Electronic Supplementary Material (ESI) for Lab on a Chip.

Supplemental material

Particle size spectrometer using inertial classification and electrical measurement techniques for real time monitoring of particle size distribution

Hong-Beom Kwon, Hong-Lae Kim, Ui-Seon Hong, Seong-Jae Yoo, Kyongtae Kim, Jangseop Han, Min-Ki Kim, Jungho Hwang and Yong-Jun Kim*

School of Mechanical Engineering, Yonsei University, Republic of Korea

* To whom correspondence should be addressed: yjk@yonsei.ac.kr

[Derivation of eqn (1), the transfer function for converting the current data set to particle size distribution]

$$I_i = eQ \cdot \int_{d_{p,min}}^{d_{p,max}} \bar{q}(d_p) \cdot C_i(d_p) \cdot N(d_p) \cdot dd_p, \qquad i = 1, 2, 3, 4 \tag{1}$$

Electrical particle-detection technique (electrical technique) is one of the established methods used for measuring the particle number concentration in aerosol-science.[1] The key points of the electrical technique are that (i) the average number of charge that particles can carry is constant if their diameter are the same, and (ii) it can be experimentally determined.[2] The average number of charge per particle ($\bar{q}$) as a function of particle size (d_p) can be fitted with power-law function where α and β are constants [eqn (S1)] .

$$\bar{q}(d_p) = \alpha \cdot d_p^{\beta} \tag{S1}$$

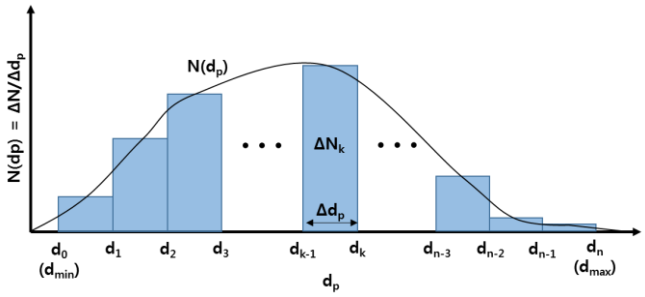

Figure S1. The arbitrary particle size distribution

Figure S1 shows the arbitrary particle size distribution [$N(d_p)$]. The set of blue blocks is the discretized size distribution with n intervals and an even spacing (Δd_p). The area of the kth block is the particle concentration in the size range of d_{k-1} - d_k [$\Delta N_k = N(d_p) \times \Delta d_p$ (N cm^{-3})]. Considering the particles are charged, the current from the kth block [I_k (C·s^{-1})] can be expressed as eqn (S2), where Q is the volumetric flow rate of sample stream (cm^3·s^{-1}) and e is the elementary charge (1.602 × 10^{-19} C).

$$\Delta I_k = Q \cdot e \cdot \bar{q}_k \cdot \Delta N_k, \qquad k = 1, 2, \ldots, n \ . \tag{S2}$$

Thus, the total current (I) from the arbitrary particle size distribution can be expressed as

$$I = \sum_{k=1}^{n} Q \cdot e \cdot \bar{q}_k \cdot \Delta N_k \ . \tag{S3}$$

However, the collection fraction at ith stage of the inertial size separator (C_i) changes depending on the size of the particles. Thus, the current measured from the ith stage (I_i) can be expressed as

$$I_i = Q \cdot e \cdot \sum_{k=1}^{n} \bar{q}_k \cdot C_{i,k} \cdot \Delta N_k = Q \cdot e \cdot \sum_{k=1}^{n} \bar{q}_k \cdot C_{i,k} \cdot N_k \cdot \Delta d_k, \qquad i = 1, 2, 3, 4 \ . \tag{S4}$$

If the size interval is very small ($n \rightarrow \infty$), eqn (S4) can be expressed as an integral form [eqn (1)].

[The technique for controlling ion concentration]

Most of the ions generated from the discharging electrode are precipitated on the ground electrode. Thus, by monitoring and controlling the current passing through the ground electrode (ion current), the ion concentration can be maintained as a constant. Figure S2 shows the simplified schematic diagram of the circuit for controlling the ion current. There is a current-to-voltage converter before the ground electrode. It detects a voltage drop caused by ion current passing through a 100 kΩ feedback resistor. OP Amp measures the difference of the two voltages: (i) the measured voltage and (ii) the target voltage from micro controlling unit (MCU). The target voltage is the value of the voltage drop when the target ion current passing through the feedback resistor. For example, if the target ion current is 2.0 μA, the target voltage is $(2.0 \times 10^{-6}\ \text{A})(100 \times 10^{3}\ \Omega) = 0.2$ V. Based on the voltage difference (target voltage – measured voltage), the OP Amp controls the discharging voltage to make the voltage difference zero by adjusting the input control voltage of the high voltage supplier. For example, if the measured voltage is smaller than the target voltage, the high voltage supplier will increase the discharging voltage, leading to the increment of the ion current.

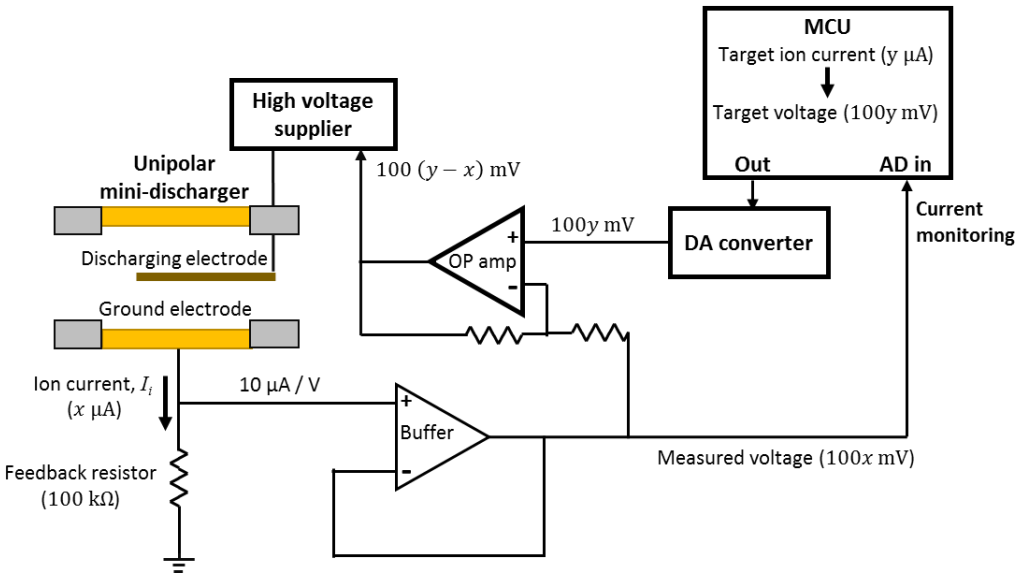

Figure S2. Simplified circuit for controlling the ion current

[Experimental setup]

Particle generation process

Figure S3 shows a schematic diagram of the simplified experimental setup for characterizing the components and overall performance of the proposed system. As shown in Fig 3 (a), compressed air passed through a high efficiency particulate air (HEPA) filter and then supplied at a flow rate of 1.55 L min^{-1} using a mass flow controller (MFC). Two types of particles were used. Monodisperse polystyrene latex (PSL) particles having a size range of 0.11 – 1 μm (Thermo Fisher Scientific, USA) were used for characterization of the unipolar mini-charger and inertial size separator, and polydisperse TiO_2 particles were used for a performance evaluation of the proposed system. The generated TiO_2 particles were initially charged by a soft X-ray charger (XRC-05, HCT Inc., KR) and passed through a differential mobility analyser (DMA; model 3081, TSI Inc., USA). The mean size of the TiO_2 particles was controlled by adjusting the voltage of the analyser, which classified the charged particles based on their electrical mobility. A diffusion dryer was used to remove moisture from the sample stream; any charged particles were neutralized by a neutralizer.

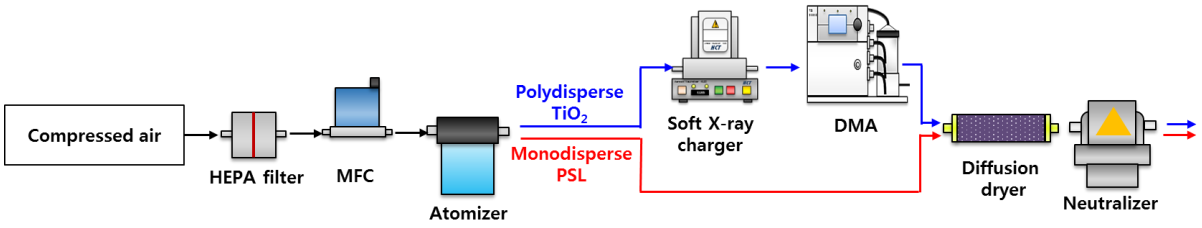

Figure S3. Schematic diagram of experimental setup for (a) generating monodisperse PSL and polydisperse TiO_2 particles

Experimental setup for humidity effect on the unipolar mini-discharger

By changing the humidity of the airstream introduced in the charger, we evaluated the robustness of the unipolar mini-discharger. The controlled humidity can be realized by using two mass flow controllers (MFCs). Figure S4 shows the experimental setup for the humidity experiment. The air stream from MFC #1 gets dried in the diffusion dryer, while the air stream from the MFC #2 passes through the water-filled bubbler and get humidified. By controlling the ratio of the two flows, the degree of humidity can be controlled.

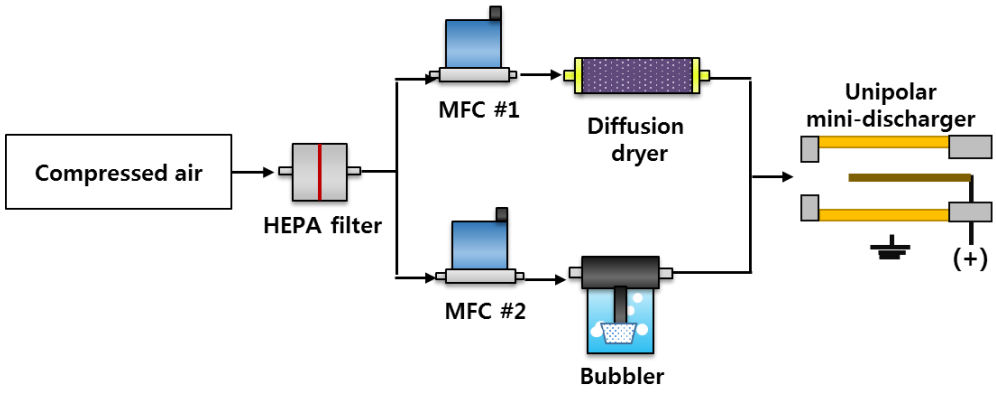

Figure S4. Schematic diagram of experimental setup for identifying the humidity effect on the unipolar mini-discharger

[Raw data sample]

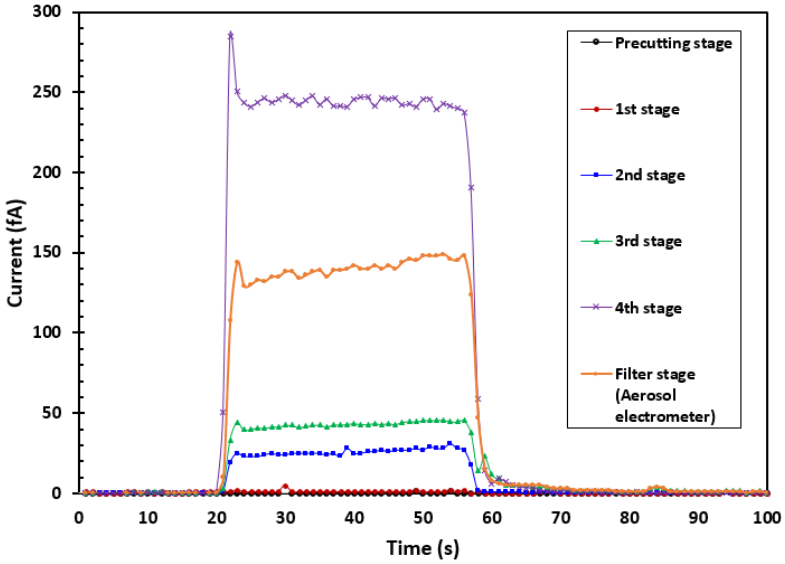

Figure S5. Measurement result of the currents at each stages when 300 nm-sized particles introduced (sampling interval: 1 s). The rise time of the electrometer was within 2 – 3 s, proving that electrical connection between the collection plate and the current-sensing electrode on the glass slide was made immediately.

[Particle material effect on the proposed system]

The electrical particle-detection technique (electrical technique) is one of the firmly-established techniques including optical and gravitational techniques.[1] The electrical technique is currently used in the high-precision instruments including the electrical low pressure impactor (ELPI; model ELPIR+, Dekati Ltd., Finland) and the fast mobility particle sizer (FMPS; model 3091, TSI Inc., USA).

The key points of the electrical technique are that (i) the average number of charge that particles can carry ($\bar{q}$) is constant if their diameters are the same, and (ii) it can be experimentally determined and fitted using power-law function when Ni·τ (ion concentration × residence time) is controlled. If particles with various properties matches the fitted curve, the electrical technique will show the same performance regardless of any incoming particles even including micro-organisms. However, in real-world situation, the fitted curve is changed by electrical properties of particles, like other techniques which have their own limitations induced by numerous properties of particles (refractive index, optical reflectivity, hygroscopicity, volatility. etc.).[1]

We conducted experiments for identifying average number of charges per particle ($\bar{q}$) as a function of particle size (d_p) with polystyrene latex, TiO_2, and Ag in the range of 0.05 – 0.3 μm. Figure S6 shows that with the increment of the conductivity of particles, $\bar{q}$ also increases, indicating that the measurement accuracy can decrease when particles with various kinds of material are introduced. However, this problem can be solved by developing and optimizing the unipolar mini-discharger.

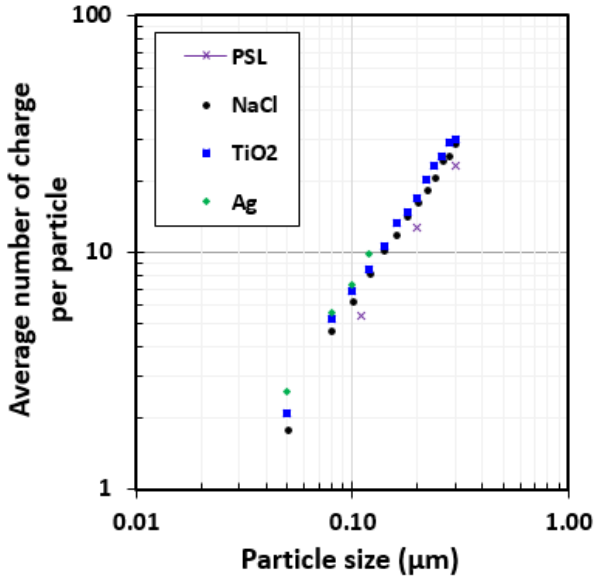

Figure S6. Average number of charge per particle as a function of particle size for various particle properties.

Leonidas Ntziachristos and co-workers reported the corona discharger which exhibits the robust performance in terms of particle material.[3] In spite of two kinds of particles [Ag (conductive), di-octyl sebacate (DOS; nonconductive)] having large difference in terms of electrical properties, both particles were well fitted to a single power-law function, meaning that particle material effect on $\bar{q}$ was negligible. Also, other researchers have reported unipolar dischargers exhibiting the similar performance in the nanometer to micrometer-size range.[4,5]

In this study, we focused on the realization of an inexpensive and compact inertial size separator and the successful operation of the entire system (discharger, inertial size separator, multi-channel electrometer, retrieval algorithm), which were validated by using test aerosol with a single composition. The optimization of each component including unipolar mini-discharger will be carried out as a future work, expected to minimize the particle material effect on the electrical technique.

[Monitoring the real-environment with the comparison of the high-precision standard instrument]

The performance of the proposed system was tested by comparing the measurement data with the scanning mobility particle sizer (SMPS, TSI Inc., USA) in our laboratory where various particles were introduced from the outside through windows and were generated from human activity, printer and other operating machines. By using Y-shaped fitting pipes and adjusting the tube length to guarantee the same transport time, airborne particles were transported to the both system at the same concentration. The whole sampling time was about 10 minutes and measured data were averaged.

SMPS is a high-precision instrument which characterize particle size distribution based on electrical mobility diameter of a particle. For the detection of particles in the size range of 0.1 - 1 μm, SMPS uses 67 discrete size bins at a logarithmically even spacing of $\Delta \log d = 0.015$.

As shown in Fig. S7, the proposed system retrieved particle size distribution to some extent; the percentage deviations of the peak size and number concentration between the proposed system and SMPS were 34.0% and 23.7 %, respectively. The proposed system could not accurately detect the particle size below 0.2 μm. This limitation has two reasons: (i) the smallest cut-off diameter in the inertial size separator was 0.26 μm, and (ii) due to stability of the algorithm, it was assumed that the both ends of the size range of interest [0.1 μm, 1.2 μm] be close to zero. These problems can be solved by adding additional stages with cut-off-diameter below 100 nm. Also, the introduced particles with various kinds of material resulted in the deviation of the peak diameter. This was because unipolar mini-charger, which has not been optimized yet, exhibited the different performance depending on electrical properties of particles.

The proposed system exhibited some limitations in monitoring the real-world environment compared with the commercial high-precision instrument. However, by developing and optimizing each component in our proposed system, the proposed system is expected to precisely monitor even the real-world environment in the future.

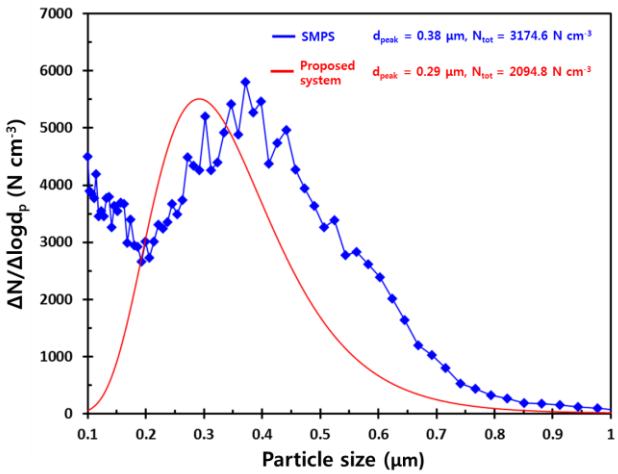

Figure S7. The comparison test results of the proposed system and SMPS in a real-world environment. Blue and red line represent the discretized size distribution from SMPS and the continuous size distribution from the proposed system, respectively.

Reference

1. P. Kulkarni, P. A. Baron and K. Willeke, *Aerosol measurement: principles, techniques, and applications*, John Wiley & Sons, 2011.
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