

# Electronic Supplementary Information

## Geometric characterization of optimal electrode designs for improved droplet charging and actuation

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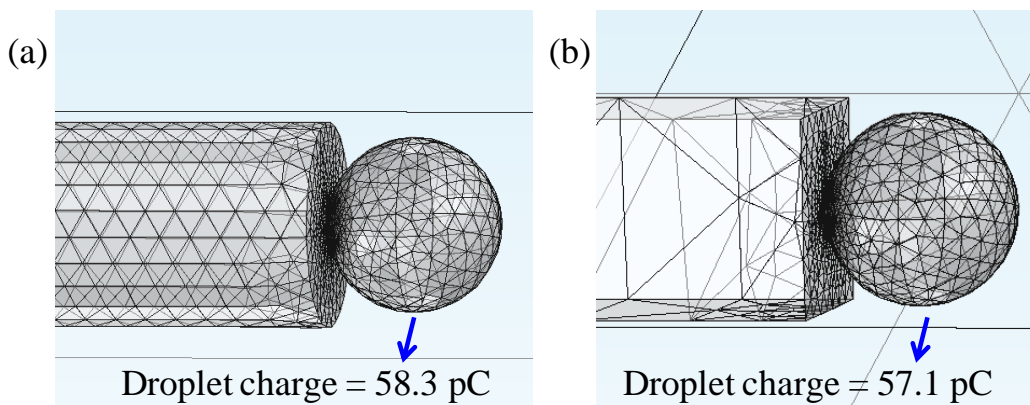
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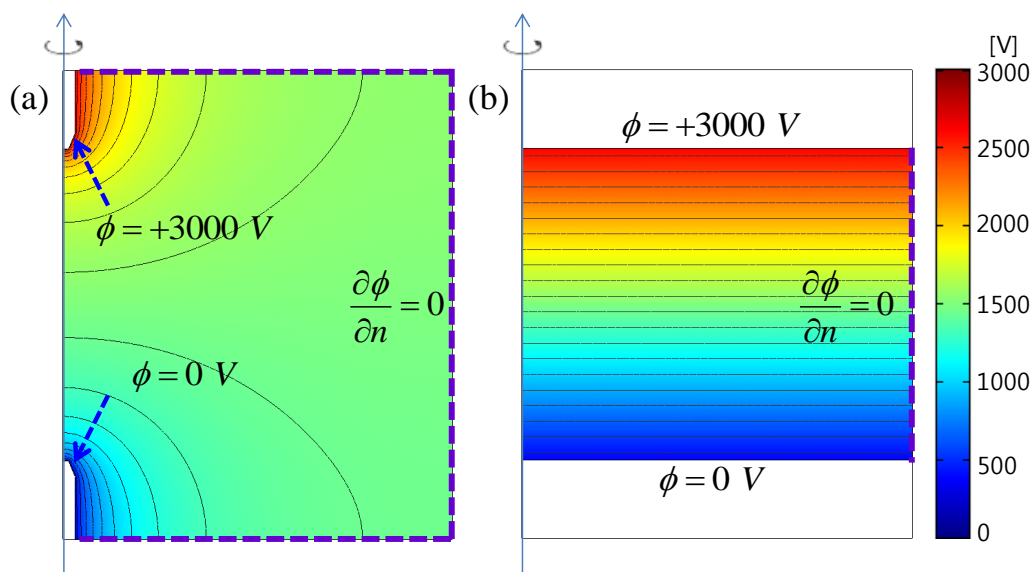
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## 1. Influence of the electrode shape on the droplet charge



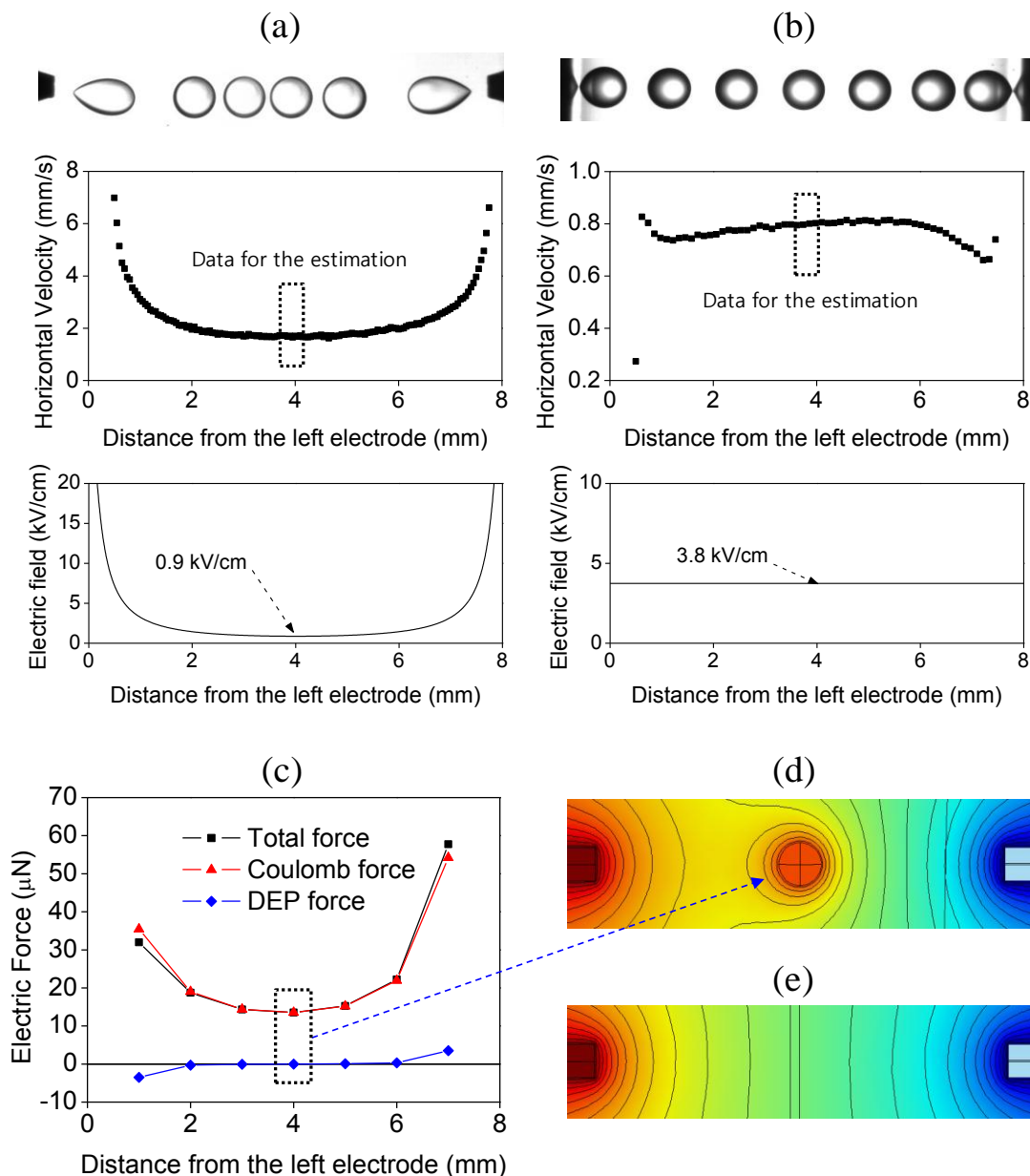
**Fig. S1.** The circular and the rectangular shape of the cross sectional area are most commonly used electrode shapes in the microfluidic devices. Therefore, numerical simulations are conducted with (a) the cylindrical electrode and (b) the rectangular parallelepiped electrode to verify the influence of the electrode shape on the droplet charge. The cross sectional surface area of two electrodes are the same ( $3.32 \times 10^{-7} \text{ m}^2$ ), the droplet volume is 100 nL and the electric field is 3 kV/cm. The droplet on the rectangular parallelepiped electrode obtains 2% less charges from the electrode than the droplet on the cylindrical electrode. This means that if the cross sectional surface area is the same, the effect of the electrode shape is relatively insignificant. Therefore, we did not consider the electrode shape as the key factor of the electrode geometry.

## 2. Electric field distribution between the electrodes



**Fig. S2** Numerical results of the electric field between the electrodes under 3.0 kV in 1000 cSt silicone oil (the dielectric constant is 2.78). The distance between the electrodes is 0.8 cm. The contour and color represent electric potential and the step size of the contour is 150 V. (a) The pin-pin electrode system. (b) The planar-planar electrode system. The dense equipotential lines near the pin-pin electrode system shows that the electric field of the pin-pin electrode system near electrode is stronger than that of the planar-planar one whereas, the sparse equipotential lines at the center of the pin-pin electrode system illustrates that the electric field of the pin-pin electrode system at the center is weaker than that of the planar-planar one, even though the two system have the same voltage difference with the same distance.

### 3. Charge estimation from the experimental images



**Fig. S3** High-speed sequential images of a translating 300 nL droplet under 3.0 kV and corresponding horizontal translational velocity profile and electric field distribution. (a) In the pin-pin electrode system. (b) In the planar-planar electrode system. As shown in electric field distribution graph, due to the highly focused electric field near the pin electrode, the droplet velocity is much faster near the pin electrode. The droplet velocity is relatively constant due to the uniform electric field in the planar-planar electrode system. Although the droplet velocity increases rapidly near the electrode in the pin-pin electrode system, the velocity in the middle region, where the velocity data extracted, is constant. Therefore, the force balance can be set using electrostatic force ( $F_E$ ) and viscous drag force ( $F_D$ ) as follows:

$$F_{Total} = F_E + F_D = QE - 4\pi\mu a U c = 0 \quad \text{with} \quad c = \frac{3\lambda + 2}{2(\lambda + 1)}, \lambda = \frac{\mu_w}{\mu} \quad (1)$$

where  $Q$  is the net charge of the droplet,  $E$  is the electric field in the middle region of the two electrodes,  $\mu$  is the viscosity of the outer medium,  $a$  is the droplet radius,  $U$  is the droplet velocity and  $\mu_w$  is the viscosity of water. Therefore, the droplet charge can be estimated from the velocity, which is measured at the center using image analyses, as follows:

$$Q = \frac{4\pi\mu a U c}{E}. \quad (2)$$

In equation (2), the electric field is important as well as the droplet velocity. However, the electric field is not uniform in the pin electrode system unlike the planar electrode system as shown in (a), (b). Thus, the electric field in the pin electrode system should be calculated numerically or analytically as shown in Fig. 2(b). Even though the electric field can be obtained from the numerical simulation, it still remains open to question about the influence of the charged droplet on the electric field distribution because the existence of the droplet carrying certain amount of charge that can distort the electric field did not considered in the calculation.

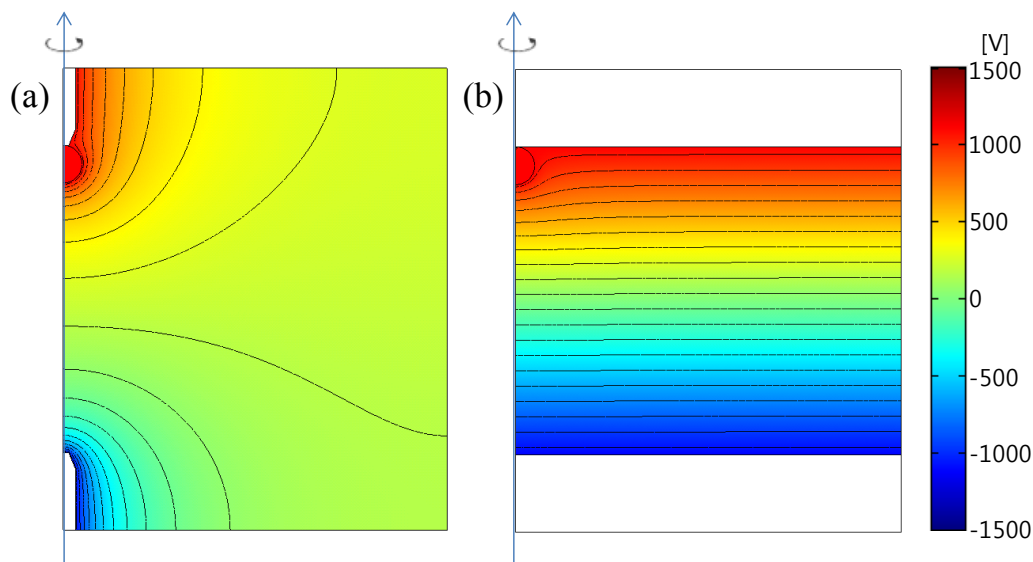
In order to verify the effect of the charged droplet on the charge estimation more clearly, the force is calculated with two different procedures. First, the force ( $F_{E1}$ ) is numerically calculated according to equation (3) with the charged droplet with known charge ( $Q$ ) on its surface as shown in (d); thus, the force ( $F_{E1}$ ) includes the effect of the electric field distortion due to the charged droplet. Second, the force ( $F_{E2}$ ) is calculated by the point charge assumption ( $F=QE$ ) with the known droplet charge ( $Q$ ) and the electric field ( $E2$ ) which is calculated without considering the charged droplet as shown in (e); thus, the force ( $F_{E2}$ ) does not include the effect of the electric field distortion due to the charged droplet. The calculated forces are  $F_{E1} = 13.5 \mu\text{N}$  and  $F_{E2} = 13.8 \mu\text{N}$  respectively. Rigorously speaking, the charge should be estimated by the force balance  $F_{E1} + F_D = F_{E1} - 4\pi\mu a U c = 0$ . Here, because both the charge and electric field are unknown, the charge should be obtained by an iterative numerical calculation to match with measured droplet velocity  $U$ . However, because the two forces ( $F_{E1}$  and  $F_{E2}$ ) are very similar (the difference is about 2%), for simplicity, we estimated the droplet charge using the point charge assumption with the electric field without considering the presence of the charged droplet.

The electric forces exerted on the droplet are also calculated numerically as shown in (c). The total force exerted on the droplet is calculated by the following surface integration:

$$F = \int_S n \cdot T dS \quad (3)$$

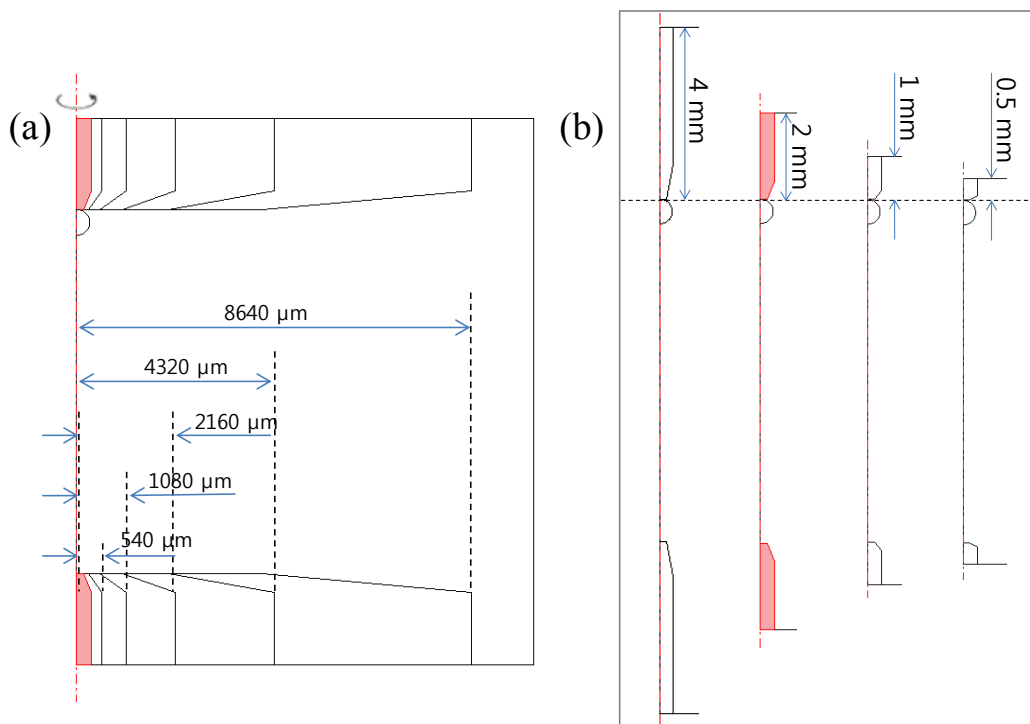
where  $n$  is the outward normal vector,  $S$  the droplet surface, and  $T$  the Maxwell stress tensor. The dielectrophoretic (DEP) force on the droplet due to the nonuniform electric field was also calculated without charge on the droplet at each position. Because the horizontal total driving force on the droplet consists of the DEP force and Coulomb force, the Coulomb force can be obtained using the difference between the total force and the DEP force from the numerical calculations. As a result, the derived Coulomb force includes all influences from the electric field distortion that result from the charged droplet near the electrodes. As shown in (c), even in the pin-pin electrode system, the DEP force in the middle region of the two pin electrodes is zero; thus, we don't need to consider the DEP force for the droplet charge estimation.

#### 4. Calculation of the charge of a droplet contacting an electrified electrode



**Fig. S4** The calculation of the charge of a droplet on the electrode under 3.0 kV in 1000 cSt silicone oil. The volume of a droplet is 0.5  $\mu\text{L}$ . The charge is calculated by integrating the surface charge density along the droplet surface (Surface charge density  $\sigma = \epsilon_{in} \frac{\partial V_{in}}{\partial r} - \epsilon_{out} \frac{\partial V_{out}}{\partial r}, V_{in} = V_{out}$ ). For thorough investigation, the charge of a droplet of various sizes (0.1 ~ 1  $\mu\text{L}$ ) under potential difference (1.8 ~ 3.0 kV) was analyzed. (a) The pin-pin electrode system. (b) The planar-planar electrode system. In the simulations, the effect of deformation of the droplet is not included for the convenience of the parametric study. The droplet in the pin-pin electrode system shows much greater deformation than the droplet in the planar-planar electrode system; thus, if the deformation effect is considered, the superior charging on the pin electrode will increase.

## 5. The numerical analysis on the effects of electrode geometries



**Fig. S5** The representative geometries of electrode for various radii and lengths. The red colored electrode geometry is the real shape of the electrode which is used for the experiment (radius is 320  $\mu\text{m}$  and length is 2 mm). The radius of electrode is varied from 86  $\mu\text{m}$  to 35 mm and the length of electrode is varied from 0 mm to 32 mm. The charge of a droplet is calculated for each combination of radius and length. (a) The geometries of electrode for various radii with fixed length. (b) The electrode geometries for various lengths with fixed radius.