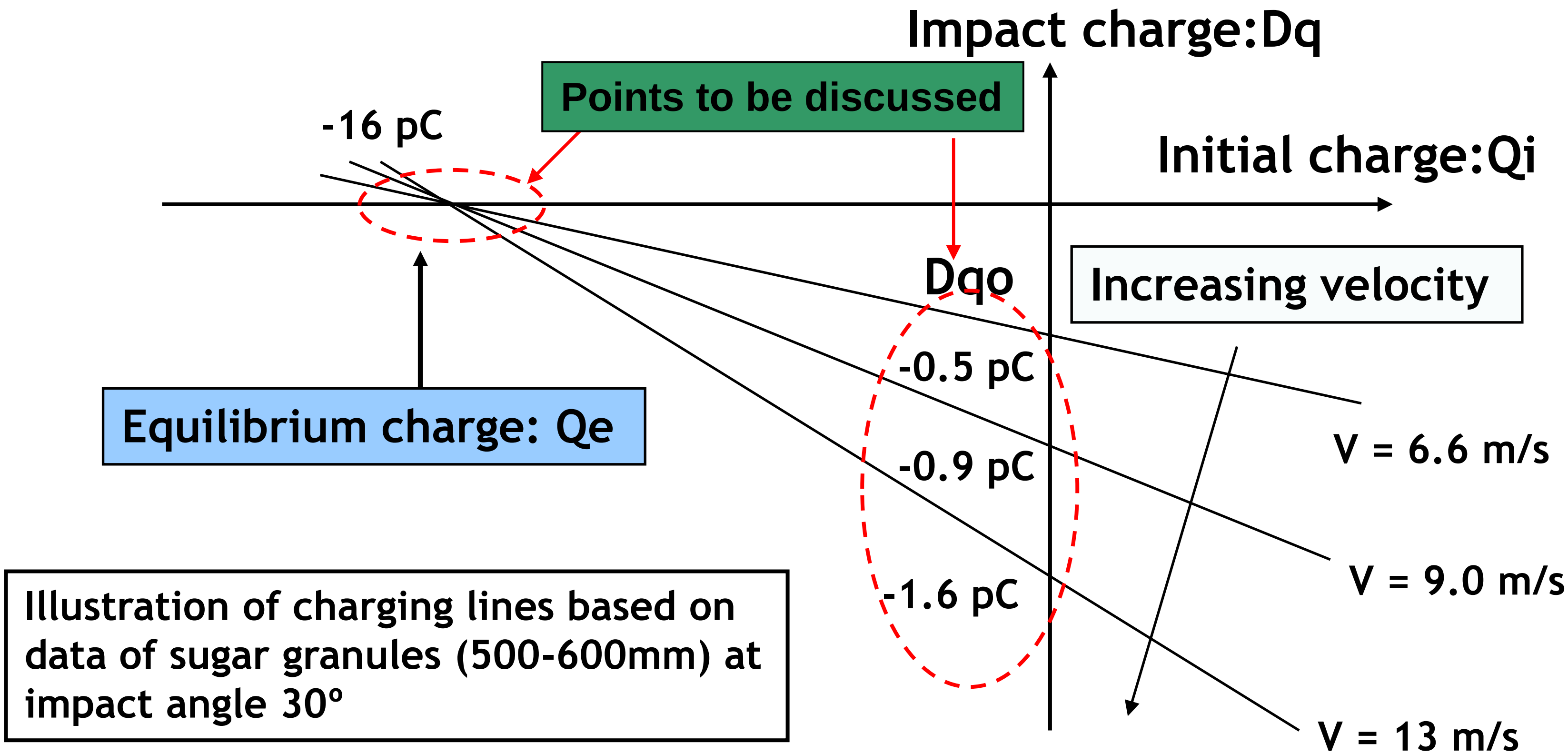
Aspirin (11m/s): OBLONG

[angle 30º
- 4mbar]



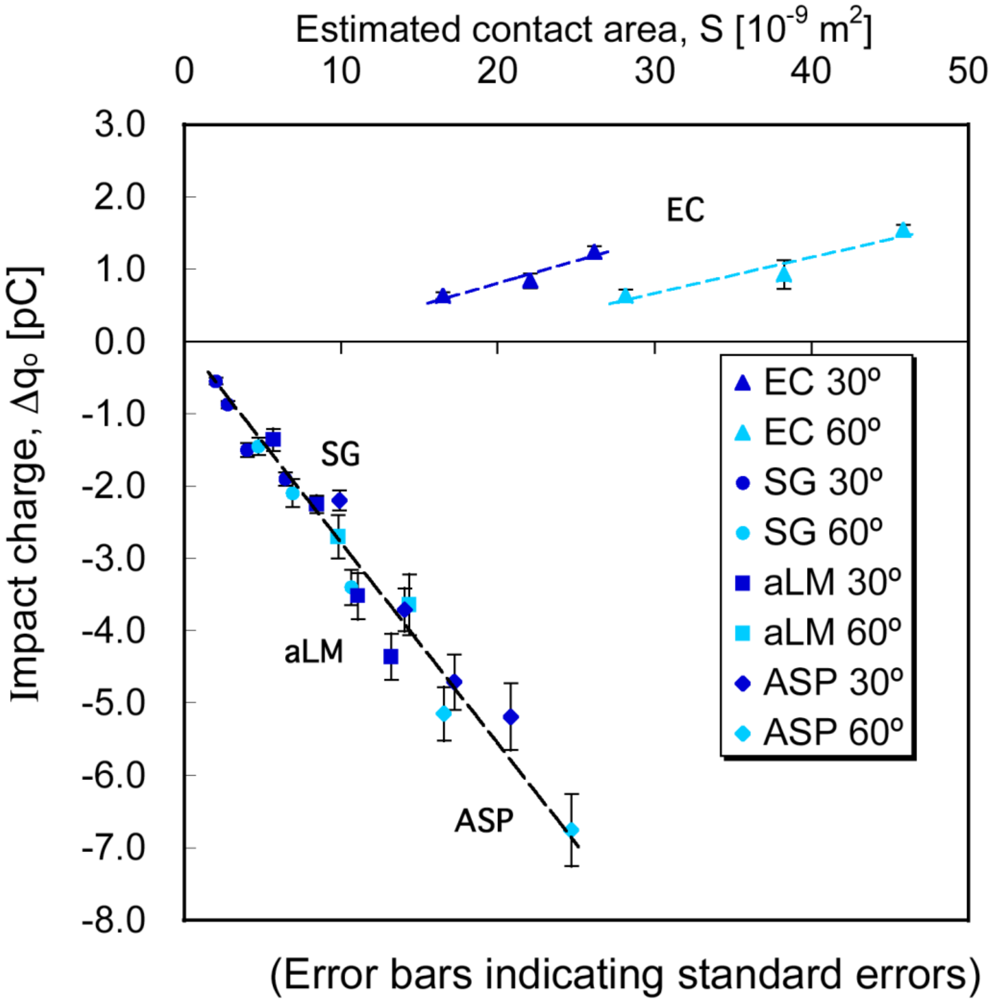
Glass beads (9.7m/s): SPHERICAL

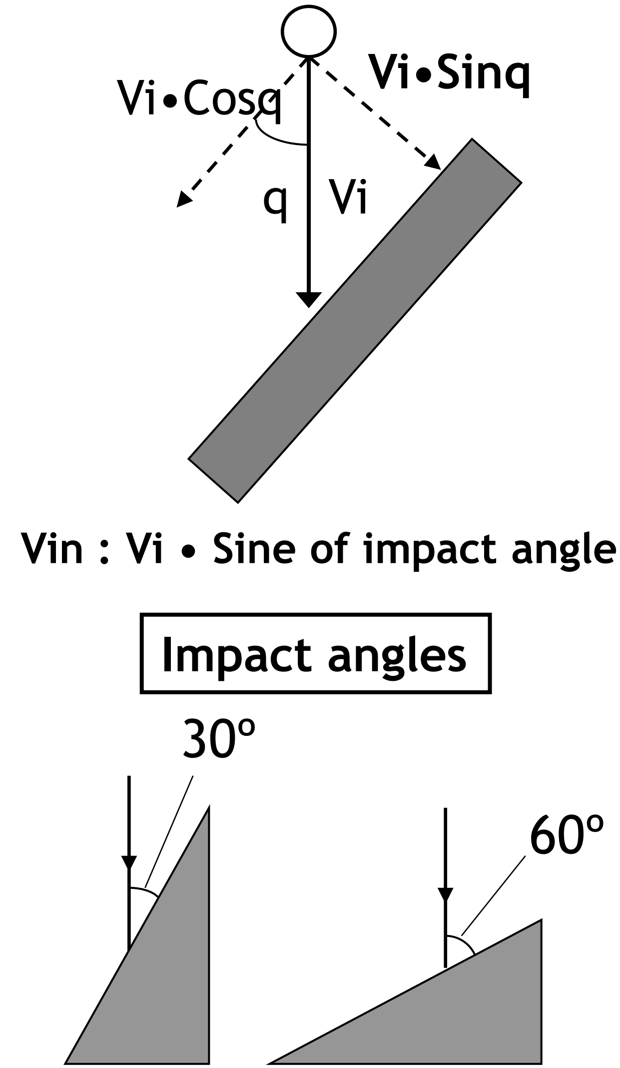
Impact charge vs Initial charge



- 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.

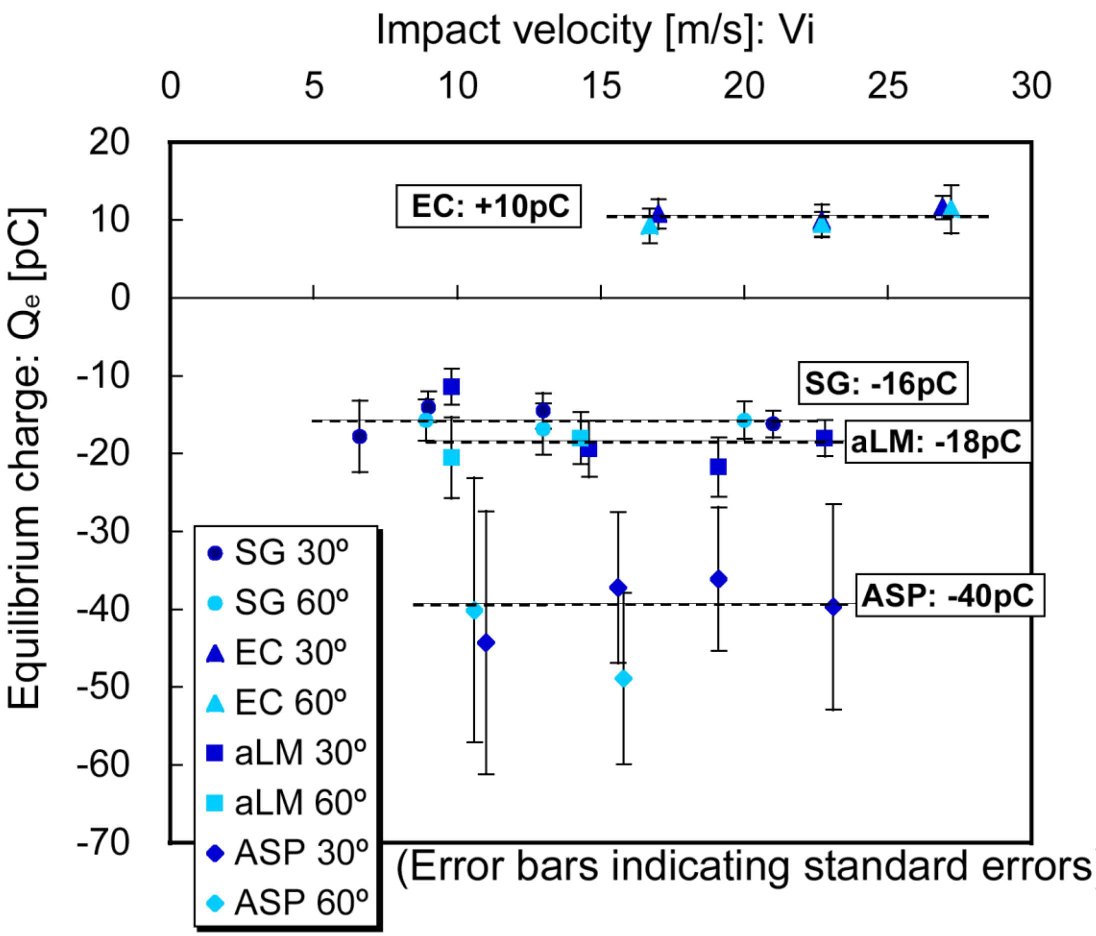
Impact Charge Dqo vs. Contact Area S





Data of SG, aLM and ASP seem to lie on one line except EC.

Equilibrium Charge: Qe



\# Qe is **independent** of impact velocity and angle.

\# Therefore, **Qe** is an *important material characteristics*.

Triboelectric Series vs Qe

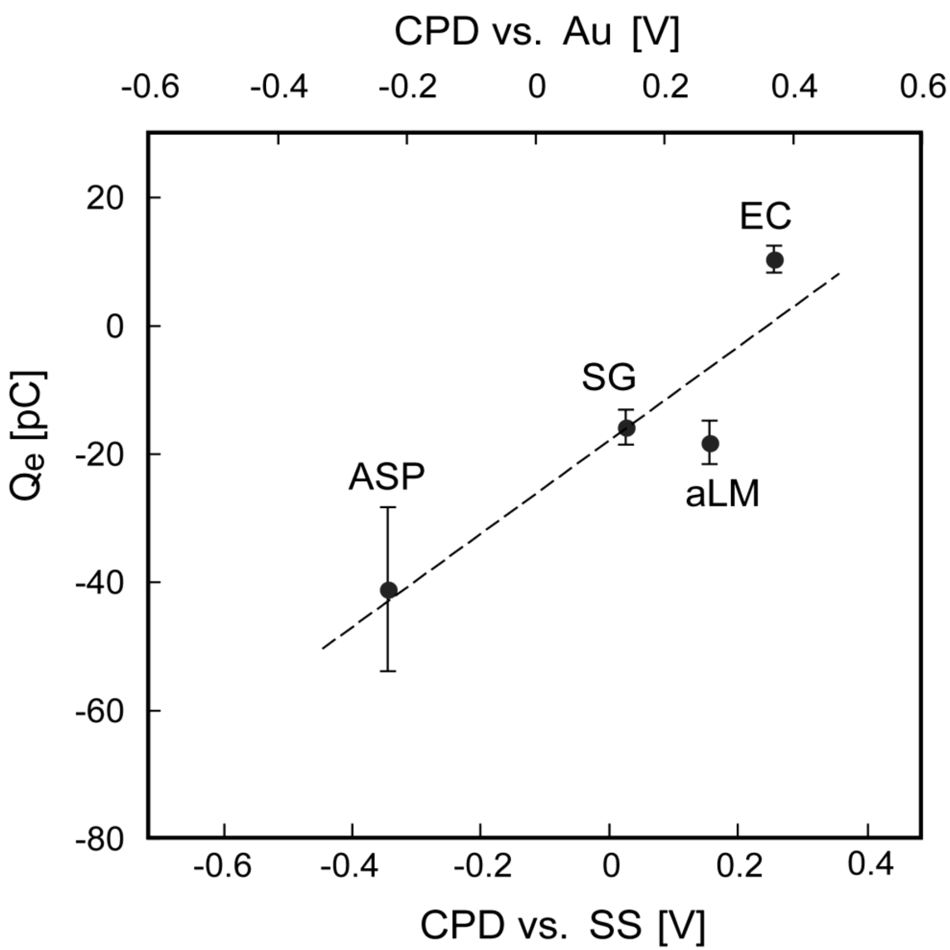
Positive end	Qe vs SS
EC	+ 10 pC
GS, Perspex, SS, Al	
aLM, SG	- 18, -16 pC
PP	
ASP	- 40 pC
PTFE	
Negative end	

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.*

Qe vs Contact Potential Difference (CPD)



CPD of sample vs SS
= CPD of sample vs Au
- CPD of SS vs Au

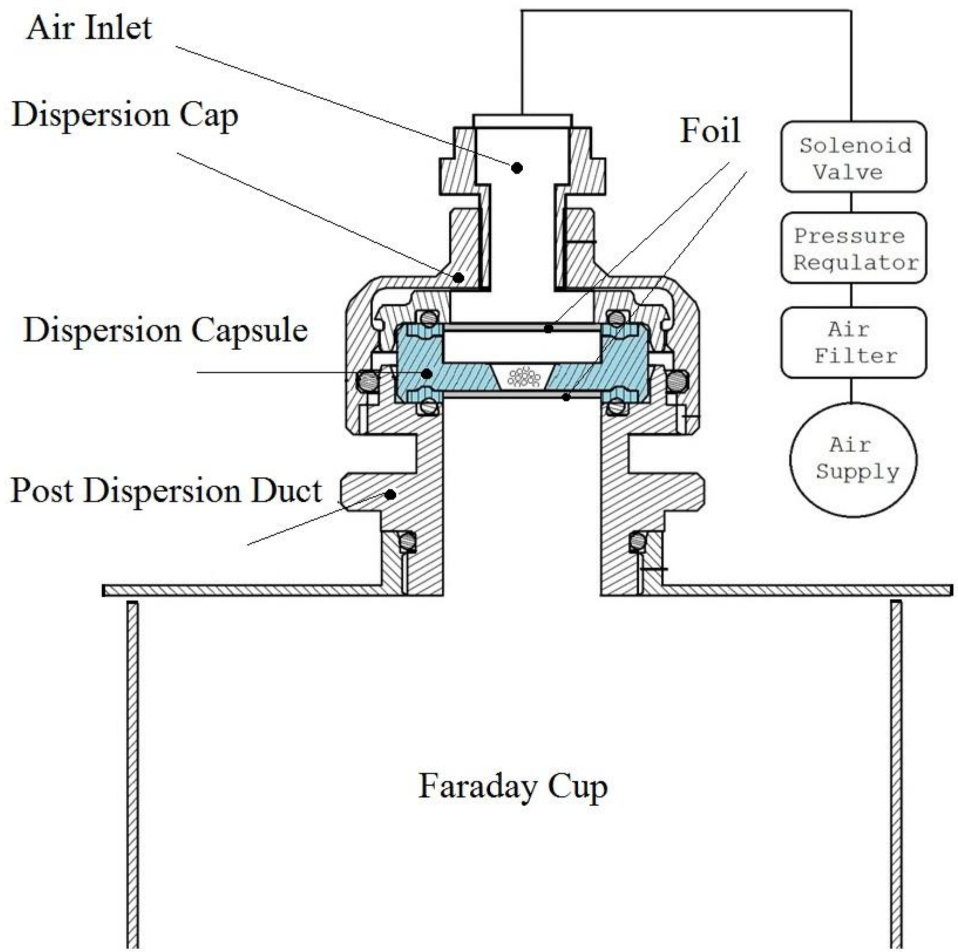
CPD of target (SS)
= +0.12 V vs Au

Contact Potential Difference (CPD) of the sample powders was measured by a system that has been developed at Kyoto University.

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

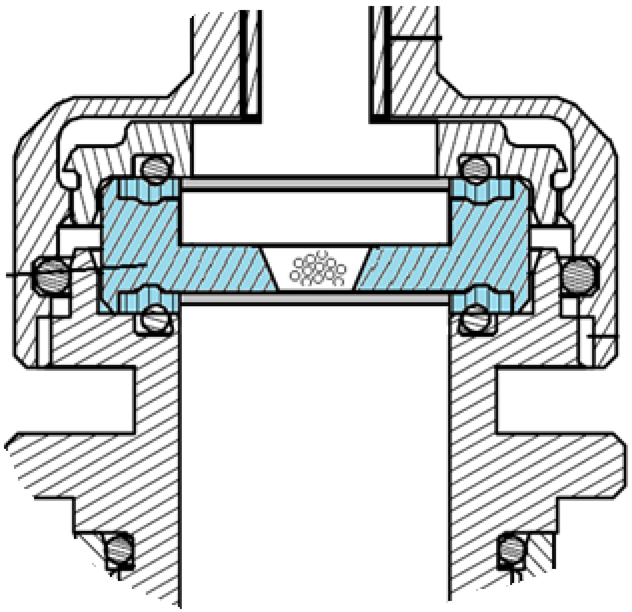
From Single Particle to Bulk

Charging by Aerodynamic dispersion by bursting foils



Zarrebini, A., Ghadiri, M., Dyson, M., Kippax, P., and McNeil-Watson, F. , "Tribo-electrification of Powders Due to Dispersion", *Powder Technology*, 250, 75-83.

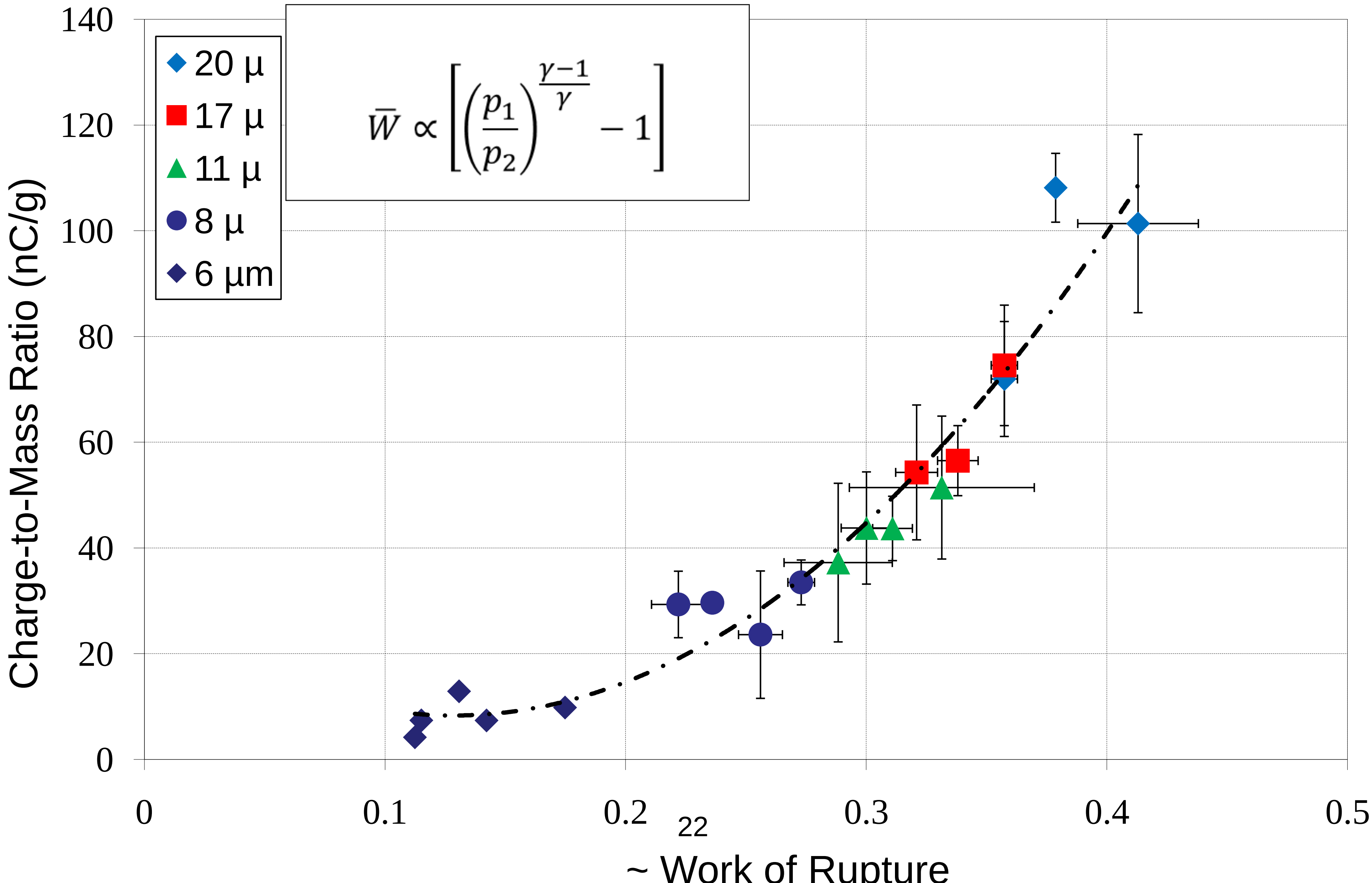
Effect of Foil Material on Tribo-electric Charging of Glass Beads



Material	Thickness (µm)
Stainless Steel	5
Copper	12.7
Aluminium	6

Methodology

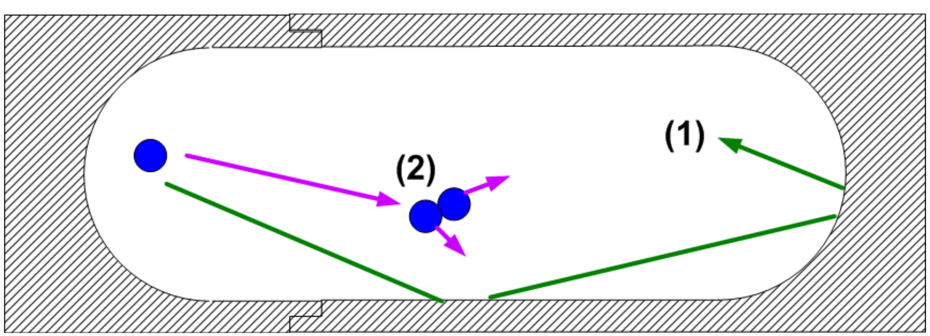
Q/M vs. Corresponding Work (Glass Beads)



Charging by Shaking

Tribo-electrification Approach:

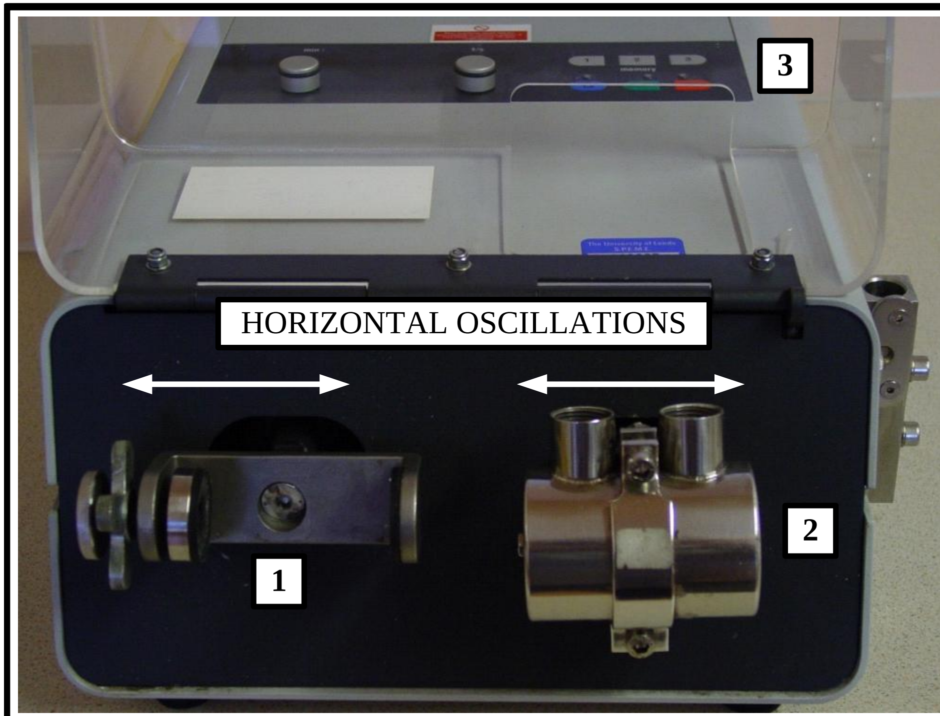
- Development of simple test method for characterising the tribo-electrification properties of powder materials, consisting of Retsch® shaking machine, set of interchangeable shaking containers made out of common industrial materials, electrometer, Faraday cup, and isolator booth.
- The container is subjected to motions in a horizontal direction:



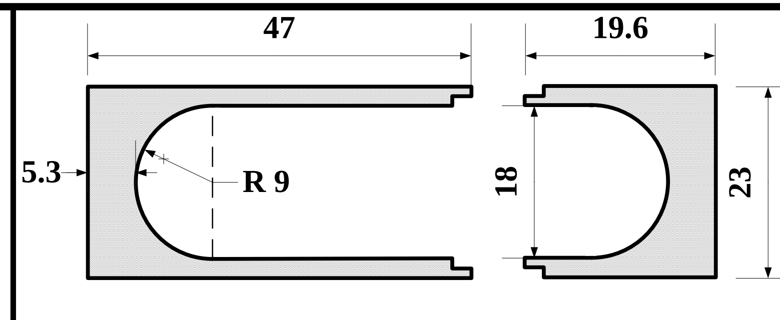
(1) Multiple particle-wall interactions
(2) Multiple particle-particle interactions
(3) Space charge effects

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



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

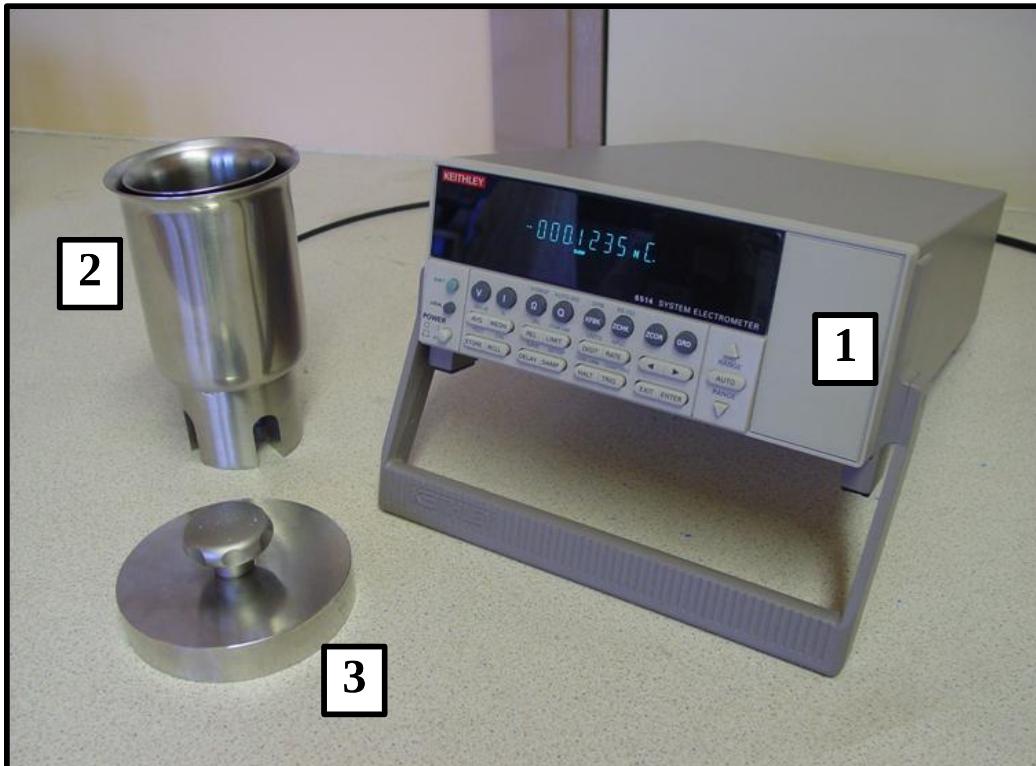


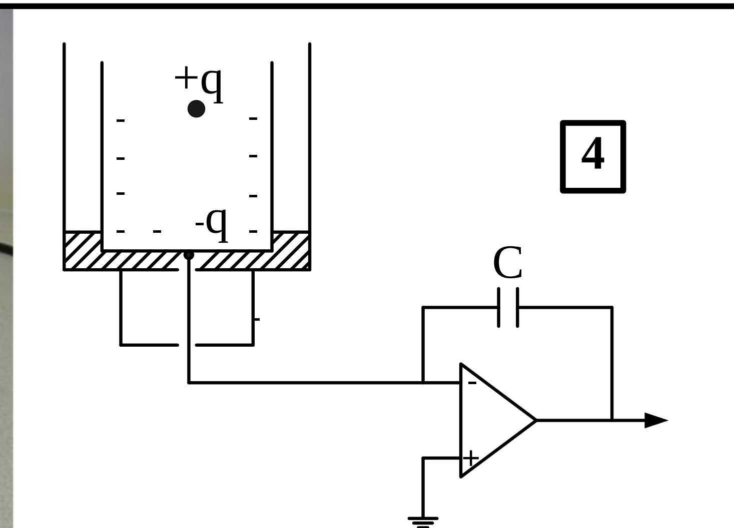
Schematic diagram of a shaking container, (all measurements in mm).

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.

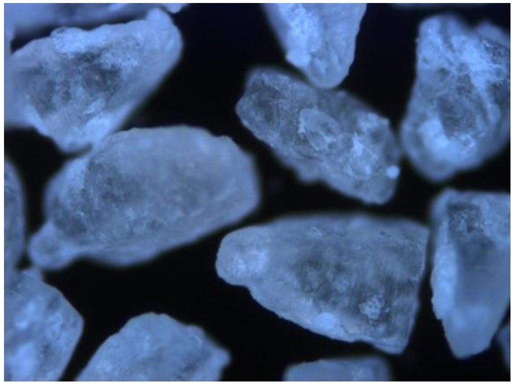




1: Electrometer, 2: Faraday cup, 3: Faraday cup lid, and 4: Schematic diagram of the setup.

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



α-LM

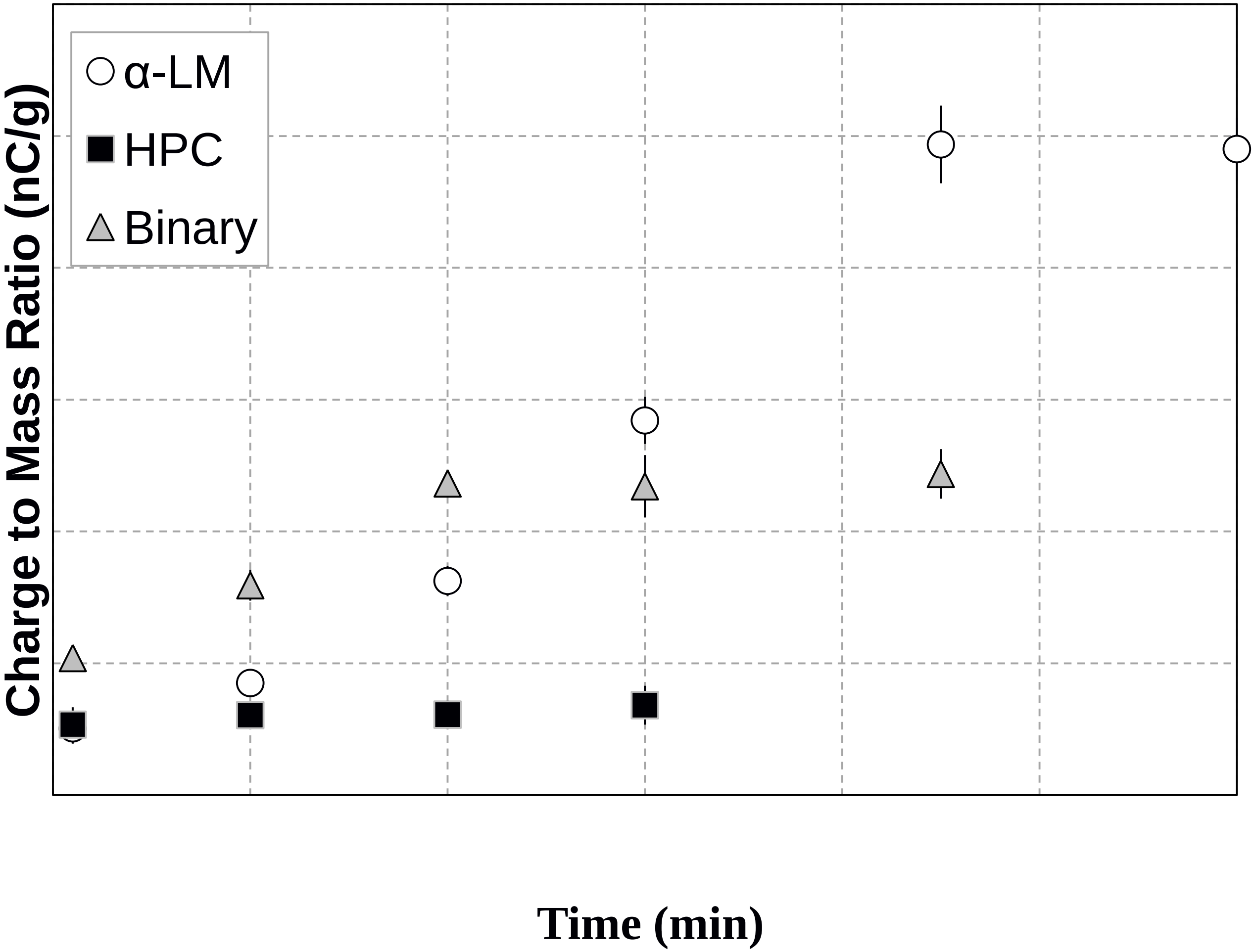
HPC

- 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.

Humidity (%)	48.2
Container	PTFE
Frequency	20 Hz



Frequency

Frequency [Hz]	Saturated Charge [nC/g]		
	α-LM	HPC	Binary
10	24.6	3.3	11.4
20	24.7	3.2	11.9
30	22.7	3.8	12.2

- **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)

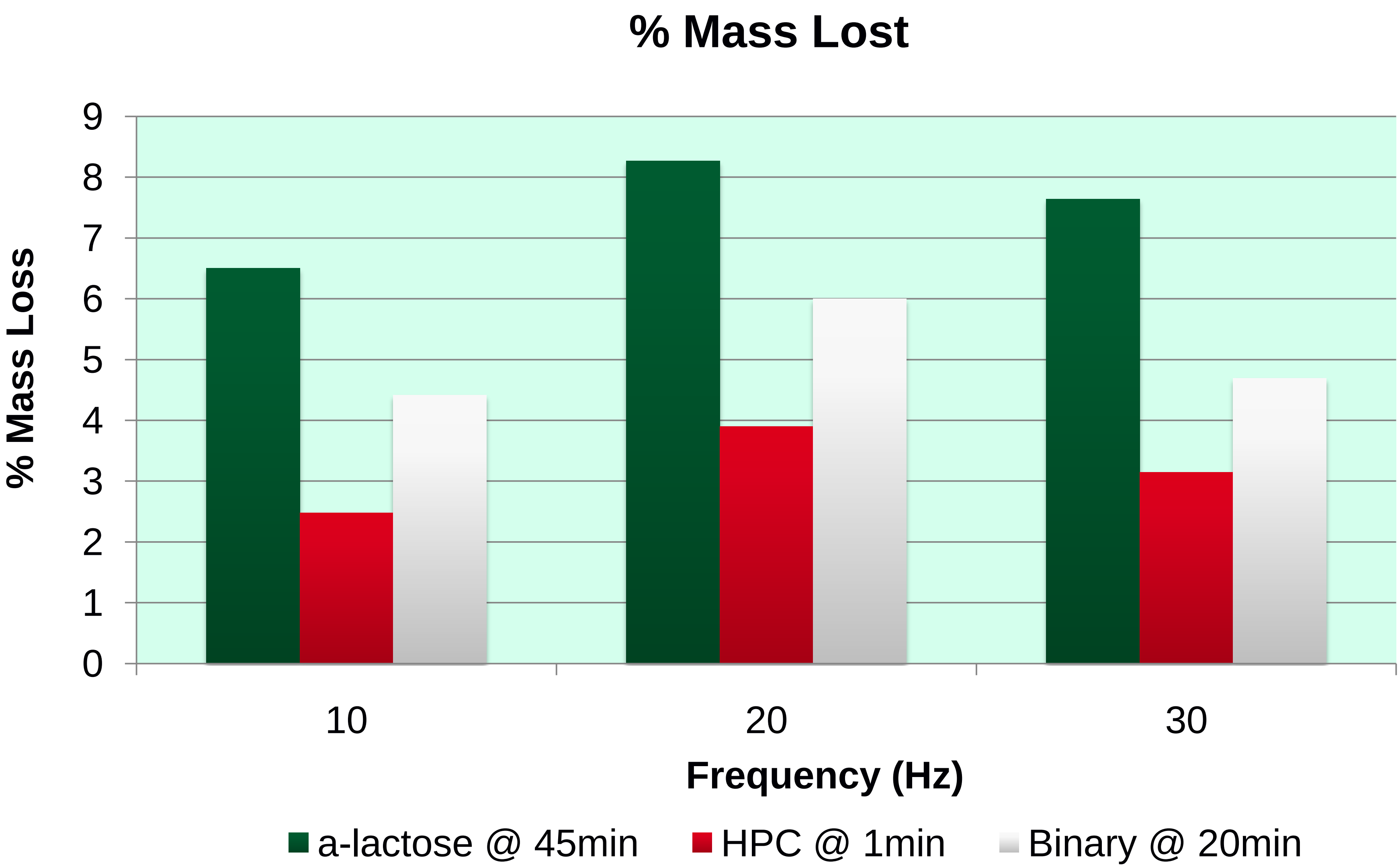
- Particles/debris adhered to the inner wall of the shaking vessel carry the highest magnitude of charge and can have a significant effect on the generated charge.

- 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.

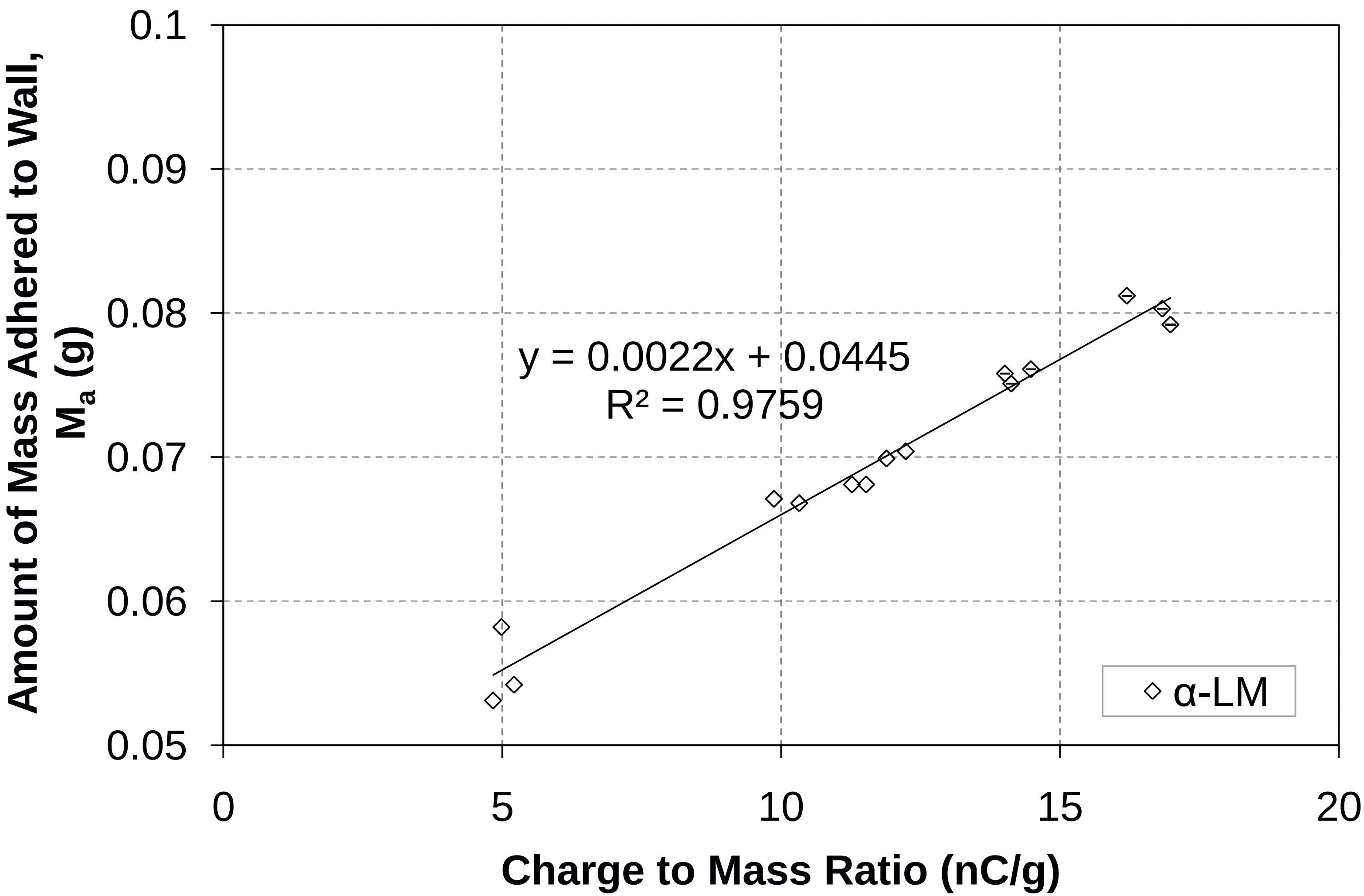
- The amount of ‘lost’ powder is a good indicator of the extent of tribo-charging that is taking place in the vessel.

Mass Loss



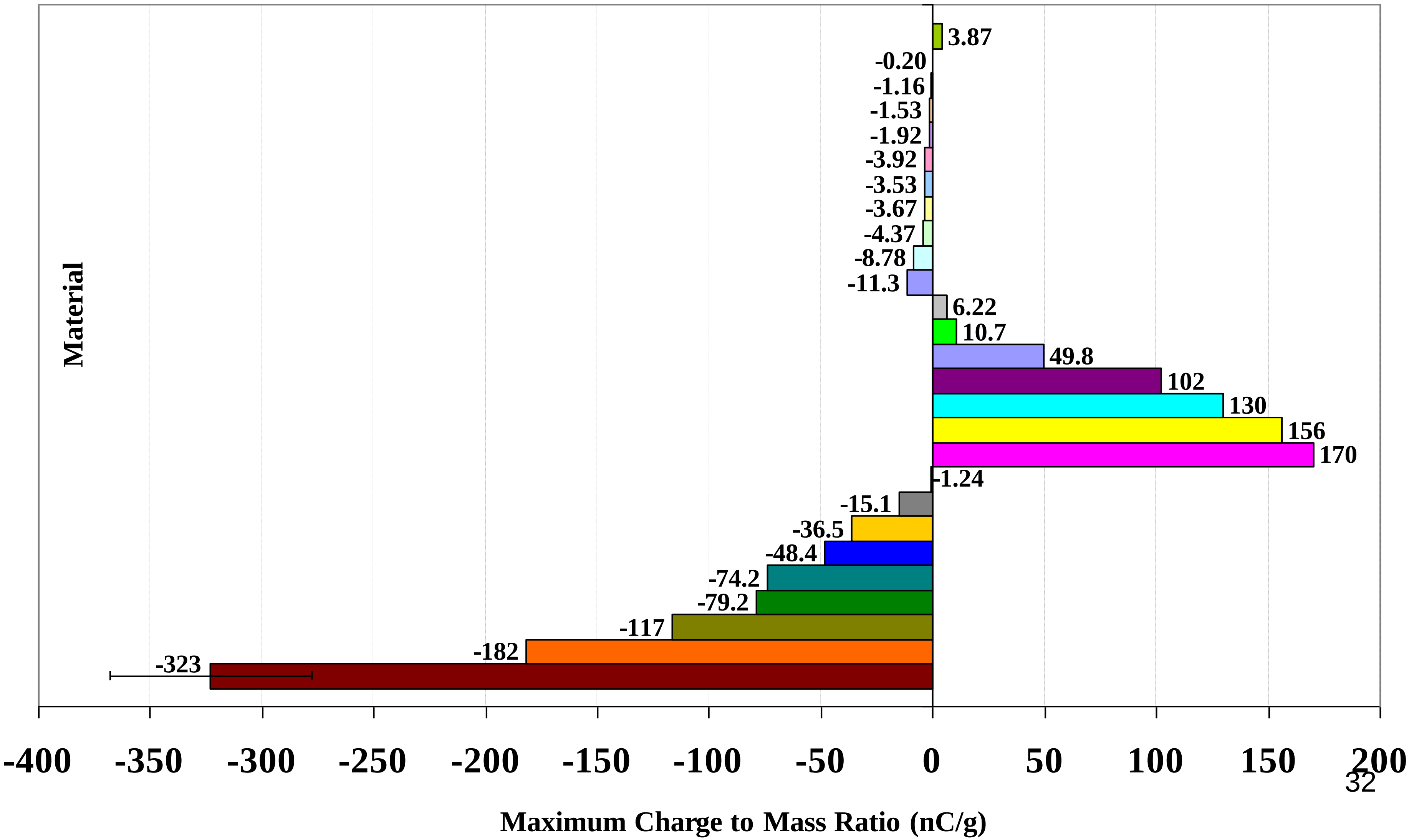
% Mass Lost
% Mass Loss
0
1
2
3
4
5
6
7
8
9
10
20
30
Frequency (Hz)
a-lactose @ 45min
HPC @ 1min
Binary @ 20min

Wall Adhesion



Chargeability: Excipients vs API

Supuk *et al*. (2012). Tribo-electrification of active pharmaceutical ingredients and excipients. Powder Technology, 217, 427-434.



Chargeability: Excipients Vs API

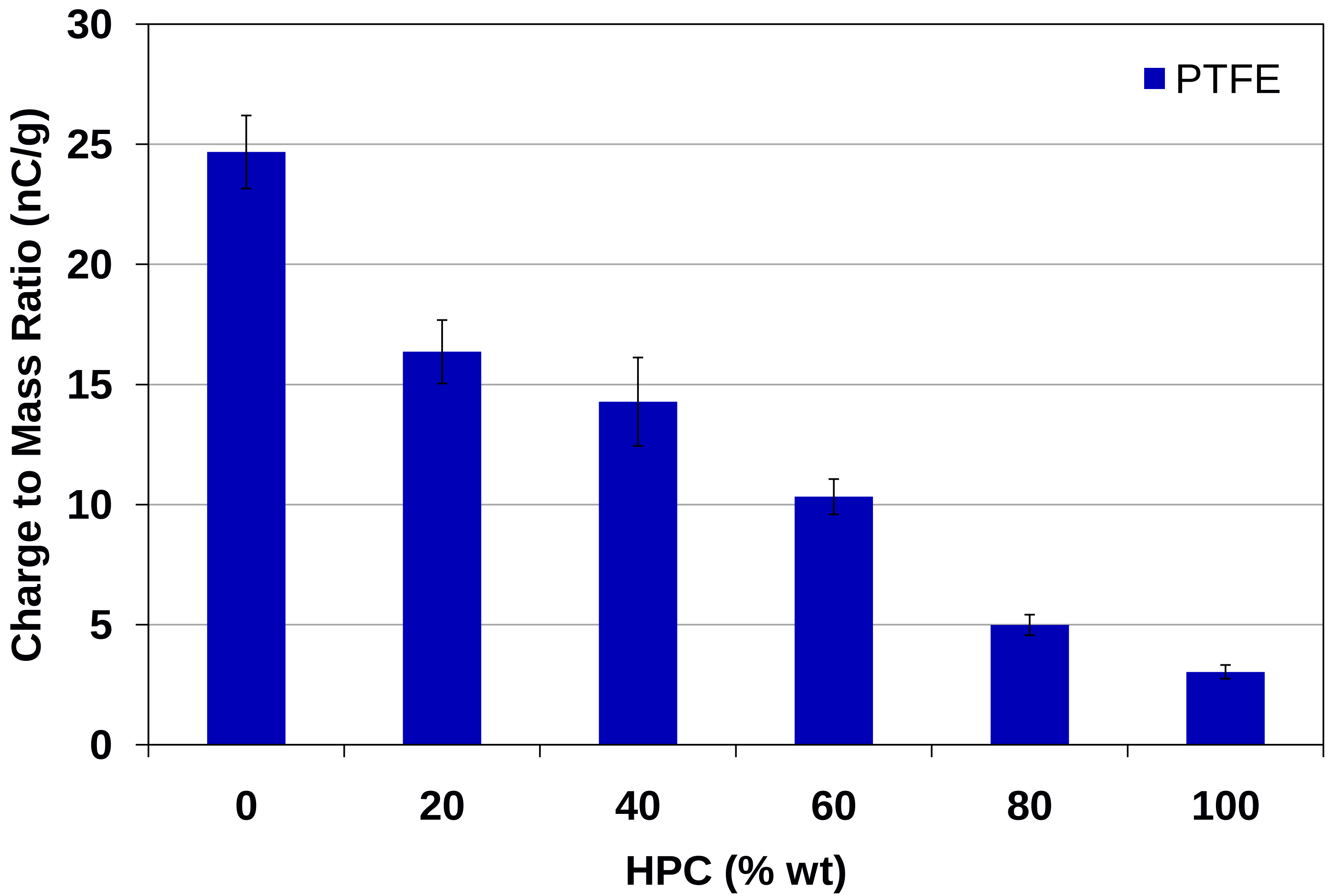
API (-)		API (+)		EXC (-)		EXC (+)	
Q_{Max}	Material	Q_{Max}	Material	Q_{Max}	Material	Q_{Max}	Material
-323	CB	170	CA4	-11.3	MAN	3.87	LA-NF
-182	KTS	156	FBP	-8.78	PAN		
-117	ASP	130	CF	-4.37	LA-250		
-79.2	SCA	102	CA3	-3.92	SID		
-74.2	AAP	49.8	CE	-3.67	MGST		
-48.4	CC	10.7	CA2	-3.53	CCS		
-36.5	THP	6.22	CD	-1.92	LA-21		
-15.1	CA1			-1.53	MCC		
-1.24	PRS			-1.16	SP		
				-0.20	DCP		
*Q_{Max} - 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 ºC and a relative humidity range of 20-55 % for selection of excipients and active materials.

Tribo-Electrification in Binary Mixtures

- 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



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:

$$q_{mixture} = x_1 q_{\alpha-LM} + x_2 q_{HPC}$$

x1 and x2 are mass fractions of α-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.

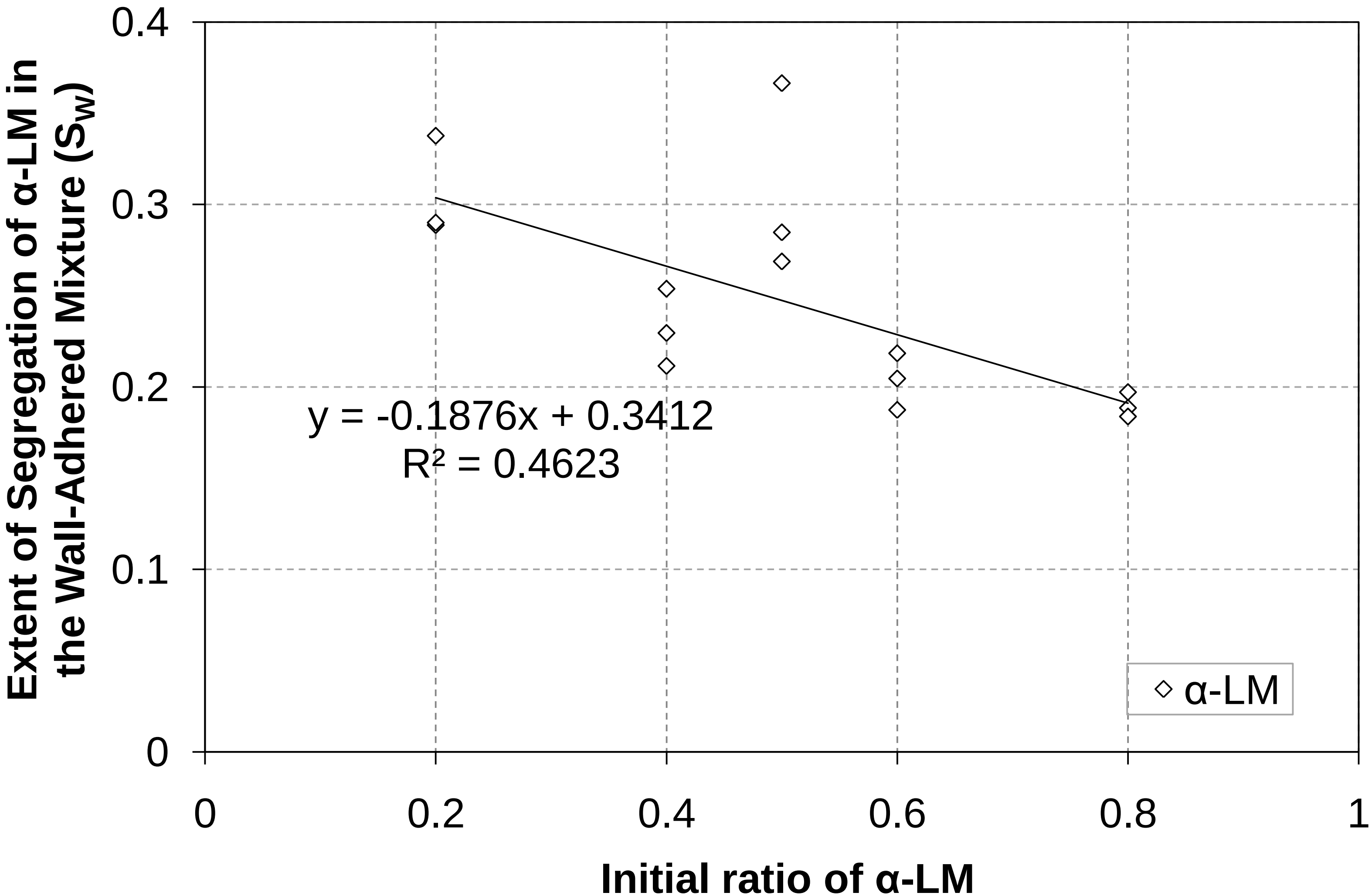
Segregation Wall-Adhered Mixture

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

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

- As SW increases from zero the more segregated the mixture on the wall is.

Segregation Wall-Adhered Mixture



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)

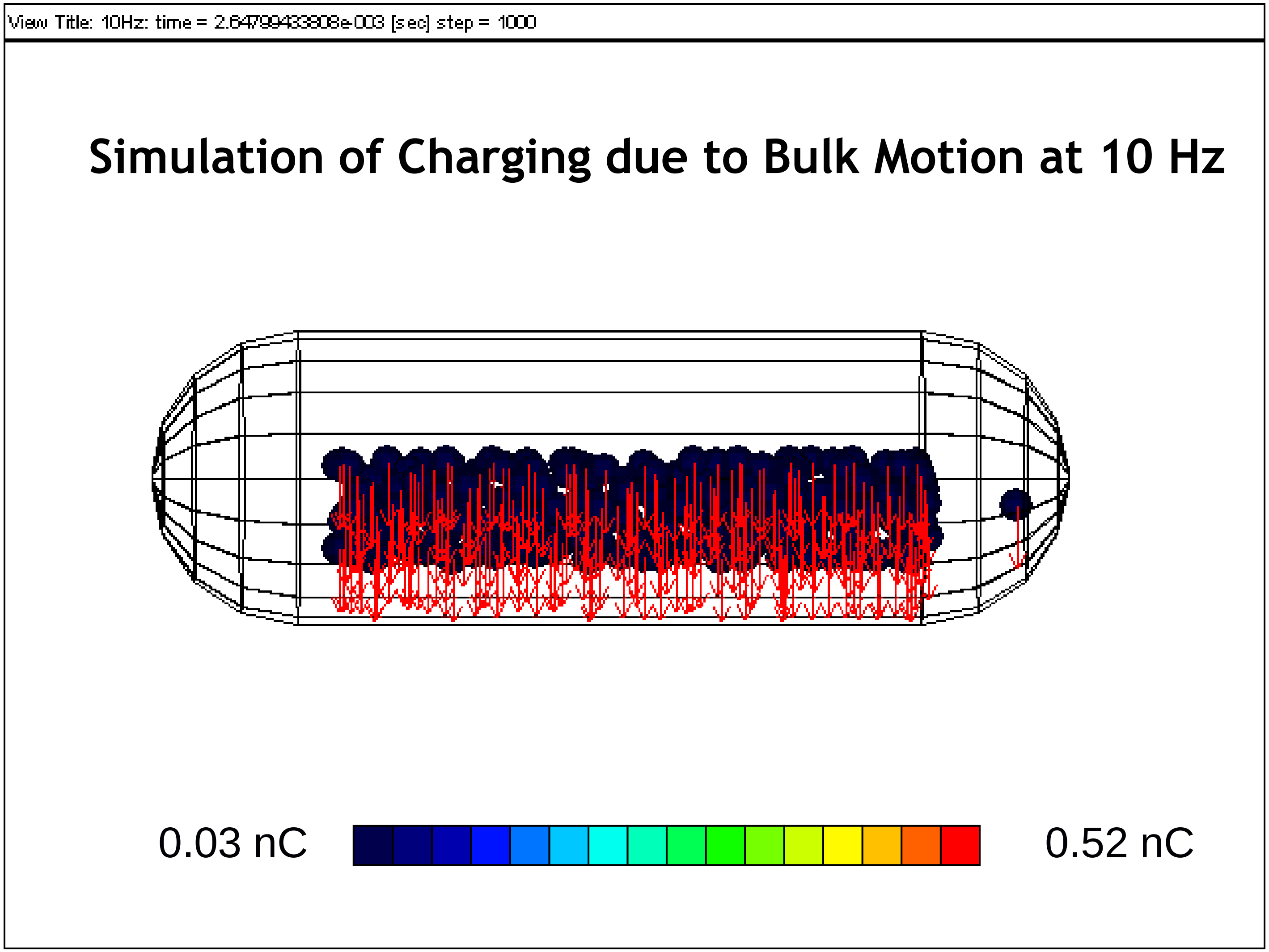
- Binary mixtures showed a decrease in the net charge as the HPC mass fraction increased.

Summary

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

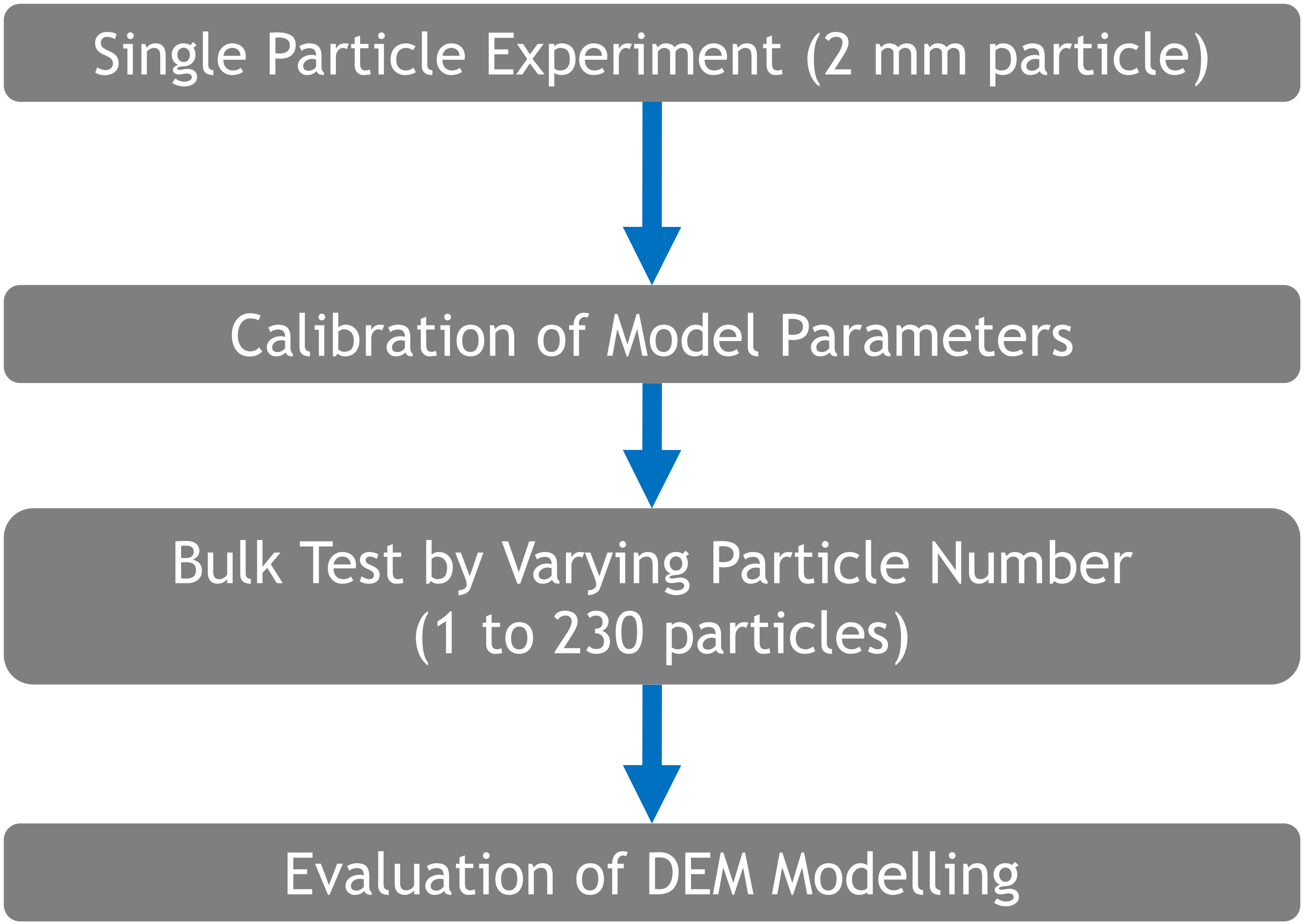
- **Wall-adhered mixture:**
  The extent of segregation is considerably higher than the main mixture with the highest value being 0.31 for the 20:80 ratio mixtures and the lowest of 0.19 for the 80:20 ratio mixtures.

Analysis of Charging by Shaking

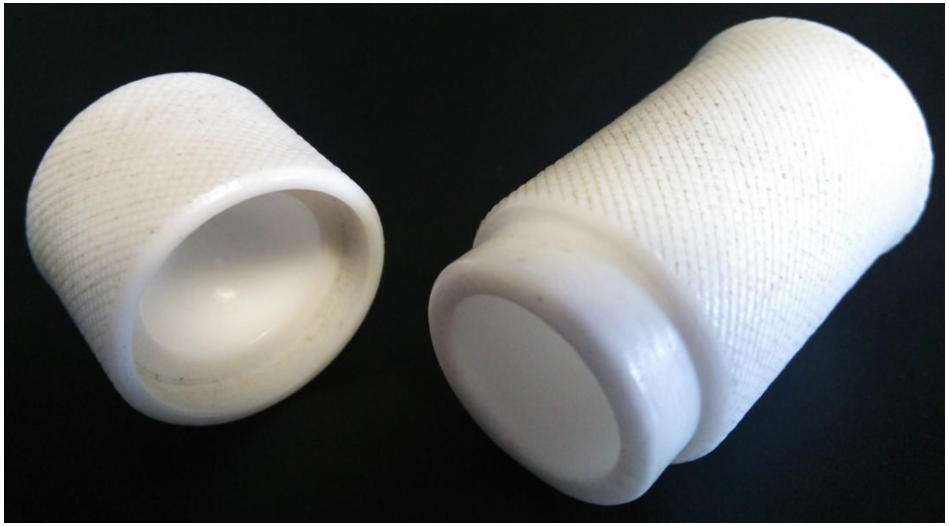


Imba M., Zarrebini A., Matsuyama T., Ghadiri M. (2013). Tribo-Electric Charging of Particles in a Shaker, Partec 2013.

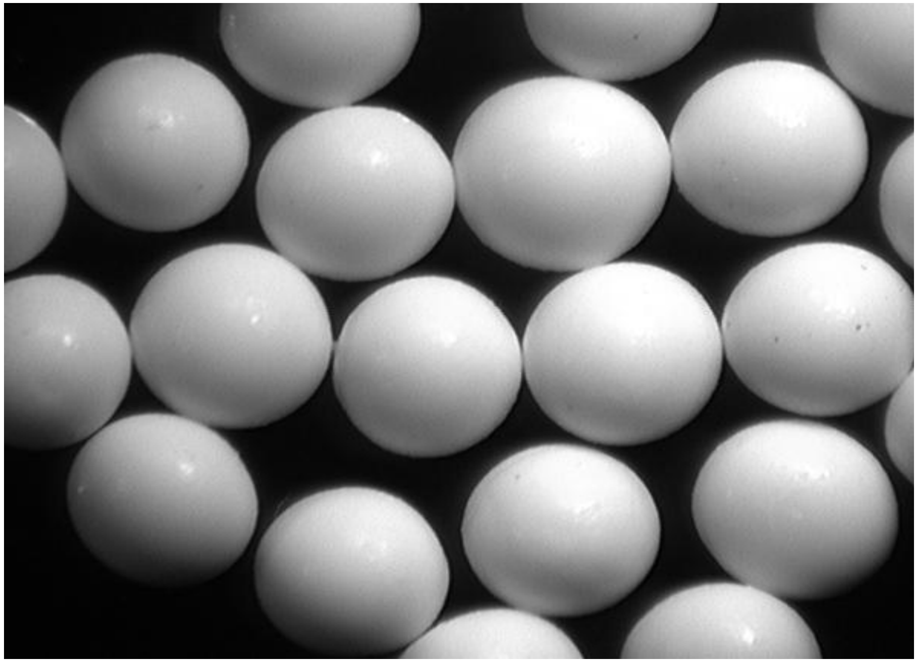
Strategy


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

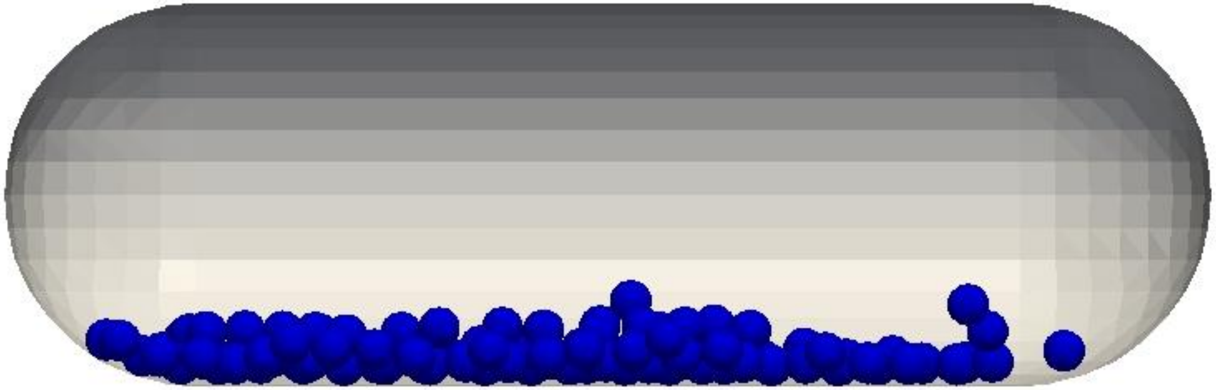
Materials



PTFE capsule



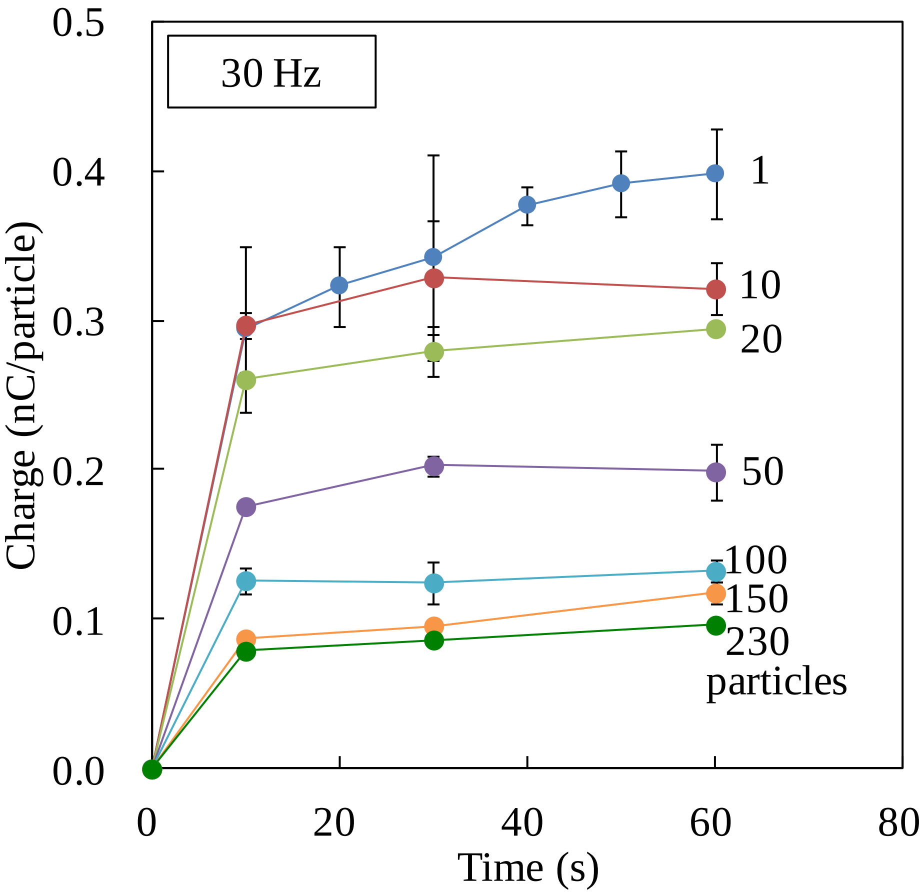
Alumina beads



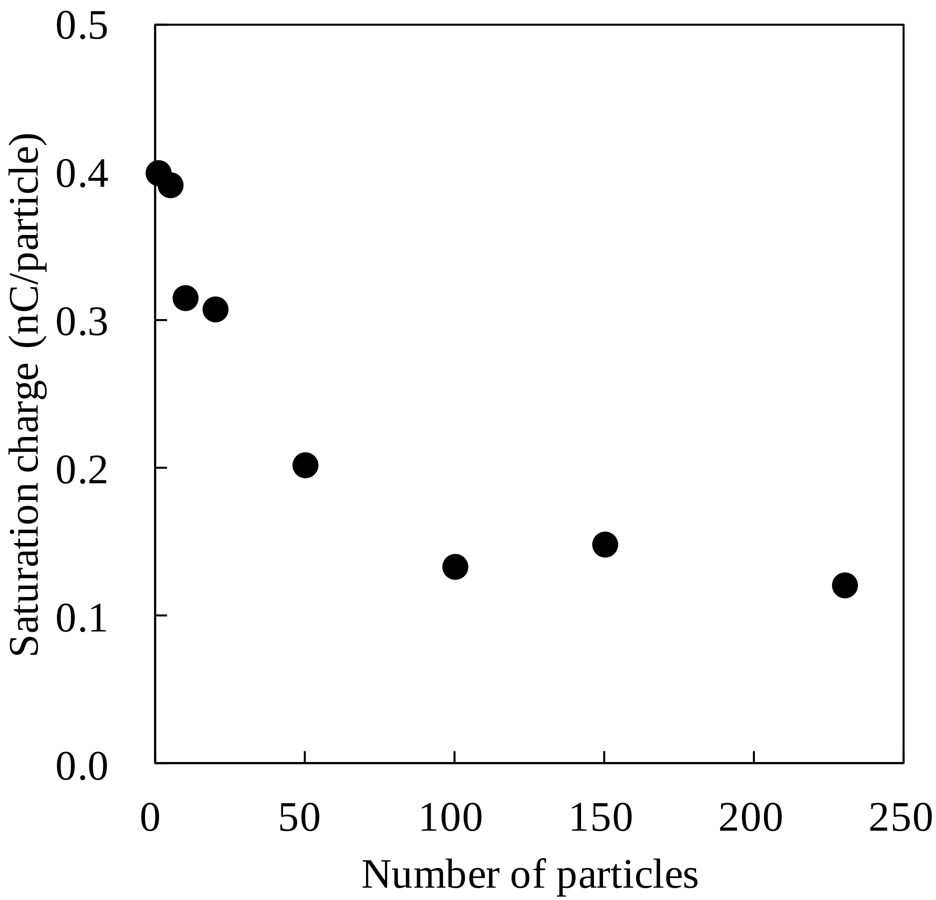
Experimental conditions

Particle	Alumina (2.01 mm)
Capsule	PTFE
Amplitude	10 mm
Frequency	30 Hz
Number of particles	1-230
Shaking time	10-60 s

Results



30 Hz
Charge (nC/particle)
0.0
0.1
0.2
0.3
0.4
0.5
0
20
40
60
80
Time (s)
1
10
20
50
100
150
230
particles



Saturation charge (nC/particle)
0.0
0.1
0.2
0.3
0.4
0.5
0
50
100
150
200
250
Number of particles

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

Terms of electrostatic models

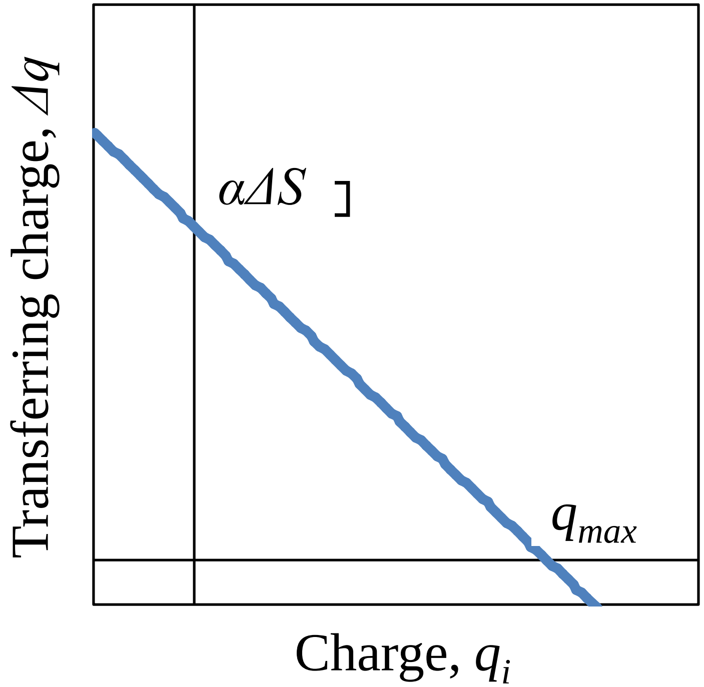
1. Contact charging model

2. Model of space charge effect

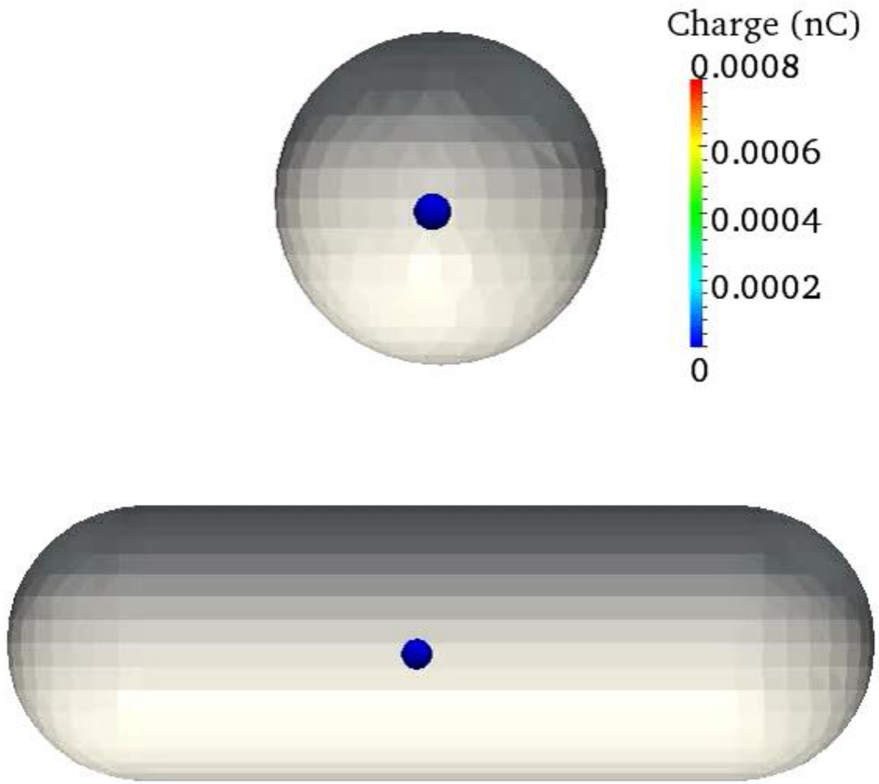
3. Boundary conditions

4. Electrostatic force

1. Contact charging model



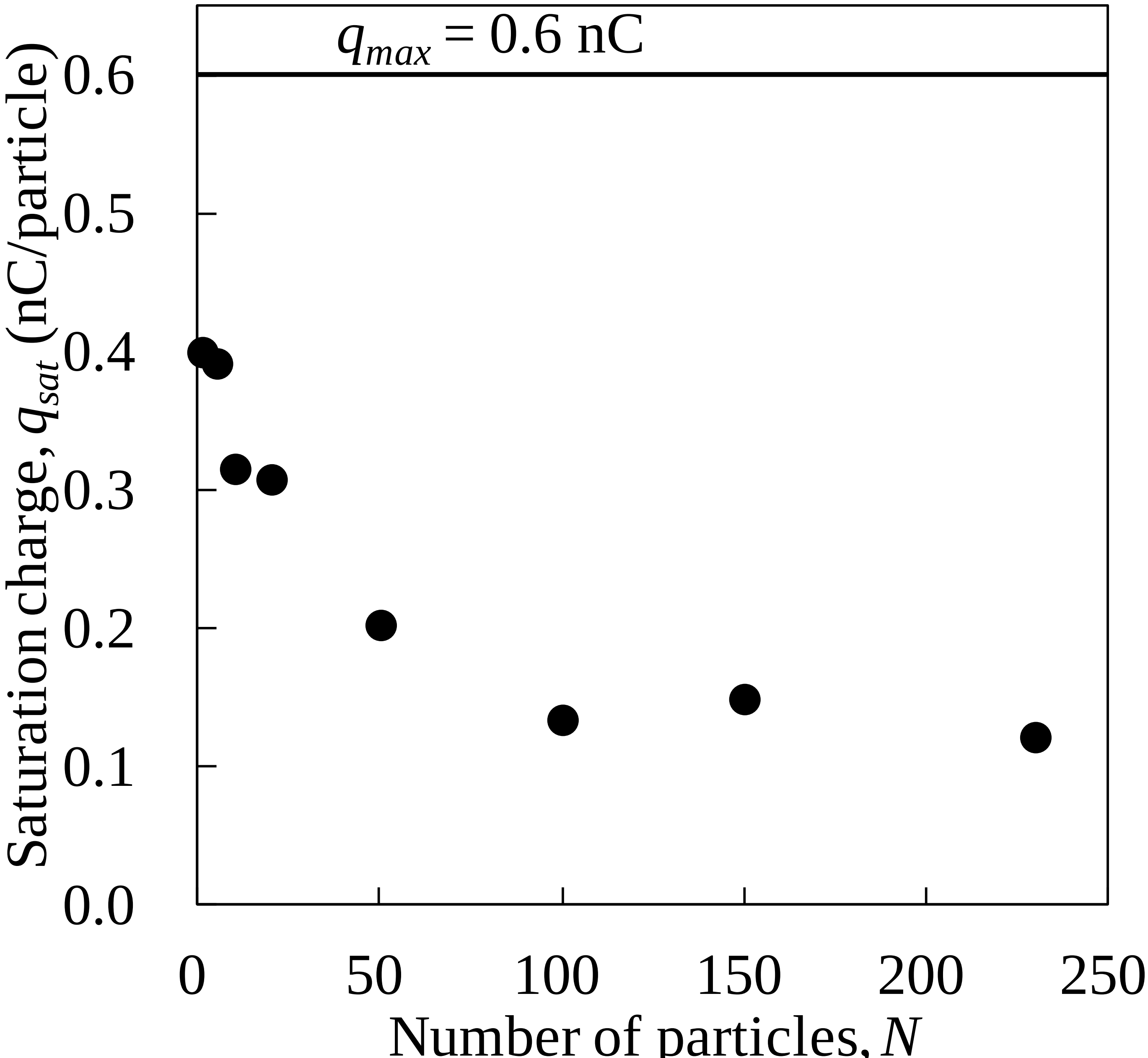
$$\frac{\Delta q}{\alpha \Delta S} + \frac{q_i}{q_{max}} = 1$$



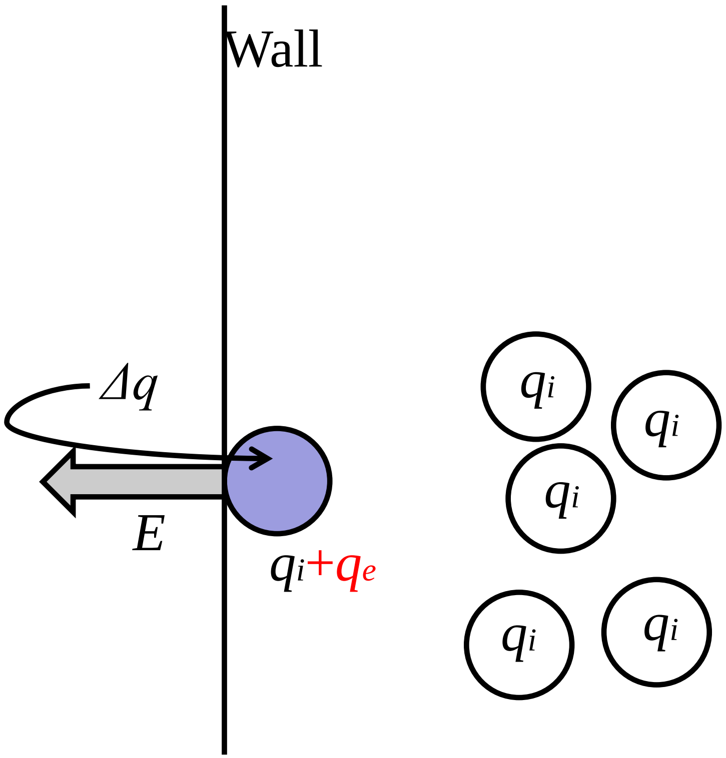
Δq (C): Transferring charge
q_i (C): Charge on particle
ΔS (m^2): Maximum contact area
q_{max} (C): Maximum charge
a (C/m^2): Coefficient ($q_{max}/\pi d^2$)

Matsusaka, S., Ghadiri, M., Masuda, H. (2000), "Electrification of an elastic sphere by repeated impacts on a metal plate", Journal of Physics D: Applied Physics, 33, pp. 2311-2319

1. Determination of Maximum charge q_{max}



2. Model of space charge effect



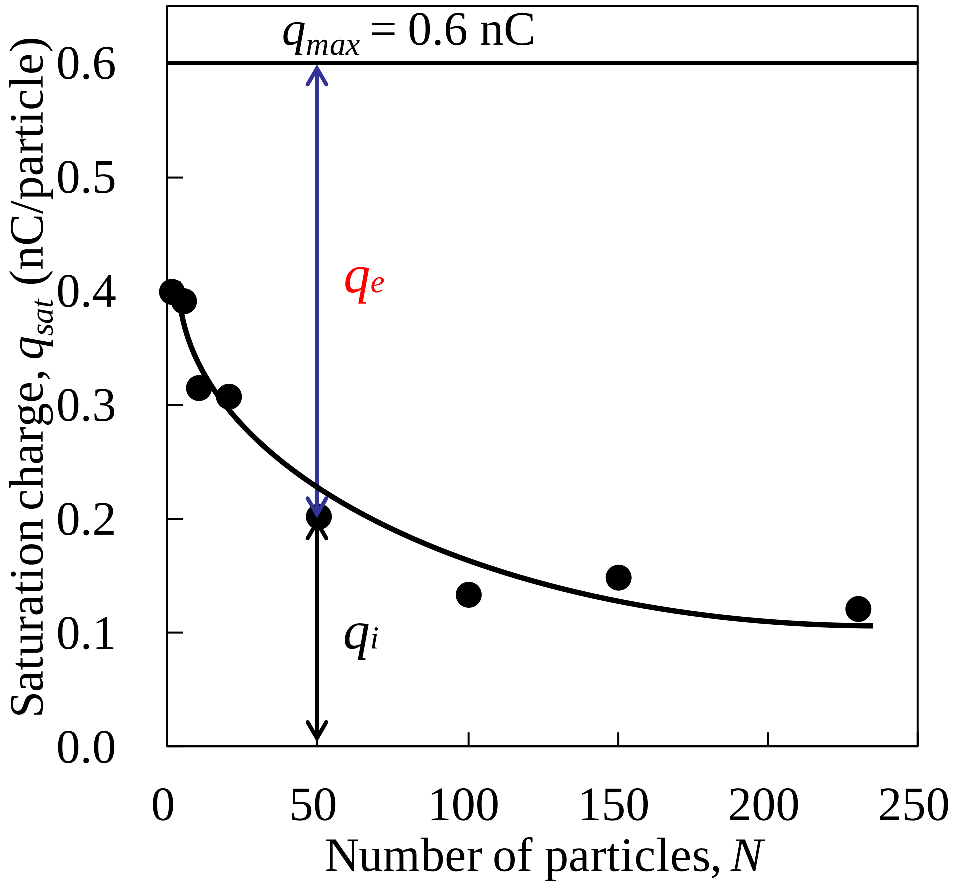
$$\Delta q = \alpha \Delta S\left(1 - \frac{q_i}{q_{max}}\right)$$

$$q_e = \pi d^2 \varepsilon_0 E$$

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

Δq (C):	Transferring charge
q_i (C):	Charge on particle
ΔS (m^2):	Maximum contact area
q_{max} (C):	Maximum charge
a (C/m^2):	Coefficient ($q_{max}/\pi d^2$)
q_e (C):	Equivalent charge
E (V/m):	Electric field at contact point

2. Model of space charge effect



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

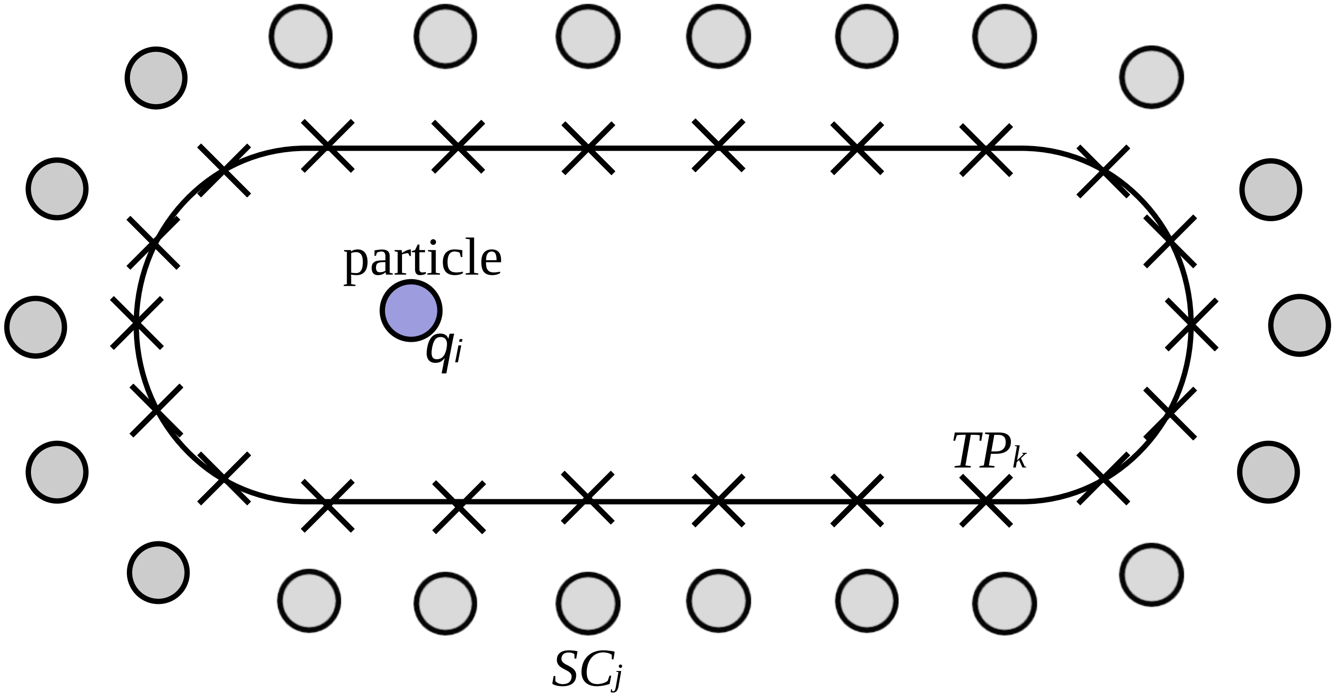
When $q_i + q_e = q_{max}$ transferring charge, Δq goes to equilibrium state

3. Boundary conditions (Simulation Charge Method)

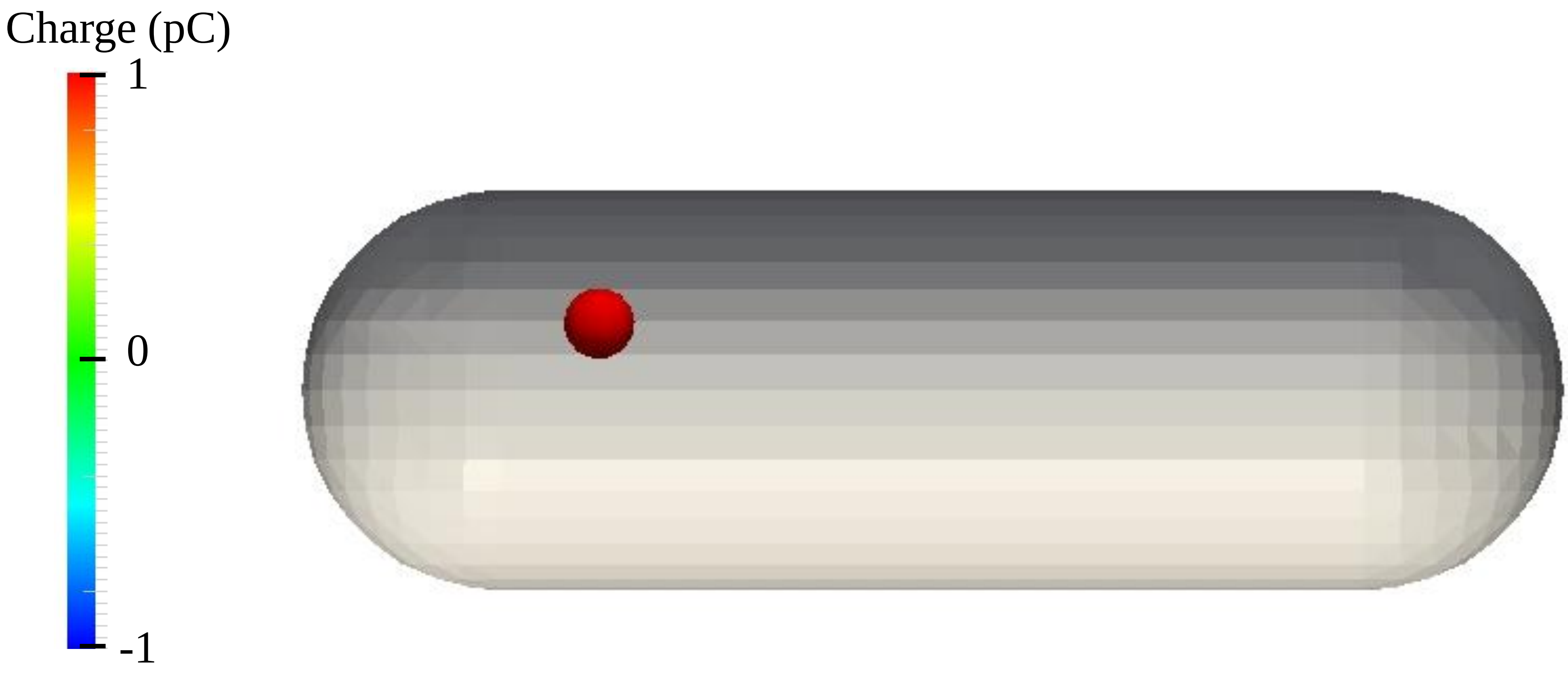
Boundary conditions

$$V_{Boundary} = 0$$

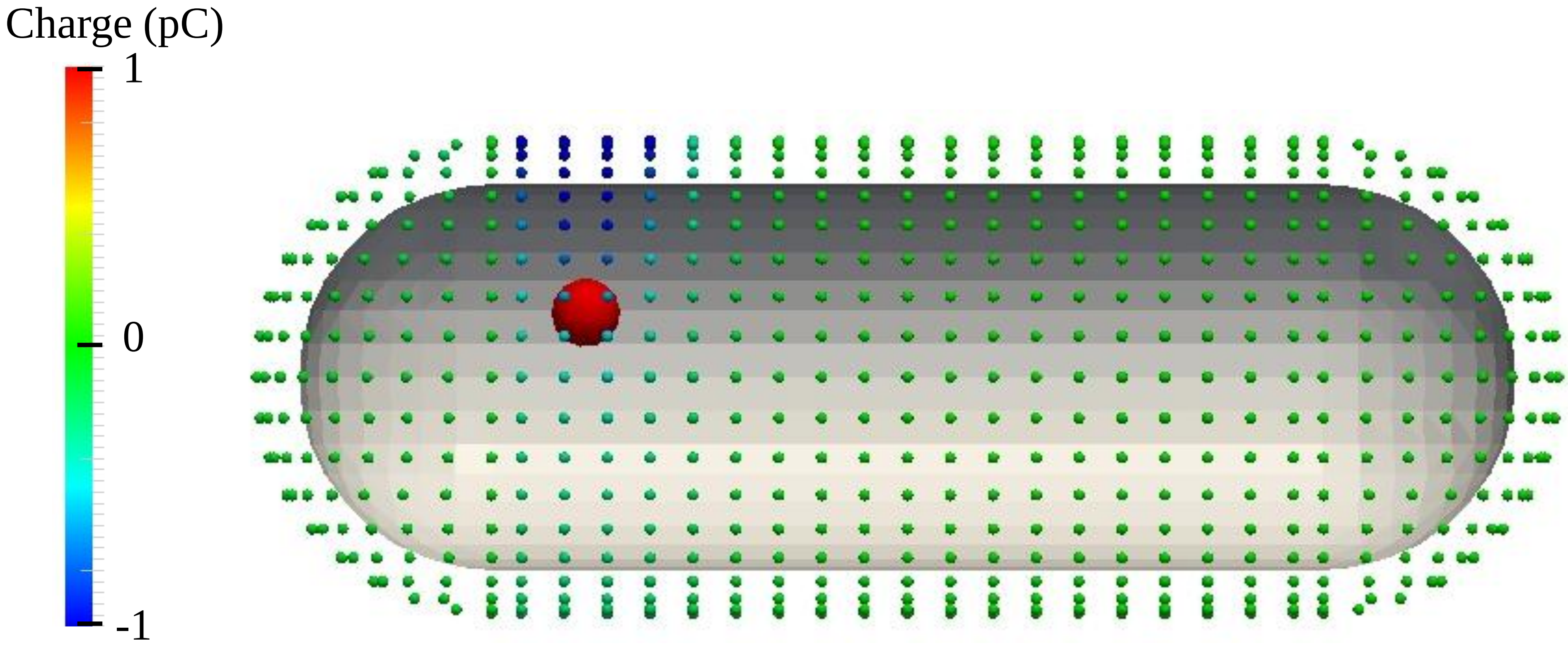
$$V_{TP_k} = \sum \frac{SC_j}{4\pi\varepsilon_0 r_{SC_j TP_k}} + \sum \frac{q_i}{4\pi\varepsilon_0 r_{q_i TP_k}}$$



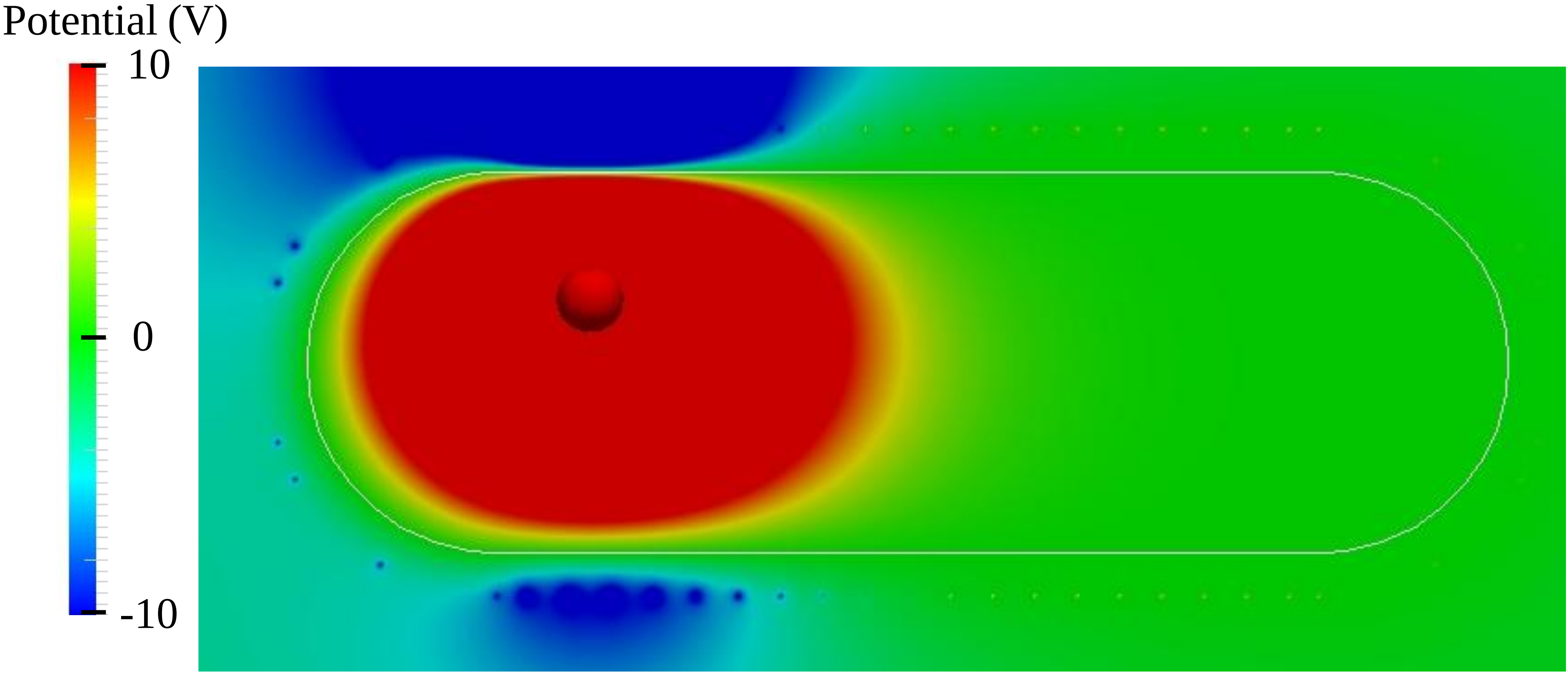
3. Simulation charge method



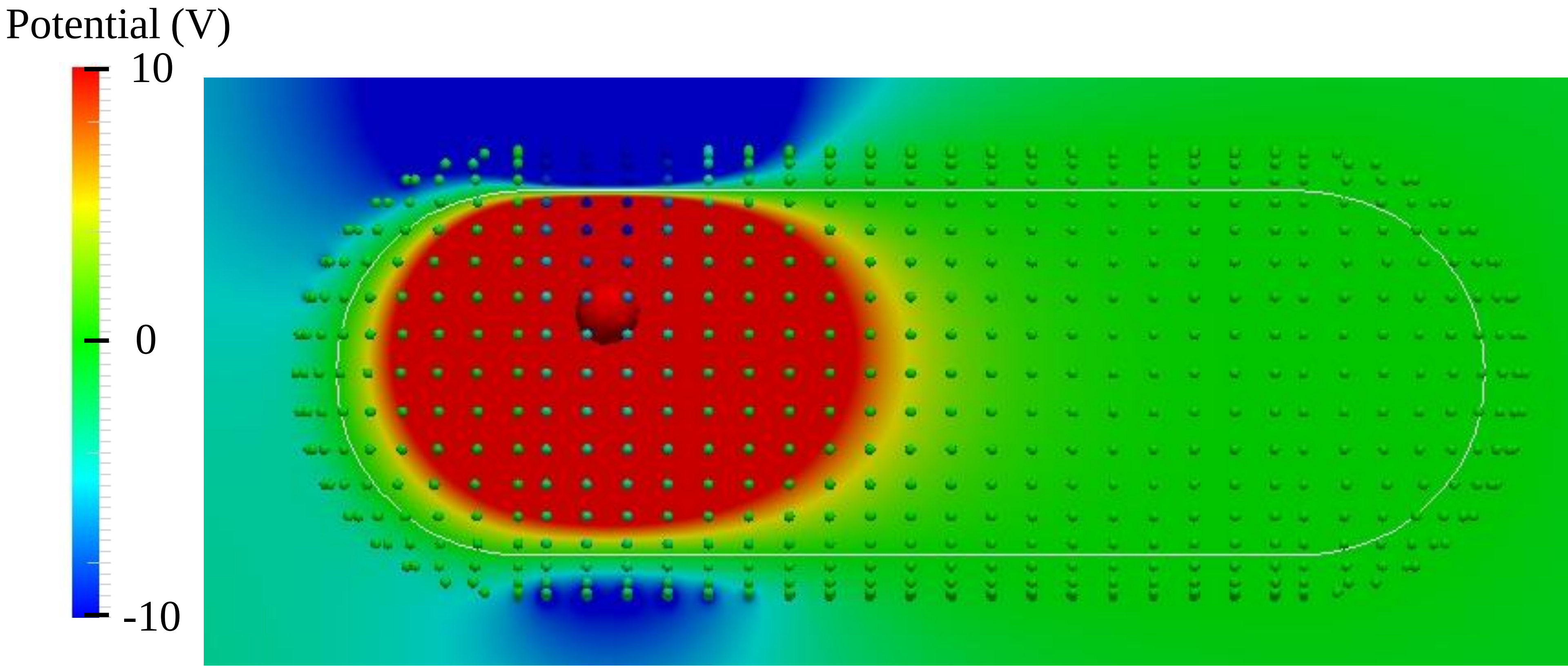
3. Simulation Charge Method



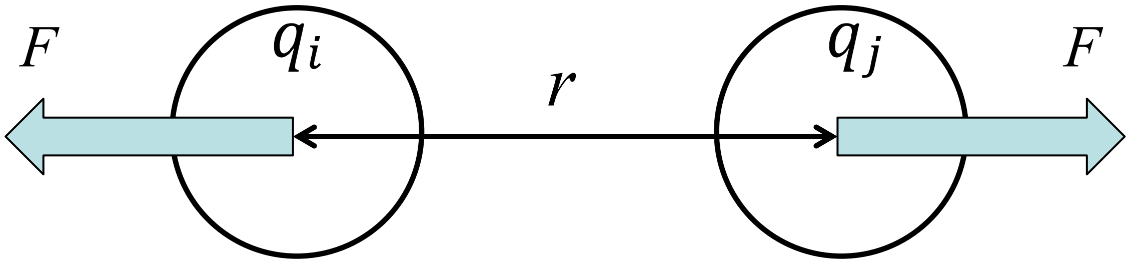
3. Simulation Charge Method



3. Simulation charge method

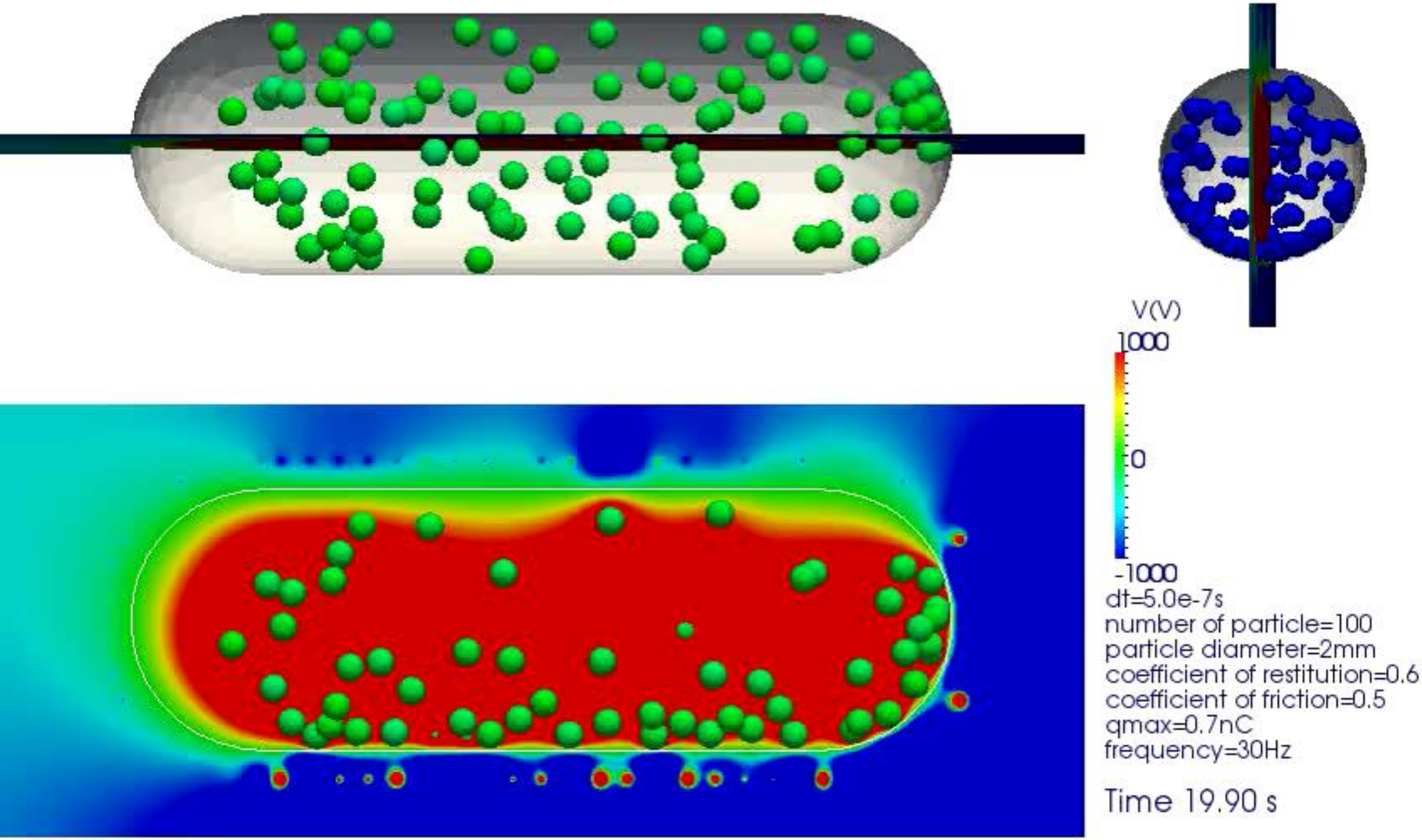


4. Electrostatic force

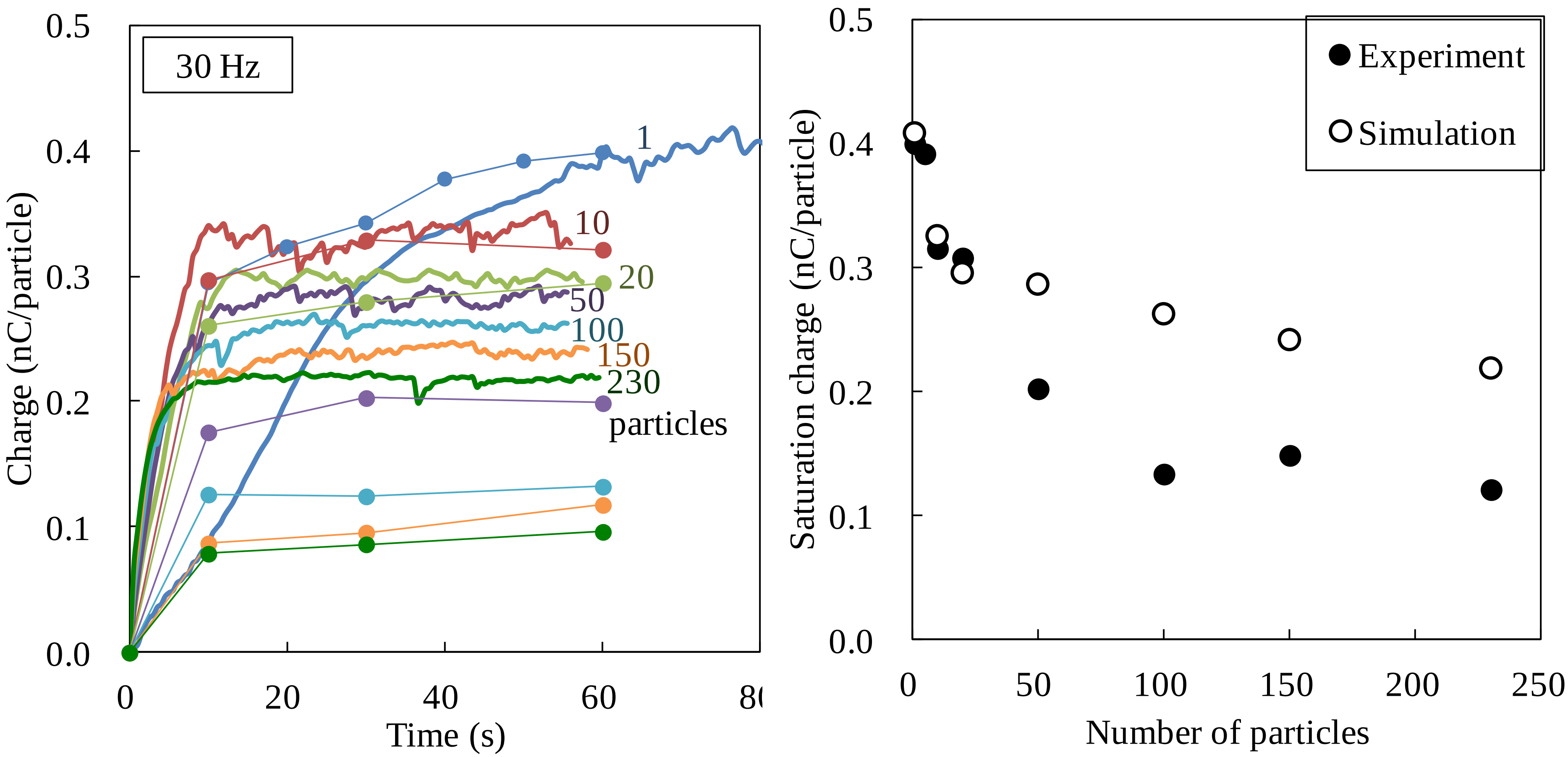


Coulomb force $F = \dfrac{q_i q_j}{4\pi r^2 \varepsilon_0}$

Simulation


V(V)
1000
0
-1000
dt=5.0e-7s
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

Simulation



Imba *et al.* (2013). Powders and Grains Conference, 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:

- Matsusaka, S., Ghadiri, M., Masuda, H. (2000), "Electrification of an elastic sphere by repeated impacts on a metal plate", Journal of Physics D: Applied Physics, 33, pp. 2311-2319.

- Watanabe, H., Ghadiri, M., Matsuyama, T., Ding, Y., Pitt, K.G., Maruyama, H., Matsusaka, S., and Masuda, H. (2007), Triboelectrification of Pharmaceutical Powders by Particle Impact, Int. J. Pharm., 334, 149-155

- Šupuk, E., Seiler, C., and Ghadiri, M. (2009), Analysis of a Simple Test Device for Tribo-Electric Charging of Bulk Powders, Part. Part. Syst. Char., 26, 7-16.

- Matsusaka, S., Fukuda, H., Sakura, Y., Masuda, H. and Ghadiri, M., 2008, "Analysis of pulsating electric signals generated in gas-solids pipe flow", Chemical Engineering Science, 63(5), 1353-1360.

- Matsuyama, T., Šupuk, E., Ahmadian, H., Hassanpour, A., and Ghadiri, M. (2009), Analysis of Tribo-Electric Charging of Spherical Beads Using Distinct Element Method, Powders and Grains 2009, Colorado, USA.

- Matsusaka, S., Maruyama, T., Matsuyama, T. and Ghadiri, M., 2010, "Triboeletric charging of powders: A review", Chemical Engineering Science, http://dx.doi.org/10.1016/j.ces.2010.07.005.

- Matsuyama, T., Supuk, E., Ahmadian, H., Hassanpour, A., Matsusaka, S. and Ghadiri, M. 2010, "Analysis of tribo-electric charging of spherical beads using distinct element method", World Congress of Particle Technology 6, Nuremberg.

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  - Hideo Watanabe
  - Enes Supuk