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A macroscopic donor–acceptor-based discrete element model for contact electrification of insulating granular materials

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\textbf{GRAPHICAL ABSTRACT}

\begin{itemize}
  \item Developed a macroscopic model for contact electrification in granular materials.
  \item Numerical outcomes quantitatively align with experimental results.
  \item Consistent reproduction of the size-dependent bipolar charging feature.
  \item Stochastic discharging, low breakdown strength, and boundary conditions improve model-experiment fit.
\end{itemize}

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\textbf{ABSTRACT}

Accurate modeling of contact electrification (CE) in insulating materials are essential for predicting and controlling their behavior. Here we propose a mesoscopic model based on the donor-acceptor mechanism, incorporating findings on surface patch features of identical dielectric materials from CE experiments. This model enables us to reproduce the charge transfer process and develop a macroscopic model that statistically captures this process. We implemented the macroscopic model in the discrete element code

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LIGGGHTS and validated its performance through comparisons with granular-vibration CE experiments. The results demonstrate the reliable performance of this model, as evidenced by the close agreement in overall particle charge distributions with experimental data. Furthermore, the model consistently reproduces the size-dependent bipolar charging feature. Our numerical tests also highlight the importance of stochastic discharging after breakdown and the influence of boundary conditions, which can reduce the mean charges and increase the variances, providing a better fit with experimental observations.

1. Introduction

Insulating granular materials are ubiquitous in nature and play a critical role in a broad range of industrial applications [1]. Images returned by past space missions reveal that granular materials are present in various size ranges and forms on many planetary bodies [e.g., the Moon, 2]. Understanding the physics of granular materials is essential for predicting and controlling their behavior, developing strategies to mitigate their potential hazards to humans and/or machinery, and designing and operating human/robotic missions that interact with the surface of planetary bodies.

The distinctive behavior and properties exhibited by granular materials can be attributed to the way that force and torque are transmitted between individual grains [3]. In addition to the short-range force caused by contacts, various long-range interactions also crucially influence granular materials’ behavior [4]. One important type of long-range interaction is the electrostatic interaction, which includes both the interaction between charged particles and the interaction between charged particles and external electric fields. For insulating materials that do not dissipate charge easily, the resulting electrostatic effect can cause particle clustering, blockages, and dust explosions [5].

Numerous experiments have shown that when the surfaces of identical insulators come into contact, static charges can be built up due to charge transfer such that one surface charges positively and the other surface charges negatively [e.g., 6–8]. This phenomenon belongs to what we commonly refer to as “tribocharging” or more generally as “contact electrification” (CE; since it may not always involve frictional movement) and has been utilized as an electrostatic control technique for grains in many industrial applications, such as electrophotography [9] and granular separation [10]. The electrostatic interactions due to CE are also expected to play a significant role in dust transport on planetary surfaces [11] and dust accretion in the evolution of the early solar system [12]. On the other hand, CE can cause hazardous charging effects and lead to quality control, maintenance and safety problems for equipment and humans under certain circumstances [13].

It is therefore essential to properly understand and model the CE process of insulating granular materials in order to accurately predict and readily control their behavior. Although the recognition of CE in identical insulating materials has a long history, the underlying physical and chemical mechanisms responsible for this phenomenon remain elusive [13–16]. This is mainly attributed to the following three aspects:

- the charge carrier species, such as electrons, ions, and nanopieces of non-neutral materials, being transferred during insulator contacts is under debate;
- the charge separation mechanisms between identical insulators are not well understood, and charging and discharging behaviors act over various timescales during the whole approach-contact-separation process;
- the effects of material surface properties and environmental conditions on charge transfer direction and magnitude add more complications.

Despite the complexities involved, previous laboratory experiments have consistently reported that the agitation of insulating bidisperse/polydisperse particle mixtures of the same material can cause distinct size-dependent bipolar charging. Specifically, many experiments found that the larger particles in the mixture tended to acquire a positive charge, while the smaller particles acquired a negative charge [e.g., 17–20], but a few other studies reported the opposite polarity [e.g., 21–24]. The direction and magnitude of charge separation and its relation with particle size differences have been utilized as important constraints to investigate the mechanisms behind the CE behavior of insulating granular materials.

One hypothesis to account for particle-size-dependent charging is that the work function, i.e., the energy required to remove one electron, is size-dependent. Gallo & Lama theoretically showed that the work function is larger for smaller particles of the same material, meaning that electrons would tend to transfer from larger particles to smaller ones [25]. However, the work function model assumed in their study might not be applicable to insulators [26]. In fact, the understanding of work functions relevant for describing insulators is still limited, and relying solely on the work-function model poorly explains insulator tribocharging phenomena [14,27]. Furthermore, even for metallic particles, the difference in work function for particles larger than 1 μm is negligible compared with the overall work function magnitude [13,28], while most of the CE experiments of insulating granular materials used particles with diameters of ~100 μm.

Another possible explanation for the bipolar charging phenomenon is that microscopic differences exist in the surface structure of particles with different sizes. The resulting variations in roughness, shape, and chemical composition can influence the contact process and affect the charge transfer tendency [26]. However, it is worth noting that these mechanisms are likely to be specific to individual experiments and cannot entirely account for the widespread and consistent behavior observed across a broad range of granular materials [13].

A more universal possible explanation for the widely observed size-dependent charging behavior is the asymmetric geometry of small and large particles. Lowell & Truscott proposed that electrons can be trapped in high-energy states in insulators and contact may cause these electrons on one surface to relax into a vacant low-energy state on another surface, and a net transfer of charge can occur during asymmetric rubbing between identical insulators [6,31]. Based on this mechanism, Lacks and co-workers showed that, in a randomly collisional granular system, the surface density of trapped high-energy electrons of the smaller particles decreases faster than that of the larger particles [32–34]. The trapped electrons have a tendency to move from the larger particles to the smaller ones, which results in the latter acquiring a negative charge.

However, Waitukaitis et al. reported that the surface density of trapped electrons, measured by thermoluminescence techniques, is orders of magnitude too small to account for the scale of charge transferred, and indicated other species, such as ions adsorbed on the surface or recruited from the surrounding gas, might be more likely to act as the charge carrier species [20]. If the charge carriers are positive ions, the small particles would be charged positively based on the asymmetric contact mechanism, which might account for the opposite charge polarity observed in some experiments [e.g., 21–24].

1 While it is unlikely that the size dependency of these microscopic features is responsible for bipolar charging, the resultant variations on the contact surfaces at the microscopic scale could indeed influence the tendency for surface “donor”/“acceptor” behavior and subsequently affect the charge transfer process [8,29,30], as elaborated in Section 2.
Furthermore, several laboratory experiments showed that charge transfer can also occur when surfaces of identical materials are contacted symmetrically [e.g., 7,8,29]. Using two atomically flat pieces of identical insulators, Apodaca et al. observed continuous charge separation of the two pieces upon repeated contacts [8]. Their experimental results indicate that the magnitude of charge transfer is directly proportional to the square root of the contact area, rather than to the contact area alone. This charging behavior suggested that same-material insulator CE may arise from equally-sized, randomly-distributed donor/acceptor sites, where unit charges are transferred during contact from a donor site to an acceptor site.

With the goal of improving the modeling capability and overall understanding of CE in insulating granular materials, this study aims to, (1) develop a charge transfer model that can systematically connect the donor–acceptor mechanism and the size-dependent bipolar charging phenomenon; (2) incorporate this model in a form that can be easily implemented in a discrete element modeling (DEM) framework; (3) quantify model parameters and influence factors by validation and calibration against experimental data.

The rest of the paper is organized as follows. In Section 2, we will use a mesoscopic CE model based on the donor–acceptor mechanism to investigate the charging behavior of two spherical particles. A macroscopic CE model will be derived and tested against the mesoscopic model. In Section 3, we will introduce the implementation of the macroscopic model in a DEM framework. Section 4 will test and calibrate the DEM numerical model against vibrated granular bed experiments. Section 5 will discuss the features and limitations of the proposed CE model in comparisons with other CE models.

2. Contact electrification: a donor–acceptor model

As discussed in the introduction, contact electrification between identical insulator materials is likely to be driven by the inherent, molecular-scale fluctuations of the material [8,29]. On microscopic scales, some surface locations on one surface tend to donate charge carriers, while other locations tend to absorb these carriers. The surface of an insulator grain can be described as having a scattered pattern of donor and acceptor elementary sites that are distributed throughout in some manner. Based on this donor–acceptor concept, Yu et al. [35] carried out statistical modeling of charge transfer for a bidisperse particle system and showed that the size-dependent charge separation behavior can be produced as a result of the asymmetric donor/acceptor numbers between the small and large particles.

In this session, based on an updated view of the role of mesoscale donor/acceptor patches [30] and the detailed consideration of electrostatic interactions, we investigate how such donor–acceptor pattern can contribute to the bipolar charging of two particles with different sizes. We first consider a mesoscopic model to investigate the charge transfer process at the mesoscale patch level, based on which we attempt to develop a macroscopic model to describe the process in terms of bulk properties such as the averaged donor/acceptor density.

2.1. Mesoscopic modeling of contact electrification between two particles

By using Kelvin force probe microscopy to inspect the surface potential of contact-electrified pieces, Baytekin et al. found that, after contact, each surface presents a random mosaic of oppositely charged regions of spatial scales up to ~450 nm [29]. Based on statistical and numerical analyses, Grosjean et al. suggested the ~450-nm mosaic size represents the length scale of a donor/acceptor patch that formed through a physically-derived nucleation process of elementary donor/acceptor sites [30]. They proposed a scale-spanning model based on the rationale that there are three length scales at play for insulator CE, i.e., the contact surface macroscopic scale $L$, the donor/acceptor patch mesoscopic scale $l$, and the donor/acceptor elementary site microscopic scale $l_0$. They showed that the exchanged charges as a function of the contact number found in the experiments of Apodaca et al. [8] can be readily produced if the elementary donor/acceptor site has a length scale $l_0$ of ~0.4 nm, provided $L \gg l_0$.

According to this scale-spanning model, we construct an idealized spherical insulating particle for CE purpose by dividing the surface into subsets of patches with the mesoscopic length scale of $l$. Each patch will be randomly assigned as either a donor or an acceptor site based on a given probability of being a donor, $p_{d,0}$ (by default, the acceptor probability $p_{a,0} = 1 - p_{d,0}$, unless otherwise specified). Fig. 1 shows an example of two particles produced via this procedure. Each donor/acceptor patch has a surface area of $r^2$.

If the particles $i$ and $j$ are colliding with each other with an approaching speed of $v_{ij}$, the contact surface can be approximately characterized by the Hertzian contact theory of deformable elastic spheres [36], i.e.,

$$A_c = \pi \frac{5kR^2\rho V^2(1 - v^2)}{2E} \frac{1}{2/5},$$

where $\rho$ is the mass density, $E$ is the Young’s modulus, and $v$ is the Poisson’s ratio of the insulating material. The effective radius $R = R_i R_j / (R_i + R_j)$, where $R_i$ and $R_j$ are the radii of particles $i$ and $j$, respectively.

This contact area defines the contact surface macroscopic scale, i.e., $L = \sqrt{A_c}$. The total number of patches on the contact surface is given as, $N_{patch} = (L/l_0)^2$. The number of elementary charge carriers on each patch is given as, $N_{elec} = (l/l_0)^2$.

We modify the procedure introduced in Apodaca et al. [8] and Grosjean et al. [30] to perform the charge transfer process between two spherical particles. The charge transfer probability $\alpha$ [8,30] is used to characterize the likelihood of charge transfer from a donor site to an acceptor site. Each donor site has one unit, $e$, charge (which can be either positive or negative). To account for the possibility of colliding at various locations during multiple contact events and the effect of repelling electric field and discharges associated with dielectric breakdown, we design the charge exchange procedure between two spheres as follows:

1. **Contact surface selection:** we randomly sample a surface area of $A_c$ on each particle. This ensures that the contact point is chosen in a way that is representative of the overall surface area and avoids bias towards certain regions.

2. **Transfer probability adjustment:** a constant $\alpha$ was assumed in [8,30] in the modeling of multiple contact events. Nonetheless, as the amount of charge increases and the repelling electric field becomes stronger, the energy needed to transfer additional charge in the same direction also increases [37–39]. This might decrease the likelihood of charge transfer during contact. To account for this effect, we adjust the transfer probability as

$$\alpha_{i \rightarrow j} = \alpha \left[ 1 - \text{sgn}(e) \frac{E_j}{E_i} \right], \quad \alpha_{j \rightarrow i} = \alpha \left[ 1 + \text{sgn}(e) \frac{E_j}{E_i} \right],$$

Fig. 1. Surface pattern examples of two particles (with diameters of 150 μm and 250 μm, respectively) with donor (dark) and acceptor (light gray) patches (with a size scale of 450 nm according to [29,30]). The donor probability $p_{d,0}$ is set to 0.3 for this case. The bright spot on each particle is the reflection of a light source in the front.
Contact electrification outcomes between two particles as derived from the mesoscopic model. The two particles have radii of \( r_i, r_j \) (as indicated in the legend, with different colors). The figure text provides the values of the remaining parameters.

4. Discharging due to dielectric breakdown: As contact between two particles continues, the charge density on each particle’s surface can increase, resulting in a higher voltage with respect to another surface during contact. If this voltage reaches the threshold value for air breakdown, the surface charge may dissipate through the air and transfer away from the surface \[ \text{(40)} \]. Additionally, dielectric breakdown of an insulating surface can induce charges to flow along its surface and onto the other particle in contact. Numerous contact charging experiments have provided direct evidence of discharges associated with dielectric breakdown \[ \text{e.g., 41–44} \]. To account for this, we use the dielectric breakdown strength, denoted by \( E_{bd} \), to constrain the upper limit on the net charge that can develop from CE. When the electric field strength at the contact point surpasses this given threshold, the charges on each particle are adjusted in order to keep the electric field strength at this threshold,

\[
\Delta Q_i = -\Delta Q_j = \begin{cases} 
4\pi\varepsilon_0 R_i^2 R_j & \text{if } E_{ij} > E_{bd}, \\
4\pi\varepsilon_0 R_i^2 R_j & \text{if } E_{ij} < -E_{bd}.
\end{cases}
\]

In Table 1, we summarize the model parameters for the mesoscopic particle CE model. The provided value ranges are based on the typical physical parameters of insulating granular materials and the donor–acceptor model \[ \text{[8,30]} \].

We employ this model to investigate the charge exchange process between two particles. The interaction and charge exchange of the two particles is determined by the four-step procedure introduced above. The initial charge of the particle is assumed to be \( Q_i \). Fig. 2 illustrates the total charge accumulated on the small particle (i.e., particle \( i \)) during sequential contacts with different model parameters (the total charge of particle \( j \) has the same amount but opposite polarity). Each panel presents the effect of each model parameter (as indicated in the legend, with different colors). The figure text provides the values of the remaining parameters.

where \( E_r \) is an empirical parameter that describes the degree of influence exerted by the repelling electric field. The electric field at the contact point can be estimated using superposition as \( E_{ij} = (Q_i/R_i^2) - (Q_j/R_j^2)/(4\pi\varepsilon_0) \). The vacuum permittivity \( \varepsilon_0 = 8.854187 \text{ pC/(V·m)} \).

3. Charge transfer: at the \( N_c \)-th contact, comparing the two selected surface contact regions, charge transfer occurs if a donor patch on particle \( i \) faces an acceptor patch on particle \( j \), and vice versa. Considering a patch pair \( m \), assuming the available donor and acceptor elements on the corresponding patches are \( N_{d,i}^{(m)} \) and \( N_{a,i}^{(m)} \), respectively, the total number of charge carriers transferred from particle \( i \) to particle \( j \) on these two patches is calculated as \( \Delta N_{a,i}^{(m)} = \sum_{a=1}^{N_{a,i}^{(m)}} X_a < a_{ij}^{(m)} (N_{a,i}^{(m)} + N_{a,j}^{(m)}) \), where \( X_a \in [0,1] \) is an independent random uniform variable. Correspondingly, the elementary donor/acceptor numerator of the involved patches decreases by \( \Delta N_{d,i}^{(m)} \). No charge transfer occurs if both patches have the same type. This calculation is repeated \( N_{patch} \) times to complete the transfer for all the patches at the contact area. In total, the net charges on particle \( i \) and \( j \) are changed by \( \Delta Q_i = \sum_{m=1}^{N_{patch}} e(\Delta N_{a,i}^{(m)} - \Delta N_{d,i}^{(m)}) \) and \( \Delta Q_j = \sum_{m=1}^{N_{patch}} e(\Delta N_{a,j}^{(m)} - \Delta N_{d,j}^{(m)}) \), respectively.

Table 1

<table>
<thead>
<tr>
<th>Description</th>
<th>Symbol</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Young’s modulus</td>
<td>( E )</td>
<td>10</td>
<td>GPa</td>
</tr>
<tr>
<td>Repelling effect strength</td>
<td>( E_{bd} )</td>
<td>1–10</td>
<td>MV/m</td>
</tr>
<tr>
<td>Breakdown strength</td>
<td>( E_{bd} )</td>
<td>1–4</td>
<td>MV/m</td>
</tr>
<tr>
<td>Macroscopic scale</td>
<td>( l )</td>
<td>5–6500</td>
<td>nm</td>
</tr>
<tr>
<td>Microscopic scale</td>
<td>( l )</td>
<td>45–6500</td>
<td>nm</td>
</tr>
<tr>
<td>Initial donor probability</td>
<td>( p_{d,i} )</td>
<td>0.0–0.9</td>
<td>–</td>
</tr>
<tr>
<td>Collision speed</td>
<td>( V_{col} )</td>
<td>1</td>
<td>m/s</td>
</tr>
<tr>
<td>Transfer probability</td>
<td>( \alpha )</td>
<td>0.05–0.3</td>
<td>–</td>
</tr>
<tr>
<td>Poisson’s ratio</td>
<td>( \nu )</td>
<td>0.22</td>
<td>–</td>
</tr>
<tr>
<td>Material mass density</td>
<td>( \rho )</td>
<td>3800</td>
<td>kg/m³</td>
</tr>
</tbody>
</table>
repelling electric field at the contact point has a diminished impact on the transfer probability. As a result, the charge exchange retardation is less efficient.

- **Breakdown strength** $E_{bd}$: this parameter controls the maximum amount of charge that can accumulate on the contacting particles and does not impact the charge exchange process until the point of saturation is reached. The final charge $|Q_{final}^{tinal}| = 4\varepsilon_\alpha E_{bd} R_i^2 l_i (R_i^2 + R_j^2)$.

- **Initial donor probability** $p_{d0}$: this parameter affects both the charge exchange efficiency and polarity by altering the amount of donor and acceptor sites on particles’ surfaces. When $p_{d0} > 0.5$, the smaller particle tends to lose charge carriers to the larger one, while the opposite occurs when $p_{d0} < 0.5$. When $p_{d0} = 0.5$, there is no size-dependent polarity as the small and large particles have the same probability to lose/obtain charge carriers in each collision.

- **Mesoscopic scale $l$**: this parameter affects the magnitude of stochastic oscillations in the charge exchange process before saturation. When $L/l < 1$, which corresponds to a scenario where the contact area is either a donor or an acceptor patch, the flow direction of the charge carriers can change with each contact, resulting in the oscillating behavior (e.g., the blue curve). When $L/l$ increases, the contact area contains more donor/acceptor patches and tends to have a donor probability close to $p_{d0}$. This leads to a more steady increase in $Q$, as there is less fluctuation in the flow direction of charge carriers.

- **Microscopic scale $l$**: this parameter has a complicated effect on the contact charging process. When $l_0 = 225$ nm, the surface number density of charge carriers is low, and the final magnitude of charge on particle $i$ is restricted by the total number of available donor and acceptor sites, i.e., $|Q_{final}^{tinal}| = 4\varepsilon_\alpha (1 - 2p_{d0})(R_i/R_0)^2$. When $l_0$ decreases to the point that the saturation state is limited by the electric breakdown strength, the final charge $Q_{final}$ becomes entirely dependent on $E_{bd}$. When $l_0$ further decreases to the extent that the total number of donor/acceptor elementary sites on a single patch, i.e., $(l/l_0)^2$, is comparable to the breakdown limit, $|Q_{final}^{tinal}|/\varepsilon_\alpha$, the evolution of the accumulated charges exhibit heavy oscillations. This is because a single contact can lead to the steady charge exchange process becomes more efficient, and a higher final charge saturation state is achieved.

Based on this understanding, we construct a macroscopic model to describe the contact charge separation process. At the beginning, the averaged surface densities of the donor and acceptor sites on particles $i$ and $j$ are given as

$$\sigma_{d} = \sigma_{a} = \frac{\rho_{d0}^2}{l_0^2} = \frac{\sigma_{d0}}{l_0}, \sigma_{a} = \frac{1 - \rho_{d0}}{l_0}.$$  

The total elementary site density $\sigma_{tot} = \sigma_{tot}^i = 1/l_0^2$.

During each collision (the variables before collision are denoted as $\cdot_0$), charge can be exchanged between particles $i$ and $j$ according to the following rule,

$$\Delta Q_{i \rightarrow j}^{(macro)} = \sigma_{d0}^i \sigma_{a0}^j \sigma_{d0} \sigma_{a0}^{-1},$$  

$$\Delta Q_{j \rightarrow i}^{(macro)} = \sigma_{d0}^j \sigma_{a0}^i \sigma_{d0} \sigma_{a0}^{-1},$$

where $\sigma_{d0}^i$ and $\sigma_{a0}^j$ are calculated from Eq. (2). If the electric field strength $E_0$ at the contact point exceeds the breakdown strength $E_{bd}$, the total charge is adjusted using Eq. (3).

After collision (the variables are denoted as $\cdot$), the donor and acceptor densities for each particle are updated accordingly, i.e.,

$$\sigma_{d}^i = \sigma_{d0}^i - \Delta Q_{i \rightarrow j}^{(macro)} / 4\varepsilon_\alpha R_i^2,$$

$$\sigma_{a}^i = \sigma_{a0}^i - \Delta Q_{j \rightarrow i}^{(macro)} / 4\varepsilon_\alpha R_i^2.$$  

We conducted contact electrification tests between two particles with varying size ratios, using both the macroscopic and mesoscopic models. The following set of parameters were used in the tests: $E_0 = 5$ MV/m, $E_{bd} = 1$ MV/m, $l_0 = 4$ nm, $p_{d0} = 0.3$, $a = 0.2$, and $R_i = 75$ μm. Particle $j$’s radius is given by the product of $R_i$ and the size ratio for each test.

Fig. 3 compares the charge separation process obtained by the two models. It can be observed that, as the size ratio increases, the charge exchange process becomes more efficient, and a higher final charge amount is acquired. This finding is consistent with previous experimental and numerical results [26,45,46]. The dependence of charging behavior on the size ratio can be attributed to several factors. One important factor is that the donor/acceptor density of the larger particle decreases more slowly with a larger size ratio during subsequent collisions, as indicated by Eq. (6). Additionally, the contact area increases with the size ratio, which can enhance the amount of charge transferred in each collision.

Notably, the results of the macroscopic model are in good agreement with the mean value obtained from the mesoscopic model. The observed consistency instills confidence in applying the macroscopic model to simulate CE in identical insulating granular materials. Furthermore, the simple format of the macroscopic model can be easily quantitatively explained.

2 Although $l_0$ poses an available donor/acceptor density limit on the final charge, previous studies have shown that the density is not likely to be a limiting factor [38,39].

3 Note that the charge polarity and magnitude are evaluated with respect to $e$, which also influences the absolute charge and polarity.
incorporated into a discrete element modeling framework, facilitating its practical application in large-scale simulations of granular materials.

3. Discrete element model with contact electrification

The discrete element method (DEM), first introduced by Cundall and Strack in the 1970s [47], is a widely used and well-established technique for simulating granular materials [48]. Early versions of the DEM only considered contact forces along the normal and tangential directions. In recent two decades, the method has been expanded to include more sophisticated contact physics to describe the interactions between modeled particles, such as rolling friction [e.g., 49], cohesion [e.g., 50], and particle breakage [e.g., 51].

Electrostatic interactions between charged particles that arise from CE have also been taken into account in the DEM framework by numerous researchers. The existing CE models can be categorized into three types:

- **Prescribing the charge transfer as a function of time.** For example, Hogue et al. [52] used an exponential equation to describe the charge transfer to a glass sphere that rolls and bounces down an inclined plane in their DEM simulations. The equation was given as $Q = Q_{sat}(1 - \exp(-t/r))$, where $Q_{sat}$ is the saturation charge determined by $E_{eq}$ and $r$ is a time constant. Matsuyama et al. [53] employed a similar format to model charge transfer in a vibrating cylindrical container. They calculated the transferred charge for each collision as $\Delta Q = A_i x_i (\phi_i - \phi_j - E_j / \delta e)$, where $\phi_i$ and $\phi_j$ are the surface potentials of the two particles and $\delta$ is the cutoff distance of the charge transfer [55]. Laurentie et al. [57] proposed to implement this model in the DEM by using the contact area increment $\Delta A$, at each timestep to calculate $\Delta Q$. They validated the model by comparing it with vibration experiments using polymer particles. After adjusting the work function of the polymers, the numerical results were observed to closely match the experimental data. Thanks to its simplicity and versatility, similar forms of this charge transfer model have been adopted by many studies in various DEM codes to investigate CE of insulating granular materials [e.g., 58–60]. Liu et al. [61] utilized this model to simulate the contact charging process of bidisperse soda-lime glass particles in vibrated beds. By assigning different effective work function values to the large and small particles, respectively, their DEM simulations were able to capture the bipolar charging behavior. However, as the difference in the work function due to particle size for an identical material is non-relevant for the considered scenario (as discussed in Section 1), this model does not explain the underlying mechanism behind the observed size-dependent bipolar charging behavior.

- **Driving the charge transfer through surface potential difference.** This concept originates from metal–metal contacts, where electrons flow from the metal with a lower work function to the other one with a higher work function [54]. This idea was extended to explain charge transfer for insulator–insulator contacts by introducing an effective work function that characterizes the surface state of an insulating material [55,56]. In the high density limit of this surface state theory, the charge exchanged between particles $i$ and $j$ can be calculated as, $\Delta Q = A_j x_j (\phi_i - \phi_j - E_j / \delta e)$, where $\phi_i$ and $\phi_j$ are the surface potentials of the two particles and $\delta$ is the cutoff distance of the charge transfer [55]. Our model belongs to the third category, which are more suitable for modeling CE between identical insulators. We implemented this model in the open-source DEM software LIGGGHTS. In the following, we provide a brief overview of the DEM model and introduce the charge exchange model.

### 3.1. Dynamical equations

In the DEM framework, the motion of a particle $i$ can be derived by solving the Newton’s equations of motion, i.e.,

$$m_{ci} \frac{d^2 \mathbf{v}_i}{dt^2} = \sum_{j=1}^{N(i)} \mathbf{F}^{(c)}_{ij} + \mathbf{F}^{(e)}_i + m_{ci} \mathbf{g}, \quad (7)$$

$$I_i \frac{d\omega_i}{dt} = \sum_{j=1}^{N(i)} \mathbf{M}^{(c)}_{ij},$$

where $m_i$ and $I_i$ are the mass and moment of inertia, and $\mathbf{v}_i$ and $\omega_i$ are the translational and rotational velocity of particle $i$, respectively. $N(i)$

![Fig. 3. Contact electrification outcomes: the macroscopic model v.s. the mesoscopic model. The results are displayed in various colors to represent different particle radii ratios. The thick curves indicate the outcomes of the macroscopic model, while the thin partially transparent curves show two simulation example outcomes of the mesoscopic model for each radii ratio case. In addition, the opaque thin curves depict the mean value obtained from 60 mesoscopic simulation outcomes, where the particle surface patch features were generated randomly in each simulation.](image-url)

$\text{Acquired charge } Q_i^c$ 

$\text{Number of collisions } N_{ci} \times 10^4$

- Driving the charge transfer through surface potential difference. This concept originates from metal–metal contacts, where electrons flow from the metal with a lower work function to the other one with a higher work function [54].
is the total number of particles that are in contact with particle $i$. $F^{(c)}$ and $M^{(c)}$ are the total force and torque due to the mechanical contact with particle $i$, and $F^{(e)}$ is the electrostatic force. $g$ is the gravitational acceleration.

### 3.2. Mechanical model

The particle contact force $F^{(c)}$ includes components along the normal and tangential directions, $F^{(c)}_{n,i}$ and $F^{(c)}_{t,i}$. We use the Hertzian contact model with a record of tangential overlap history to compute the contact force. The torque acting on particle $i$ due to the tangential force is calculated as $M^{(c)}_{t,i} = n_{i,c} \times F^{(c)}_{n,i}$, where $n_{i,c}$ is the vector connecting the center of particle $i$ with the contact point. In this study, we do not consider rolling friction, as it is of lesser importance in the highly agitated granular system under consideration. The contact mechanical model has been implemented in the original version of LIGGGHTS. We refer the reader to the user website\(^4\) for detailed description of the mathematical formulae of the normal and tangential contact forces (the formulae can also be found in [58]).

### 3.3. Electrostatic forces

In this study, we assume that the charge is homogeneously distributed on the surface of each particle, and the dielectrophoretic effects due to particle polarization are not taken into account. This simplification is a typical treatment used in previous DEM CE models [e.g., 57, 58, 61].

The electrostatic force $F^{(e)}$ acting on particle $i$ is given as

$$ F^{(e)}_{i} = \frac{Q_i}{r_i} E(x_i), \quad (8) $$

where $Q_i$ is the particle charge, and $E(x_i)$ is the strength of the electric field at particle $i$’s mass center $x_i$. The electric field $E(x_i)$ can be evaluated though Coulomb’s law using the principle of superposition, i.e.,

$$ E(x_i) = E_{ext} + E_{long}(x_i) + \frac{1}{4\pi \varepsilon_0} \sum_{\substack{j \neq i, j \notin \text{contact} \\text{pair} \\text{for} \ D_i \to d_{cut-off}}} \frac{Q_j n_{ij}}{||x_i - x_j||^2}, \quad (9) $$

where $n_{ij} = (x_i - x_j)/||x_i - x_j||$. $E_{ext}$ represents an electric field that originates from an external source, such as two electrodes ($E_{ext} = 0$ V/m in the current study). $E_{long}(x_i)$ represents the long-range component of the electric field that arises due to the influence of charged particles located beyond a certain cutoff distance, $d_{cut-off}$, from particle $i$ in the granular system. The final term in the equation accounts for the short-range component of the electric field due to charged particles located within $d_{cut-off}$.

The long-range component of the electric field, $E_{long}(x_i)$, can be calculated implicitly from the charge distribution by solving Poisson’s equation [e.g., 58] or using a hybrid form of particle–particle particle–mesh method [e.g., 63]. In this study we do not account for $E_{long}(x_i)$. This choice is justified in the context of our application, i.e., modeling of granular vibration experiments. As shown in Section 4.2, the maximum charge-to-mass ratio of a particle is on the order of $10^{-4}$ C/kg, and the electrostatic force between two particles in contact with each other is comparable to the gravitational force. By setting the cutoff distance $d_{cut-off}$ to five times the maximum particle radius, we find that the electrostatic force acting on a particle from another particle located outside of $d_{cut-off}$ reduces to about 1% of the gravitational force. Furthermore, because the particles frequently collide with the boundary and other particles during the shaking of the container, which significantly alters their trajectories, the long-range electrostatic force is not likely to have a substantial impact on the overall behavior of the particles for the considered scenarios.

\[4\] https://www.cfdem.com/media/DEM/docu/pair_gran.html

### Table 2

Macroscopic DEM CE model parameters (nominal test parameters are underlined).

<table>
<thead>
<tr>
<th>Description</th>
<th>Symbol</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Young’s modulus</td>
<td>$E'$</td>
<td>10</td>
<td>MPa</td>
</tr>
<tr>
<td>Repelling effect strength</td>
<td>$E'_f$</td>
<td>[5, 10, 15]</td>
<td>MV/m</td>
</tr>
<tr>
<td>Breakdown strength</td>
<td>$E'_d$</td>
<td>[1, 1.5, 2]</td>
<td>MV/m</td>
</tr>
<tr>
<td>Carrier charge</td>
<td>$\epsilon$</td>
<td>$1.6 \times 10^{-19}$</td>
<td>C</td>
</tr>
<tr>
<td>Effective surface area fraction</td>
<td>$S_A$</td>
<td>Wall. 0.1</td>
<td>–</td>
</tr>
<tr>
<td>Microscopic scale</td>
<td>$a$</td>
<td>[1, 4, 16]</td>
<td>nm</td>
</tr>
<tr>
<td>Mass ratio</td>
<td>$m_{co}$</td>
<td>[8, 4, 2, 1, 0.5]</td>
<td>–</td>
</tr>
<tr>
<td>Initial donor probability of mobile particles</td>
<td>$p'_{d0}$</td>
<td>[0.1, 0.3, 0.5]</td>
<td>–</td>
</tr>
<tr>
<td>Initial donor probability of wall particles</td>
<td>$p'_{d0}$</td>
<td>[0.0, 0.1, 0.3]</td>
<td>–</td>
</tr>
<tr>
<td>Initial acceptor prob. of wall particles</td>
<td>$p'_{a0}$</td>
<td>[0.0, 0.3, 0.7]</td>
<td>–</td>
</tr>
<tr>
<td>Transfer probability</td>
<td>$q'$</td>
<td>[0.1, 0.2, 0.3]</td>
<td>–</td>
</tr>
<tr>
<td>Poisson’s ratio</td>
<td>$\nu'$</td>
<td>0.22</td>
<td>–</td>
</tr>
<tr>
<td>Material mass density</td>
<td>$\rho'$</td>
<td>3800</td>
<td>kg/m$^3$</td>
</tr>
<tr>
<td>Restitution coefficient</td>
<td>$e_r$</td>
<td>0.9</td>
<td>–</td>
</tr>
<tr>
<td>Friction coefficient</td>
<td>$\mu_t$</td>
<td>0.5</td>
<td>–</td>
</tr>
<tr>
<td>Timestep</td>
<td>$\Delta t$</td>
<td>$5 \times 10^{-7}$</td>
<td>s</td>
</tr>
</tbody>
</table>

Although the dielectrophoretic effects and $E_{long}(x_i)$ are not accounted for in the current study with the consideration of computational efficiency and their relatively small influence on the particles’ trajectories, their influence may be important in less agitated scenarios, such as particle flow and deposition [52, 63]. Therefore, in future investigations, these effects should be considered in the DEM model when appropriate.

### 3.4. Charge exchange model

We use the macroscopic model introduced in Section 2.2 to characterize the charge exchange between two particles in contact. For clarity, we describe the 3D format of the charge exchange model here. The superscript $t$ is used to distinguish the macroscopic DEM CE model parameters (as summarized in Table 2) from those used in the macroscopic two-particle CE model. The charging flux of particle $i$ is determined by summing up the charges transferred from all other particles in contact with it, i.e.,

$$ Q_i = \sum_{j \neq i} N_{ij} = \sum_{j \neq i} \frac{\Delta Q_{ij}}{\Delta t}, \quad (10) $$

where $\Delta t$ is the DEM integration timestep.

The net charge transferred from particle $j$ to particle $i$ at one timestep is given as (the variables at time $t$ and $t + \Delta t$ are denoted as $^t$ and $^t+$ respectively),

$$ \Delta Q_{ij} = \Delta Q_{ij}^t - \Delta Q_{ij}^{t+} $$

$$ = H(\Delta A) \frac{\Delta A}{\sqrt{\rho^2}} \left( a_{ij}^{t+} s_{ij}^{t+} - a_{ij}^t s_{ij}^t \right) e. \quad (11) $$

where $\Delta A$ is the change in the contact area between particles $i$ and $j$, and $H(\cdot)$ is the Heaviside function. Charge transfer only occurs when the two particles are approaching each other, i.e., $\Delta A > 0$, to avoid the cancellation of the charging effect [57].

To ensure computational feasibility in DEM simulations, a small Young’s modulus is often employed, resulting in a non-physically large contact area. We introduce a scaling factor $A = (E'/E)^{(2/5)}$ in Eq. (11) to correct this, where $E$ is the actual Young’s modulus and $E'$ is the value used in DEM simulations.
The transfer probabilities are derived according to the electric field at the contact point \( x_{ij} \),

\[
a'_{i-j} = \begin{cases} 
1 - \text{sgn}(\varepsilon) & \frac{E'(x_{ij}) \cdot n_{ij}}{E^b} \\
1 + \text{sgn}(\varepsilon) & \frac{E'(x_{ij}) \cdot n_{ij}}{E^b} 
\end{cases}.
\]

(12)

The donor and acceptor densities at time \( t + \Delta t \) are updated as,

\[
\sigma_{dj}^+ = \sigma_{dj}^- - \frac{\Delta Q_{ij}}{4\varepsilon f'_{ij} R_j^2}, \quad \sigma_{ak}^+ = \sigma_{ak}^- - \frac{\Delta Q_{ij}}{4\varepsilon f'_a R_j^2}, \\
\sigma_{dk}^+ = \sigma_{dk}^- - \frac{\Delta Q_{ij}}{4\varepsilon f'_{ij} R_k^2}, \quad \sigma_{ak}^+ = \sigma_{ak}^- - \frac{\Delta Q_{ij}}{4\varepsilon f'_a R_k^2}.
\]

(13)

In contrast to Eq. (6), Eq. (13) includes a parameter, \( f'_{ij} \), which is used to characterize the effective surface area of wall particles in DEM simulations. This is because these wall particles are fixed to the container and only a fraction of their surface area can come in contact with mobile particles (see the gold patches in Fig. 4B for example). According to the computation of the visibility of the wall particles to the mobile particles via the ray-tracing technique, \( f'_{ij} \) is set to 0.1 for the wall particles. For mobile particles, \( f'_{ij} = 1 \), indicating that their entire surface area can exchange charges with other particles.

To check if the electric field strength at the contact point surpasses the breakdown strength, the electric field is updated by taking into account the exchanged charge \( \Delta Q_{ij} \) given by Eq. (11). \( \Delta Q_{ij} \) needs to be modified if \( \|F(x_{ij})\| > F^b_{\text{bd}} \). Previous studies have shown that the amount and distribution of charges on a particle after dielectric breakdown could be stochastic, and the charges can be reduced significantly below that set by the breakdown limit [42,43]. Therefore, we consider three possible ways to treat the charging state of the two particles in contact after dielectric breakdown:

1. Upper-limit treatment: extra charge transfer drives the electric field strength at the contact point to \( E^b_{\text{bd}} \), i.e.,

\[
\Delta Q_{ij}^{(1)} = \begin{cases} 4\varepsilon f'_{ij} R_j^2 \frac{E'(x_{ij}) \cdot n_{ij} - E_{\text{bd}}'}{R_k^2 + R_j^2}, & \text{if } E'(x_{ij}) \cdot n_{ij} > E_{\text{bd}}'; \\
4\varepsilon f'_{ij} R_j^2 \frac{E'(x_{ij}) \cdot n_{ij} + E_{\text{bd}}'}{R_k^2 + R_j^2}, & \text{if } E'(x_{ij}) \cdot n_{ij} < -E_{\text{bd}}'; \\
0, & \text{otherwise}. 
\end{cases}
\]

(14)

The total exchanged charge \( \Delta Q_{ij}^{(1)} \) used to evaluate the charge transfer in Eq. (10) is the sum of \( \Delta Q_{ij} \) and \( \Delta Q_{ij}^{(1)} \).

2. Lower-limit treatment: particles in contact become conductive and charges are evenly distributed between the two particles, i.e.,

\[
\Delta Q_{ij}^{(2)} = \begin{cases} \frac{Q_j - Q_i}{2}, & \text{if } |E'(x_{ij}) \cdot n_{ij}| > E_{\text{bd}}'; \\
\Delta Q_{ij}, & \text{otherwise}. 
\end{cases}
\]

(15)

3. Stochastic treatment: a stochastic scenario where a random value is generated to find the exchanged charge between the above two limiting cases, i.e.,

\[
\Delta Q_{ij}^{(3)} = \begin{cases} \Delta Q_{ij}^{(1)} + X(\Delta Q_{ij}^{(2)} - \Delta Q_{ij}^{(1)}), & \text{if } |E'(x_{ij}) \cdot n_{ij}| > E_{\text{bd}}'; \\
\Delta Q_{ij}, & \text{otherwise}. 
\end{cases}
\]

(16)

\( X \in [0, 1] \) is an independent random uniform variable.

As the extra transferred charges arise from the breakdown of air and/or dielectric materials near the contact site, we assume the donor/acceptor densities are unaffected by this process. In Section 4.3.1, the performance of these treatments will be tested.

4. Modeling of size-dependent charging of same-material dielectric grains

One of the key advantages of the DEM simulation is the ability to track the physical state of individual particles, which enables us to analyze the charge amount of each particle as well as the overall bulk charge polarity between different populations. Recent advancements in experimental tribocharging measurement techniques, such as those described in Waitukaitis & Jaeger [64] and Carter & Hartzell [65], have made it possible to measure the charges of individual particles. In this section, we will apply the implemented DEM CE model to study the charging behavior of insulating granular materials in a vibrating container and use the experimental data of Carter & Hartzell [24] to validate and calibrate the DEM model.

4.1. Contact electrification experiments

The CE experiments conducted by Carter & Hartzell [24] used zirconia-silica spherical beads comprising two size populations. These beads have a material density of 3800 kg/m³, and each population’s size distribution approximately follows a normal distribution. The two populations were mixed together with a designed mass ratio, denoted as \( m_{ij} = m_{\text{large}}/m_{\text{small}} \), and poured into a cylindrical container. To ensure collisions occurred only between grains of the same material, the
interior of the stainless steel container was coated with a polyurethane glue and covered with a thin layer of the zirconia-silica beads, as illustrated in Fig. 4A.

The container was then placed inside a vacuum chamber and connected to a shaker. Once the vacuum chamber reached the operational pressure of 30 μTorr, the shaker was activated with a frequency of 14 Hz, vertically shaking the grains for 5 min. Afterwards, the bottom lid of the container was rotated to a position that allows releasing a narrow stream of grains through a 1-mm-diameter hole (see Fig. 4A). At the same time, a high-speed camera was released to film the grains as they fall between two electrodes. The charge amount of each grain can be measured by analyzing the lateral movement of the grains in the external electric field from the obtained images.

4.2. DEM modeling setup

We replicated the experimental setup in the DEM simulation scenario through three steps:

1. The grain-coated container is created by randomly depositing particles from a granular mixture with a mass ratio of \( m_b = 1 \) onto a cylindrical wall and subsequently removing the interior particles.
2. The test sample, consisting of mobile particles, is generated with the designed mass ratio and packing efficiency, and settled under Earth’s gravity inside the container.
3. The container, represented by the wall particles, is vertically shaken at 14 Hz with an amplitude of 3 mm to thoroughly mix the grains and induce contact electrification.

To resemble the experimental samples, the small and the large particles have a mean diameter of 150 μm and 250 μm, respectively, with a standard deviation of 10 μm. In the CE experiments, the container contains millions of mobile particles with a packing efficiency of ~0.6. Simulating such a vast number of particles is computationally expensive. Therefore, in the simulations, a smaller container with a 1:6 ratio compared to the experiment is used. This reduces the number of mobile particles to ~10,000 while maintaining the same packing efficiency. This approach is appropriate since the collisional probability of individual particles is primarily influenced by the packing efficiency.

Initially, all particles are neutral. The simulated shaking continues for 3 min, during which all tests (except the test with \( l_b = 16 \) mm) reach a saturation charging state. Fig. 4B illustrates the initial configuration and the vibration-induced charging process of an example DEM test, clearly showing contrasting charge polarities between the large and small particles.

4.3. Comparison between numerical and experimental results

We conducted a series of tests to study the performance and reliability of the DEM CE model. Table 2 lists the adopted values of the model parameters. During the investigation of a specific model parameter, the remaining parameters were set to the underlined nominal values. For example, except in the investigation of the boundary condition, all tests assumed zero initial donor and acceptor probabilities for wall particles, as justified by the reuse of the container in the experiments and the potential unavailability of donors/acceptors on the wall particles.

4.3.1. Effect of charging state after dielectric breakdown

Our DEM CE model considers three treatments to compute the charging state of a particle pair in contact after dielectric breakdown, i.e., Eqs. (14)–(16). Fig. 5 compares the charging processes and outcomes of the simulations that use these three treatments. Initially, the particles were charged in a similar manner in all the three cases (see Fig. 5D). As time progressed to around 70 s, the electric field strength between a substantial number of collided particle pairs reached the breakdown threshold. This led to a divergence in the charging state of the particles due to discharging.

The upper-limit treatment drives the averaged charge of the small and large particle populations to continuously increase until all the particles are in the proximity of the breakdown states. As shown in Fig. 5A, the final charge-size distribution exhibits a narrow region near the upper charging limit for both populations. In comparison, the lower-limit and the stochastic treatments produce more scattered charge distribution (Fig. 5B, C, F), which is more consistent with the distribution found in the experiment (Fig. 5E).

The measured mean charge-to-mass ratios for the case of \( m_b = 8 \) in [24] were \( \overline{Q}_S = (2.3 \pm 3.4) \times 10^{-5} \text{ C/kg} \) and \( \overline{Q}_L = (-0.1 \pm 1.2) \times 10^{-5} \text{ C/kg} \) for the small and large particles, respectively. By contrast, the upper-limit case gives \( \overline{Q}_S = (8.5 \pm 4.1) \times 10^{-5} \text{ C/kg} \) and \( \overline{Q}_L = (-1.0 \pm 0.8) \times 10^{-5} \text{ C/kg} \), the lower-limit case gives \( \overline{Q}_S = (5.1 \pm 3.4) \times 10^{-5} \text{ C/kg} \) and \( \overline{Q}_L = (0.6 \pm 1.0) \times 10^{-5} \text{ C/kg} \), and the stochastic case gives \( \overline{Q}_S = (6.2 \pm 3.5) \times 10^{-5} \text{ C/kg} \) and \( \overline{Q}_L = (-0.7 \pm 0.9) \times 10^{-5} \text{ C/kg} \). The lower-limit and stochastic treatments exhibit smaller mean charges and larger standard-deviation-to-mean-charge ratios, showing a better fit.
to the experimental data (the mean charges are a few times larger, but can be reduced by adjusting the breakdown strength or boundary condition as will be shown later). Therefore, we conclude that these two treatments are more appropriate to describe the charging state after dielectric breakdown occurs. In particular, the stochastic treatment can characterize the stochastic discharging behaviors observed in various experiments [42,43]. In the following, we use this treatment throughout the DEM simulations.

4.3.2. Effect of CE model parameters

In Section 2.1, we have characterized the influence of the CE model parameters on the charge transfer between a particle pair from the mesoscopic perspective. Here we conduct a similar exercise to investigate their influence on the collective charging process of a granular system.

Fig. 6 illustrates the charge exchange process with various CE model parameters. The role each model parameter plays in the charge exchange efficiency and the final accumulated mean charge amounts of the large and small particle populations qualitatively aligns with the findings from the mesoscopic model (Fig. 2). The size-dependent bipolar charging behaviors are robustly reproduced in all the simulations, including the standard deviation, using the same color code. The horizontal lines represent the experimental results, with solid lines indicating the mean values and dashed lines representing the 1-σ interval.

A more effective way to bring the numerical mean value into closer agreement with experimental values is to reduce the breakdown strength $E_{bd}'$. By performing a linear fit of the mean values with respect to $E_{bd}'$, a strength of 0.56 MV/m would yield mean charge-to-mass ratios of $2.3 \times 10^{-5}$ C/kg and $-0.3 \times 10^{-5}$ C/kg for the small and large particle populations, respectively. These values closely resemble the experimental results.

However, 0.56 MV/m is substantially smaller than the typical air breakdown limit, e.g., $\sim 3.45$ MV/m for nitrogen gas at atmospheric pressure [44]. Moreover, at the operational pressure of 30-μTorr during the CE experiments, the air breakdown limit would be even higher by an order of magnitude according to Paschen’s law [66]. Nonetheless, we did observe small increases in the chamber pressure up to 100-μTorr when the shaker was activated, possibly due to the outgassing of the particles during collision. If this is true, the local pressure around a particle may be considerably higher than the chamber pressure, potentially reducing the breakdown limit. For instance, Manning et al. [67] experimentally measured the discharge voltage between spherical electrodes spaced 1.5 mm apart. Their results showed a discharge voltage of 100 to 1000 V at ambient pressures ranging from 0.01 to 100 torr, which aligns with the breakdown strength level derived from our study. Additionally, charges could concentrate on local regions of a particle’s surface, resulting in significantly higher local electric stresses at those locations. These localized electric strengths have the potential to exceed the typical breakdown strength.

4.3.3. Effect of boundary condition

To account for potential differences in surface charge carrier properties between wall and mobile particles, the implemented code allows for the assignment of different initial donor/acceptor probabilities to different types of particles. Apart from the nominal case, where $p_{aw0} = p_{aw} = 0$, we investigate two additional scenarios to test the influence of boundary conditions, i.e., $p_{aw0} = 0.3$, $p_{aw0} = 0.7$ (Test #1) and $p_{aw0} = 0.1$, $p_{aw0} = 0.3$ (Test #2). In the first scenario, the initial donor/acceptor probabilities of the wall particles are assumed to be the same as those of the mobile particles. In the second scenario, the available donors/acceptors on the wall particles are fewer compared to the mobile particles (representing the potential consumption in previous tests in the CE experiments).

Fig. 7 illustrates the charging outcomes under different boundary condition settings. The presence of wall particles capable of donating and accepting charges adds complexity to the evolution of particle charges. Initially, with an effective surface area fraction of 0.1, the wall
particles exhibit a higher decreasing rate in surface donor densities and tend to acquire charge carriers from the mobile particles. Consequently, both Test #1 and Test #2 show a positive leap in the mean charge of the wall particles (Fig. 7A). As time progresses, the collisional frequency between the mobile particles themselves surpasses that between the mobile and wall particles. This leads to a faster decrease in donor densities of the mobile particles, especially the smaller ones, causing the wall particles to give away carriers and resulting in a reduction in their mean charge evolution. Furthermore, due to generally lower collision probabilities, the lateral wall particles are more likely to donate carriers, while the bottom wall particles have a greater tendency to absorb carriers. This is evident from Fig. 4B, where the bottom wall particles are positively charged, while a significant fraction of the lateral ones are negatively charged.

Fig. 7B–E show that the initial donor and acceptor probabilities of the wall particles significantly influence the charge distribution of the mobile particles. The mean charge evolution and final distribution of the large and small groups in Test #1 are similar to those in the nominal test, given the same initial donor/acceptor probabilities. However, due to the substantial amount of charge carriers going to the wall particles, the final mean value of the small mobile particles is lower than that of the nominal test. In Test #2, the wall particles have a lower initial donor-to-acceptor ratio, resulting in a higher probability of absorbing charge carriers compared to Test #1. Consequently, the overall charge of the small mobile particles is further decreased, and the particle charge distribution exhibits a more scattered pattern. Test #1 yields $Q_L = (3.4 \pm 3.1) \times 10^{-5}$ C/kg and $Q_W = (-0.7 \pm 1.0) \times 10^{-5}$ C/kg, and Test #2 yields $Q_L = (3.4 \pm 4.5) \times 10^{-5}$ C/kg and $Q_W = (-0.7 \pm 1.3) \times 10^{-5}$ C/kg. The fact that the mean values and the standard-deviation-to-mean-charge ratios obtained in Test #2 align better with the experimental outcomes suggests that the boundary condition may play an important role in the charging state of the mobile particles in the CE experiments.

Fig. 8. DEM simulation outcomes for different large-to-small particle mass ratios $m_{l/s}$. Panel (A) displays the mean charge per particle of the large (blueish colorcode), small (reddish colorcode), and wall (greenish colorcode) particle populations in the three tests. The charge-to-mass ratio distributions of all particles for Test #1 and #2 are presented in Panels (B) & (C), respectively. Panels (D) & (E) show the charge probability distribution of the three populations in Test #1 and #2.

4.3.4. Effect of mass ratio

The CE experiments of Carter & Hartzell [24] also investigated the impact of the mass ratio between the large and small particle populations on the charging outcomes. To further validate the predictive capability of our DEM CE model, we conducted a series of simulations to investigate the effect of $m_{l/s}$.

As shown in Fig. 8, the simulations consistently demonstrate size-dependent polarity segregation across all tested mass ratios, with small grains predominantly charging positively and large grains charging negatively. The time taken to reach a steady state slightly decreases with a smaller $m_{l/s}$. This can be attributed to the increased total number of particles when the mass fraction of small particles is increased while maintaining the same packing efficiency. Fig. 8B clearly illustrates a trend where the absolute mean charge value decreases for the small particle population and increases for the large particle population with a smaller $m_{l/s}$. Notably, this trend aligns with the experimental results represented by the gray triangles in the plot. Moreover, the overall particle charge distributions at the end of the simulations exhibit a strong quantitative agreement with the experimental data, as illustrated in Figs. 8C and 8D, taking the case of $m_{l/s} = 2:1$ as an example.

5. Discussion and perspective

The conducted tests and comparisons with experimental results provide strong evidence of the robustness and reliability of our proposed macroscopic CE model, highlighting its potential for wide applicability in the modeling of tribocharging in insulating granular materials. In this section, we provide an overview of the model’s features and limitations, shedding light on its strengths and areas for further improvement.

5.1. Model features

Our CE model is built upon the fundamental concept that charge transfer between insulators is driven by the surface charge carrier density, as previously proposed by Lowell & Truscott [6,31] and Lacks et al. [32–34]. In order to enhance the model’s accuracy and capture
the intricacies of charge exchange between insulating materials, we incorporate recent findings related to donor–acceptor surface patches, as revealed by experimental studies [8,29] and statistical analyses [30]. This allows us to develop a mechanistic mesoscopic model that takes into account the CE-relevant surface characteristics of insulating materials. In addition, our CE model accounts for the electrostatic and stochastic discharging effects. By integrating these factors, our model robustly reproduces the observed size-dependent bipolar charging phenomena, exhibiting mean values and charge distributions that closely match experimental data.

Unlike the phenomenological-type models that require fitting empirical parameters based on post-experiment tribocharging outcomes, our model parameters, such as the breakdown strength, the transfer probability, the microscopic scale size, have explicit physical meanings and can be obtained through independent experimental measurements [e.g., 8,29,30,67]. This feature enables more reliable predictions and meaningful interpretations of tribocharging outcomes, and has the potential to provide a deeper understanding of the underlying physical processes involved in tribocharging phenomena. For instance, our model successfully replicates the large standard-deviation-to-mean-value ratio observed in experiments, suggesting that the size-dependent bipolar charging is primarily due to the difference in the surface area rather than in the surface potential. In contrast, when attempting to achieve the same large standard deviation using the effective work function model [57], fine-tuning of the modeling parameters and inclusion of distributed surface work function values are necessary [61].

Furthermore, our modeling outcomes reveal the presence of two mechanisms that can influence the polarity of the small and large particle populations, i.e., the polarity of the charge carriers and the surface donor-to-acceptor probability ratio. In the case of positive charge carriers, the tribocharging process can result in large particles charging positively and small particles charging negatively when $p_d > p_a$; Conversely, when $p_d < p_a$, the large particles tend to charge negatively, while the small particles tend to charge positively. These polarity trends are reversed by considering negative charge carriers, i.e., the large particles would charge negatively when $p_d > p_a$. In situations where $p_d = p_a$, each collision event causes an equal amount of charge carriers being lost and attained, resulting in no net change in the particle’s charge. These insights offer a plausible explanation for the disparate observations of particle polarity and the absence of size-dependent charging behavior reported in different experiments [e.g., 17,20,21], highlighting the complex interplay between charge carriers and surface properties in determining the polarity of particles during the tribocharging process.

5.2. Model limitations

In our CE model, we assume that charges and donor/acceptor elements are uniformly distributed on the surfaces of particles when calculating the exchanged charges and considering the electrostatic effect. However, as particles approach each other closely, the forces arising from polarization become important. These forces can cause particles with the same or opposite polarity to aggregate, forming clusters or chains [68]. The presence of these aggregated configurations can strongly influence the translational and rotational dynamics of the involved particles [69]. Depending on the stability of the particle configuration formed by these aggregates, there may be a preferential orientation for continuous contact among the particles, which can have a non-negligible impact on the subsequent charge exchange process over time.

To accurately capture the behavior of a granular system, particularly when it is close to a quasi-static state, it is necessary to account for particle polarization and the associated dielectrophoretic effects in the CE model. This requires a detailed particle surface model that considers the non-uniform distribution of charges and donor/acceptor elements, such as those proposed in previous studies [70–72], to characterize the particle charge distribution and its interaction with the dielectric properties of the material.

However, it is computationally expensive to simulate a large-scale granular system composed of >1,000 particles while considering the detailed charge and donor/acceptor distribution on the particles’ surfaces. In cases where the granular system is not in a quasi-static state and involves a large number of particles, it is reasonable to assume a uniform surface distribution of charges and donor/acceptor elements. Under such circumstances, our CE model can be appropriate for modeling the charge exchange process and dynamics of the granular system.

In cases where the influence of non-uniform charge distribution and polarization effects are important and the particle resolution cannot be compromised, a computationally tractable avenue for modeling the insulator CE process based on the donor–acceptor mechanism is to explore a probability distribution function-based approach. Such a probability distribution function for describing the charge/donor/acceptor surface distribution could be formulated according to local electric fields, electrostatic interactions with neighboring particles, and the historical record of contacts. This probabilistic perspective could offer a more comprehensive representation of the inherently heterogeneous charge distribution patterns on insulator surfaces. Further investigations, both theoretically and experimentally, in this direction hold potential for advancing the understanding and modeling capabilities of contact charging in insulators.

6. Conclusions

In this study, we propose a macroscopic model for contact electrification in insulating granular materials. This model incorporates the concept of surface charge carrier density and donor–acceptor surface patches, with the consideration of the electrostatic and stochastic discharging effects. By applying this model to simulate the vibration process of a granular system consisting of particles of two primary sizes, we demonstrate the model’s robustness and reliability through comparisons with experimental data.

The numerical tests successfully reproduce the observed size-dependent bipolar charging phenomena by accurately capturing the charge distributions as well as the mean values and standard deviations of the different particle populations. Notably, the low mean values of the small and larger particle populations suggest a potential low air breakdown strength of approximately 0.56 MV/m. This observation indicates the occurrence of effects such as outgassing or localized charge concentration on particle surfaces during the experiments. We also identify the influence of charge carriers and surface donor-to-acceptor probability ratio on the polarity of particle populations with different sizes. Furthermore, the simulation outcomes highlight the importance of stochastic discharging after dielectric breakdown and the influence of boundary conditions, which enable improved alignment with experimental observations. These findings contribute to the understanding of contact electrification of insulating granular materials and provide insights for future improvements and applications of the model.

CRediT authorship contribution statement

Yun Zhang: Conceptualization, Investigation, Methodology, Software, Validation, Visualization, Writing – original draft. Ali Ozel: Software, Writing – review & editing. Christine M. Hartzell: Conceptualization, Funding acquisition, Resources, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.
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