8-7 Classification of Monodisperse Aerosol Particles using an Adjustable Soft X-ray Charger

Classification of Monodisperse Aerosol Particles using an Adjustable Soft X-ray Charger

Bangwoo HAN[1,2], Manabu SHIMADA[1*], Kikuo OKUYAMA[1] and Mansoo CHOI[2,3]

[1]Department of Chemical Engineering, Graduate School of Engineering, Hiroshima University,
Higashi-Hiroshima 739-8527, Japan
[2]School of Mechanical and Aerospace Engineering, Seoul National University, Seoul 151-742,
South Korea
[3]National CRI Center for Nano Particle Control, Institute of Advanced Machinery and Design,
Seoul National University, South Korea

Submitted to POWDER TECHNOLOGY
on November 8, 2002.
revised on February 19, 2003

*Address correspondence to Dr. Manabu Shimada, Department of Chemical Engineering, Graduate School of Engineering, Hiroshima University, 4-1, Kagamiyama 1-chome, Higashi-Hiroshima 739-8527, Japan. E-mail: smd@hiroshima-u.ac.jp

Running title: ADJUSTABLE SOFT X-RAY CHARGER

ABSTRACT

At high concentrations of ambient bipolar ions, charged aerosol particles show a stationary charging state as a function of particle size. When the particle diameter exceeds 0.1 μm, a considerable fraction of multiply charged particles is present, which hinders size classification of particles, when a differential mobility analyzer is used. However, at low ion concentrations, particle charging by the ions can be suppressed, and in principle, the production of multiply charged particles can be reduced even when the particle size is larger than 0.1 μm.

In this study, a use of a power-adjustable soft X-ray emitter was investigated for the first time as the ionization source of an aerosol charger. The bipolar ion concentration was found to be adjustable between 10^{10} and 10^{13} ions/m^3. Through proper control of the bipolar ion concentration in the soft X-ray aerosol charger, multiply charged particles were reduced to less than 5% of the singly charged particles while the singly charged particles remained at a relatively high concentration with most of the particles in size range of 0.1-1.0 μm. Therefore, highly monodisperse aerosol particles in the 0.1-1.0 μm diameter range could be generated using this technique with a commercialized differential mobility analyzer.

Keywords: Soft X-ray; Bipolar charging; Monodisperse; Aerosol particle; Photoionization

1. INTRODUCTION

The generation of monodisperse aerosol particles is essential in the study of size-dependent aerosol properties (e.g. diffusion coefficient or electrical mobility) as well as the performance evaluation of aerosol instruments such as impactors, cyclones and filters. A typical procedure for generating monodisperse particles is to classify polydisperse particles on the basis of particle mobility using a differential mobility analyzer (DMA). While the DMA functions well when aerosol particles smaller than about 0.1 μm are involved, it becomes difficult to apply to particles larger than 0.1 μm because equi-mobility particles obtained by DMA classification contain a considerable amount of multiply charged species with sizes larger than those of the singly charged particles.

Aerosol particles in the 0.1-1.0 μm diameter range are very important in the study of atmospheric aerosols such as cloud nucleation and gas-aerosol reactions. A method for classifying monodisperse or nearly monodisperse atmospheric aerosols is necessary for such purposes. Romay-Novas and Pui [1] used a technique that involved combining a DMA and a micro-orifice impactor to remove undesirable multiply charged particles of larger sizes. This impactor technique was efficient in producing monodisperse particles in a relatively large particle size range (≥0.5 μm). However, an adjustment of the impactor cutoff point is necessary at each time the size of the aerosol particles is changed. Furthermore, the loss of singly charged particles is too high owing to the dilution of the aerosol in the impactor. Gupta and McMurry [2] used a low-activity radioactive (0.09 μCi ^{63}Ni) charger to reduce the production of multiply charged particles due to low ion concentration. This technique has advantage over the impactor technique in the sense that it is not affected by the density of the particle material. It is also more effective for small size particles (≤0.5 μm) than the impactor technique.

In the charger technique, the aerosol concentration and flow rate as well as the ion concentration are dominant parameters, thus the input aerosol conditions strongly affects the resulting charging state of the charger. However, the ion production rate is hardly alterable in the low-activity radioactive source charger, thus, the charging time (or aerosol flow rate) must be controlled in order to acquire a suitable monodispersity aerosol, each time the size of particles is changed. It is difficult to control the charging time to obtain particles of an adequate monodispersity in the overall particle size range of 0.1-1.0 μm, because the charging time depends not only the aerosol flow rate but the pattern of aerosol flow in the charger as well, and is, hence, difficult to estimate. If the ion concentration of the charger could be controlled, the particle charging state can be easily regulated so as to be largely singly charged in a variety of aerosol conditions (input particle sizes and concentrations) without the need to change the aerosol flow rate.

In fact, Gupta and McMurry [2] reduced the ion concentration by selecting a low-activity radioactive source. It is, however, by no means practical to adjust the source activity according to particle size and concentration. In this study, a soft X-ray photoionization charger equipped with an adjustable X-ray power was used instead of a low-energy radioactive charger. A photoionization charger using soft X-ray (<10 keV) can be more safely and easily handled than radioactive chargers. That is, no special radioactive manager is necessary and the device can be simply turned on and off. In our previous studies, it was shown that a soft X-ray charger was capable of generating very stable and a high concentration of bipolar ions [3] and it had a high unipolar charging efficiency in the nanoparticle size range of less than 50 nm [4]. By using the power-adjustable soft X-ray charger, bipolar ion concentrations in the charger can be changed, and thus, it becomes easy to obtain nearly monodisperse particles of 0.1 - 1.0 μm in diameter by classification using the DMA.

2. THEORETICAL APPROACH

Our previous studies [3,4] showed that the charging of aerosol particles using a soft X-ray charger could be well evaluated by the diffusion bipolar/unipolar charging theory. However, in those studies, only time-independent equilibrium charging states were considered, and thus, the analytical solutions ($\mathrm{d}n_p/\mathrm{d}t = 0$ for all p) were calculated to estimate the experimental charging states. However, in this study, it becomes necessary to calculate time-dependent changes of particle charging state to determine ion concentrations and charging time which are appropriate to bring particles into the nearly singly charged state.

The basic equations of bipolar diffusion charging can be expressed as follows [5]

$$\frac{\mathrm{d}n_\mathrm{i}^+}{\mathrm{d}t} = -\alpha n_\mathrm{i}^+ n_\mathrm{i}^- - \sum_{p=-\infty}^{+\infty} \beta_p^+ n_p n_\mathrm{i}^+ + S, \qquad (1)$$

$$\frac{\mathrm{d}n_\mathrm{i}^-}{\mathrm{d}t} = -\alpha n_\mathrm{i}^+ n_\mathrm{i}^- - \sum_{p=-\infty}^{+\infty} \beta_p^- n_p n_\mathrm{i}^- + S, \qquad (2)$$

$$\frac{\mathrm{d}n_p}{\mathrm{d}t} = \beta_{p+1}^- n_{p+1} n_\mathrm{i}^- - \beta_p^- n_p n_\mathrm{i}^- + \beta_{p-1}^+ n_{p-1} n_\mathrm{i}^+ - \beta_p^+ n_p n_\mathrm{i}^+, \qquad (3)$$

where n_p is the number concentration of p-charged particles. β_p^+ and β_p^- the combination coefficients of a p-charged particle with a positive and negative ion, respectively. α and S are the recombination constant ($=1.6\times10^{-12}$ m^3/s; [6]) and the production rate of bipolar ions, respectively. n_i^+ and n_i^- are the concentrations of positive and negative ions, respectively. Equations (1) and (2) represent time-dependent changes in number concentrations of positive and negative ions, respectively. The first, second and third terms on the right-hand sides of these equations indicate the loss rate due to the recombination of bipolar ions, the loss rate due to collision with particles, and the production rate due to soft X-ray photoionization. Equation (3) represents the changes in concentrations of p-charged particles (n_p) with time. Each term on the right-hand side of the equation expresses the gain or loss rate of p-charged particles due to collisions with ions. The Fuchs theory [7] was used to calculate the value of β_p because of its good agreement with many charging experiments [8,9]. In the calculation, we used 130 amu and 100 amu for mass of positive and negative ions, and 1.1×10^{-4} and 1.3×10^{-4} m^2/(V·s) for electrical mobility of positive and negative ions, respectively, as were used in our previous studies [3,4].

Calculations of the single and double charge ratio n_1/n_T, n_2/n_1 ($n_T=\sum_{-\infty}^{\infty} n_p$) with the particle diameter for different ion concentration (n_i), particle concentration (n_T) and charging time (t_r) are shown in Figs. 1(a)-1(c), respectively. Charge ratios are derived from positively charged particles because all experiments in this study deal with positively charged particles. The number concentration of positive ions, n_i^+ is assumed to be the same as that of negative ions, n_i^-. Most of the charged particles appear to be singly charged at ion concentrations lower than the particle concentration and at a relatively short charging time. Therefore, as the ion concentration or charging time is decreased, particles of better monodispersity will be obtained by classification using the DMA. However, a lower ion number and shorter charging time also lead to singly charged particles to be smaller, which is undesirable. In this study, the fraction of the two charged particles was targeted to be about $n_2/n_1\sim5\%$ ($\sigma_g\sim1.12$) in overall range in order to obtain a suitable concentration of n_1.

3. EXPERIMENTAL SETUP

Figure 2 shows the soft X-ray aerosol charger developed and used in this study. It is equipped with a soft X-ray emitter (Photoionizer C4870, Hamamatsu Photonics, Japan) having current (0-200.0 μA) and voltage (1.60-10.00 kV) adjusters to change the X-ray power. A soft X-ray is emitted from a circular beryllium window 15 mm in diameter at a solid angle of about 120°. The entire charger is enclosed with a cylindrical polytetrafluoroethylene container with a wall thickness of over 3 mm to prevent X-rays from escaping outside. The upper and lower walls of the chamber are made of stainless steel and act as electrodes for the ion concentration measurements. For the rest of experiments to generate nearly singly charged particles, no voltage was applied to the charger. The predicted charging time of aerosols in this charger is about 0.16 s for the carrier gas of 1 *l*/min. This value was obtained by the best fit between the calculated and experimental data in our previous study which used the same charger as this one [4].

Experiments were conducted in two separated ways. One was intended to prove the reduction of multiply charged particles under low ion concentration conditions using standard monodisperse polystyrene latex (PSL) particles. Another was to verify the monodispersity of the particles classified from a polydisperse aerosol using a DMA combined with the soft X-ray charger.

Figure 3(a) shows the setup used in the first experiment using PSL particles. PSL particles (STADEX, JSR Co., Japan) were dispersed into air using a pressurized nebulizer (Nanomaster, JSR Co., Japan) and were then passed through a dry column to eliminate water in the aerosol. They were then fed into a ^{241}Am neutralizer and an electrostatic precipitator (applied with a dc voltage of ~10 kV) to bring the aerosol particles into uncharged state. The resulting uncharged, monodisperse particles were fed into the soft X-ray charger followed by a DMA (Model 3071, TSI inc., USA) and a condensation nucleus counter (CNC, Model 3022A, TSI inc., USA) to determine the charging state of the particles charged by the soft X-ray charger. N_2 gas was used as a carrier gas and 1 *l*/min of aerosol flow rate was introduced into the X-ray charger in all experiments. Aerosol and sheath gas flow rates were maintained at a ratio of 1:10 in the DMA.

Figure 3(b) shows the apparatus used to classify monodispere particles from polydisperse aerosols generated with the spray drying method. By means of the spray drying method, particles larger than 0.1 μm in diameter and of excellent sphericity could be produced and the mean size of the particles could also be controlled from 0.1 to 1.0 μm by changing the colloidal nanoparticle concentration [10]. The particles prepared by the spray drying method were therefore, very suitable as test particles to validate the soft X-ray charger. Nanometer-sized silica sols (Snowtex, Nissan Chemical Ind., Ltd.) were used as the precursor. The silica sols were nebulized into air using an ultrasonic nebulizer (NE-U11, Omron Corp.). The droplets were then transported by the carrier N_2 gas into the tubular furnace. Inside the furnace, the droplets were evaporated, and finally, dry spherical SiO_2 particles were produced. Table 1 shows the experimental conditions used in generating polydisperse silica particles using the spray drying method. The concentration of spray-dried silica particles was about 2×10^6 particles/cm^3 and the mean diameters were approximately 0.3 and 0.6 μm for the each case of sol concentrations 0.01 and 0.1 M. The geometric standard deviations were about 1.40~1.45. The particles were introduced successively into a ^{241}Am charger and an electrostatic precipitator as was done for the PSL aerosols. The uncharged polydisperse particles were then fed into the soft X-ray charger followed by the DMA. The newly-charged and classified particles by the soft X-ray charger and DMA series were collected on the surface of chopped Si wafers by an electrostatic collector, using an applied voltage of 8 kV. The collection time was 1-2 h. The morphology of the collected SiO_2 particles was examined using a Field Emission-Scanning Electron Microscope (FE-SEM; S-5000, Hitachi, Ltd.).

4. RESULT AND DISCUSSION

Figure 4 shows the change in measured ion current with applied voltage. To measure this, the inlet and outlet of the charger were closed and an electric field was applied between the two electrodes of the charger. The ion current induced by the resulting electric field in the chamber was measured by means of an electrometer (Model 6512, Keithley, USA). The number concentration of bipolar ions can be evaluated by the following equation [11]:

$$n_i = \sqrt{I_0/(\alpha e V)}, \tag{4}$$

where, I_0 is the saturation current in the charger, e the elementary electrical charge and V the volume of charger ($=1.64\times10^{-4}$ m^3). Bipolar ion number concentrations n_i predicted from Eq. (4) are shown in Table 2. By adjusting the X-ray power, the ion number concentration of the charger could easily be regulated to the values of our choice. However, ion concentrations ($n_i = 10^9 \sim 10^{11}$ ions/m^3) corresponding to the experimental conditions to bring particles into the mostly singly charged state was impossible to measure because of the high current noise of our system. Therefore, more rigorous calculations corresponding to the experimental conditions could not be conducted.

Figures 5(a) and 5(b) show the charging states of the 0.207 μm and 0.791 μm PSL particles, respectively. The case of maximum power of soft X-ray represents the bipolar equilibrium charging state. Although not shown in this paper, identical results could be obtained when a soft X-ray charger (with a maximum power) was replaced by a ^{241}Am charger. This is consistent with our previous findings [3] where the soft X-ray charger generating a high number of bipolar ions could bring aerosols into the well-known equilibrium charging state as the ^{241}Am charger. In the equilibrium state, multiply charged particles were easily detected and the fraction of multiply charged particle increased with particle size. Even particles charged to five elementary charges could be observed for the 0.791 μm PSL particles whereas triple charged particles have their maximum for the size of 0.207 μm. However, as the X-ray power was decreased, the fractions of multiply charged particles were drastically decreased. At proper low-power conditions, singly charged particles were mostly dominated.

Figures 6(a) and 6(b) show the charge ratios of positively charged particles, n_2/n_1 and n_1/n_T, as a function of PSL particle size, respectively.[†] Circles, squares and triangles show the experimental results for different soft X-ray power conditions and the dotted line shows the calculated values using Eq. (3) for the bipolar equilibrium charging state. For the maximum power case, experimental values are in good agreement with the calculated ones, which again proves that the soft X-ray can bring aerosols into steady state charge distributions. The fraction of doubly charged particles increases with increasing particle size, irrespective of the ion concentration. At an X-ray power of 30.0 μA, 3.00 kV, singly charged particles were largely obtained in a particle range of less than 0.5 μm. While very high-monodispersity particles could be generated for the 0.1~0.2 μm (n_2/n_1 = 1~2%), the fraction of singly charged particles was too low (n_1/n_T =

[†] Total particle numbers (n_T) of Fig. 6(b) could not be measured directly owing to the residue particles detected by CNC. They were estimated from the calculated equilibrium charging ratios and

2~3%). If the X-ray power was slightly increased, more singly charged particles will be obtained while the fraction of doubly charged particles may be preserved at about 5%. For the case of larger particles, much lower ion concentration condition (i.e. 15.0 μA, 3.00 kV) was needed to remove doubly charged particles (n_2/n_1~5% for the 0.603, 0.791 μm PSL particles). Even in that case, a non-negligible amount of the doubly charged fraction, n_2/n_1 survived for the case of the 1.008 μm particle. To acquire a better monodispersity for the 1.0 μm particles, a slightly lower ion concentration may be necessary but at the cost of singly-charged particles' concentration ($n_1/n_T < 5\%$).

Figures 7(a)-(d) show FE-SEM images of SiO_2 particles classified by the DMA which was preset to obtain the particles 0.3 μm and 0.6 μm in diameter in the spray drying method. Figures 7(a) and 7(b) show the particles 0.3 μm in diameter for the maximum power case (i.e. ordinary bipolar equilibrium charging case) and 30.0 μA, 3.00 kV case, respectively. Figures 7(c) and 7(d) show the particles of 0.6 μm at the maximum power and 15.0 μA, 3.00 kV case, respectively. The size distributions of these particles were determined by randomly sampling 300 particles from the FE-SEM photographs, and are shown in Figs. 8(a)-8(d). As can be seen from the figures, multiply charged large particles are easily classified with singly charged particles by the DMA for the maximum ion concentration cases. In contrast to this, for the low ion concentration conditions, the fraction of undesirable multiply charged particles was drastically decreased and monodisperse particles ($\sigma_g \sim 1.15$) were largely obtained.

5. CONCLUSION

Using a power-adjustable soft X-ray charger, the production of mostly singly charged aerosols in the diameter of 0.1-1.0 μm range was investigated. The soft X-ray charger can adjust the bipolar ion concentration at will, and thus, low ion concentration conditions for generating nearly singly charged particles in the overall 0.1-1.0 μm range can be simply prepared. Through the proper control of the bipolar ion concentration in the soft X-ray charger, aerosols of good monodispersity could be generated under a variety of particle size and concentration conditions. To our regret, ion number concentrations of the soft X-ray charger as a function of various X-ray powers could not be measured owing to the limit of our system. If the ion concentrations were known with the X-ray power in our system, it would become easier to select the conditions for generating moniodsperse particles in the 0.1-1.0 μm range using this technique.

ACKNOWLEDGMENT

The authors thank Mr. Junichi Nakahara (Graduate School of Engineering, Hiroshima University) for helping the experiment. This work was supported in part by the New Energy and Industrial Technology Development Organization (NEDO)'s “Nanotechnology Materials Program - Nanotechnology Particle Project” based on funds provided by the Ministry of Economy, Trade and Industry, Japan (METI). And Grant-in-Aids for Scientific Research by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT) are gratefully acknowledged. This work was also supported in part by the Creative Research Initiatives from the Korean Ministry of Science and Technology (Center for Nano Particle Control). B. Han thanks the Brain Korea 21 (BK 21) project for financial support.

NOMENCLATURE

D_{p} = particle diameter [μm]

D_{pp} = preset particle diameter of DMA [μm]

e = elementary electrical charge ($=1.6021\times10^{-19}$) [C]

I_0 = saturation current of bipolar ions in the charger [A]

n = number concentration of particle [particles/m^3]

n_{i} = number concentration of positive ion (or negative ion) [ions/m^3]

S = production rate of bipolar ions in unit volume [ions/(m$^3\cdot$s)]

t = time [s]

t_{r} = charging time [s]

V = effective volume of the charger [m^3]

Z_{p} = electrical mobility of particle [m^2/(V$\cdot$s)]

α = recombination constant of bipolar ions ($=1.6\times10^{-12}$) [m^3/s]

β = combination probability between ion and particle [m^3/s]

Subscripts

p = carrying p elementary unit charges

T = total

0 = uncharged

1 = singly charged

2 = doubly charged

Superscripts

$+$ = positive

- = negative

Reference

[1] F. J. Romay-Novas, D. Y. H. Pui, Generation of monodisperse aerosols in the 0.1-1.0 μm diameter range using a mobility classification-inertial impaction technique, Aerosol Sci. Technol. 9 (1988) 123-131.

[2] A. Gupta, P. H. McMurry, A device for generating singly charged particles in the 0.1-1.0 mm diameter range, *Aerosol Sci. Technol.* 10 (1989) 451-462.

[3] M. Shimada, B. Han, K. Okuyama, Y. Otani, Bipolar charging of aerosol nanoparticles by a soft X-ray photoionizer, *J. Chem. Eng. Jpn.* 35 (2002) 786-793.

[4] B. Han, M. Shimada, M. Choi, K. Okuyama, Unipolar charging of nanosized aerosol particles using soft X-ray photoionization, *Aerosol Sci. Technol.* (2002) accepted.

[5] T. Seto, T. Nakamoto, K. Okuyama, M. Adachi, Y. Kuga, K. Takeuchi, Size distribution measurement of nanometer-sized aerosol particles using DMA under low-pressure conditions, *J. Aerosol Sci.* 28 (1997) 193-206.

[6] J. Bricard, *The Problem of Atmospheric and Space Electricity* (S. C. Coroniti, ed.). Elsevier, Amsterdam, (1965) 82-117.

[7] N. A. Fuchs, On the stationary charge distribution on aerosol particles in a bipolar ionic atmosphere, *Geofis. Pura Appl.* 56 (1963) 185-193

[8] M. Shimada, S.-J. Cho, K. Okuyama, T. Tamura, M. Adachi, T. Fujii, Removal of submicron aerosol particles by a tubular particle-removal device using UV/photoelectron method, *J. Aerosol Sci.* 28 (1997) 649-661.

[9] M. Shimada, K. Okuyama, Y. Inoue, M. Adachi, T. Fujii, Removal of airborne particles by a device using UV/photoelectron method under reduced pressure conditions, *J. Aerosol Sci.* 30 (1999) 341-353.

[10] F. Iskandar, I. W. Lenggoro, T.-O. Kim, N. Nakao, M. Shimada, K. Okuyama,

Fabrication and characterization of SiO_2 particles generated by spray methods for standards aerosol, *J. Chem. Eng. Jpn.* 34 (2001) 1285-1292.

[11] B. Y. H. Liu, D. Y. H. Pui, Electrical neutralization of aerosols, *J. Aerosol Sci.* 5 (1974) 465-472.

Table 1. Experimental conditions used in the spray drying method

Precursor	Nanocolloidal SiO_2
Sol concentration	0.01-0.1 M
Sol particle diameter	5 nm
Frequency of the ultrasonic nebulizer	1.7 MHz
Furnace temperature	300 °C
Carrier gas flow rate	1 *l*/min

Table 2. Estimated bipolar ion concentration generated by the soft X-ray charger with a variety of X-ray power

Indicated X-ray power	Saturation current, I_0	Ion concentration, n_i [ions/m^3]
200.0 μA, 10.00 kV[a]	40 nA	3.1×10^{13}
200.0 μA, 5.00 kV	3.9 nA	9.6×10^{12}
100.0 μA, 5.00 kV	2.0 nA	6.9×10^{12}
30.0 μA, 3.20 kV	< 0.1 pA[b]	$< 4.9\times10^{10}$
30.0 μA, 3.00 kV	< 0.1 pA[b]	$< 4.9\times10^{10}$
15.0 μA, 3.00 kV	< 0.1 pA[b]	$< 4.9\times10^{10}$

[a] Maximum for both current and voltage

[b] Saturation currents could not be measured owing to current noises. 0.1 pA indicates the order of maximum noise current.

FIGURE CAPTIONS

Figure 1. Theoretical estimation of the single and double charge ratios, n_1/n_T, n_2/n_1, with the particle diameter for different (a) ion concentrations, n_i (b) particle concentrations, n_T and (c) charging times, t_r. Thick lines in (a) indicate equilibrium charging state.

Figure 2. Schematics of the power-adustable soft X-ray charger.

Figure 3. Experimental setup for generating nearly singly charged aerosols using the soft X-ray charger and DMA: (a) polystyrene latex (PSL) particles, and (b) polydisperse SiO_2 particles generated by a spray drying method.

Figure 4. Ion currents versus voltage applied to the charger with a variety of soft X-ray power.

Figure 5. Charge distributions of PSL particles of (a) 0.207 μm and (b) 0.791 μm diameter under maximum power and low power conditions of the soft X-ray charger. +1~+5 indicate the number of charge units on the PSL particles. Some residue particles originated from Latex suspension were also detected at the high mobility parts of the graphs.

Figure 6. Change in charge ratios, n_2/n_1 and n_1/n_T, for PSL particles classified by DMA with particle diameter under a variety of ion concentration conditions.

Figure 7. Scanning electron micrographs of particles classified by the soft X-ray charger and DMA from polydisperse SiO_2 particles: (a) D_{pp} = 300 nm and maximum power, (b) D_{pp} = 300 nm and I = 30.0 μA, V = 3.00 kV, (c) D_{pp} = 600 nm and maximum power and (d) D_{pp} = 600 nm and I = 15.0 μA, V = 3.00 kV.

Figure 8. Size distributions of classified SiO_2 particles obtained from SEM photographs: (a) D_{pp} = 300 nm and maximum power, (b) D_{pp} = 300 nm and I = 30.0 μA, V =

3.00 kV, (c) $D_{pp} = 600$ nm and maximum power and (d) $D_{pp} = 600$ nm and $I =$ 15.0 μA, $V = 3.00$ kV.

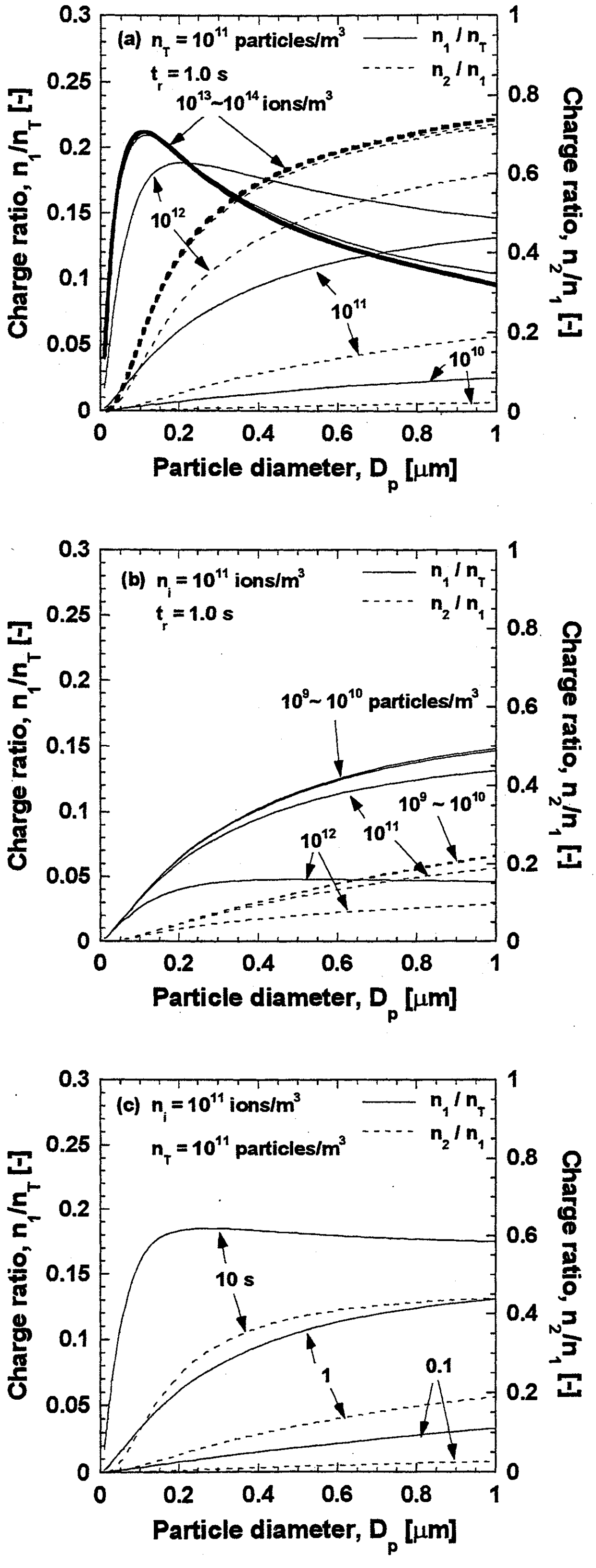


Figure 1. Han et al.

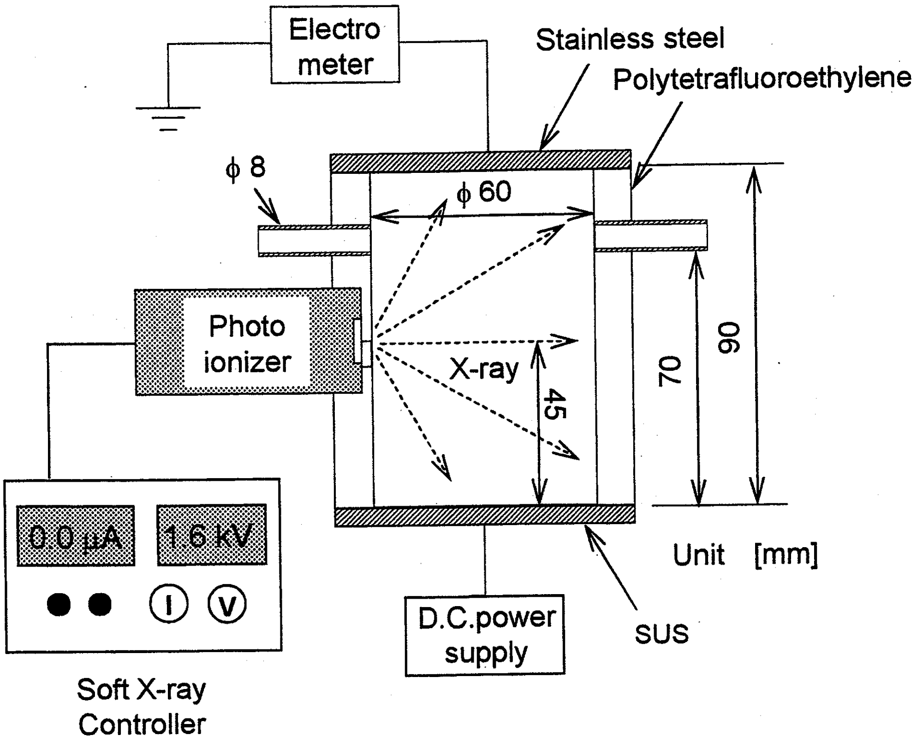
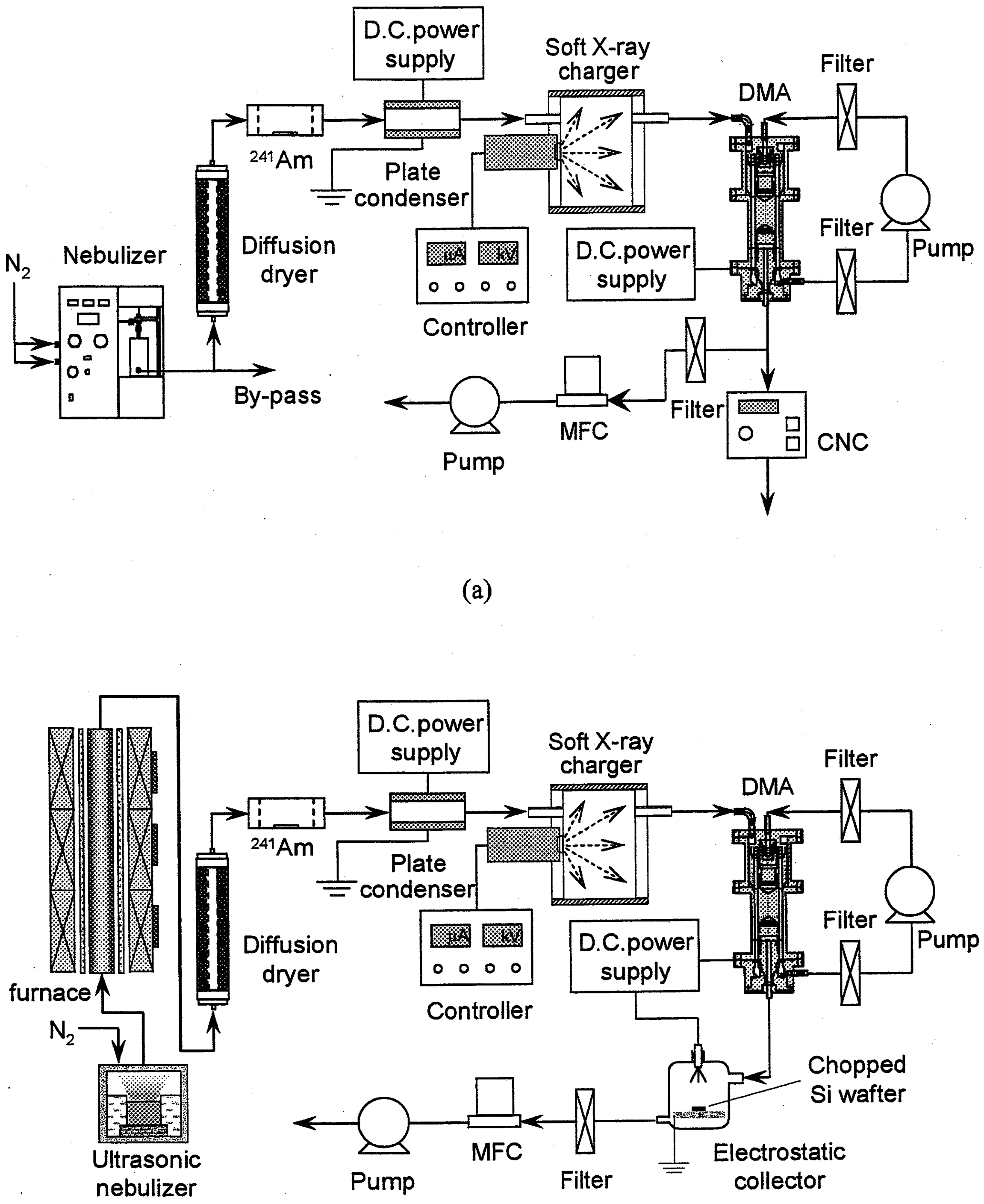


Figure 2. Han et al.



(a)

(b)

Figure 3. Han et al.

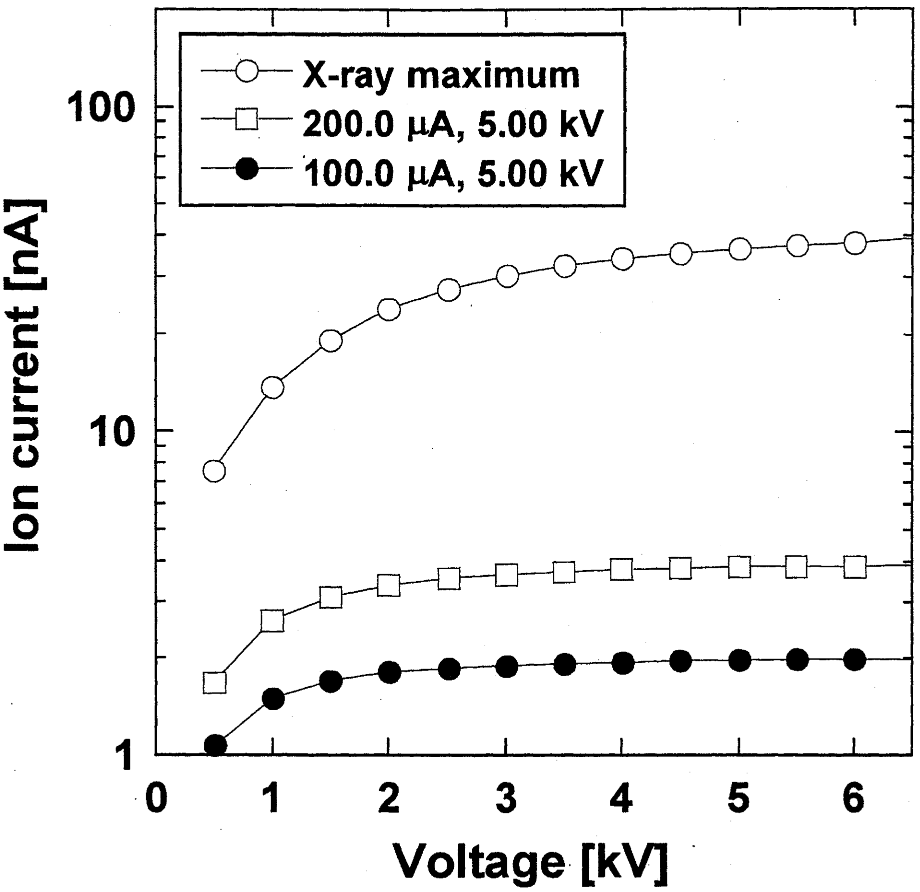


Figure 4. Han et al.

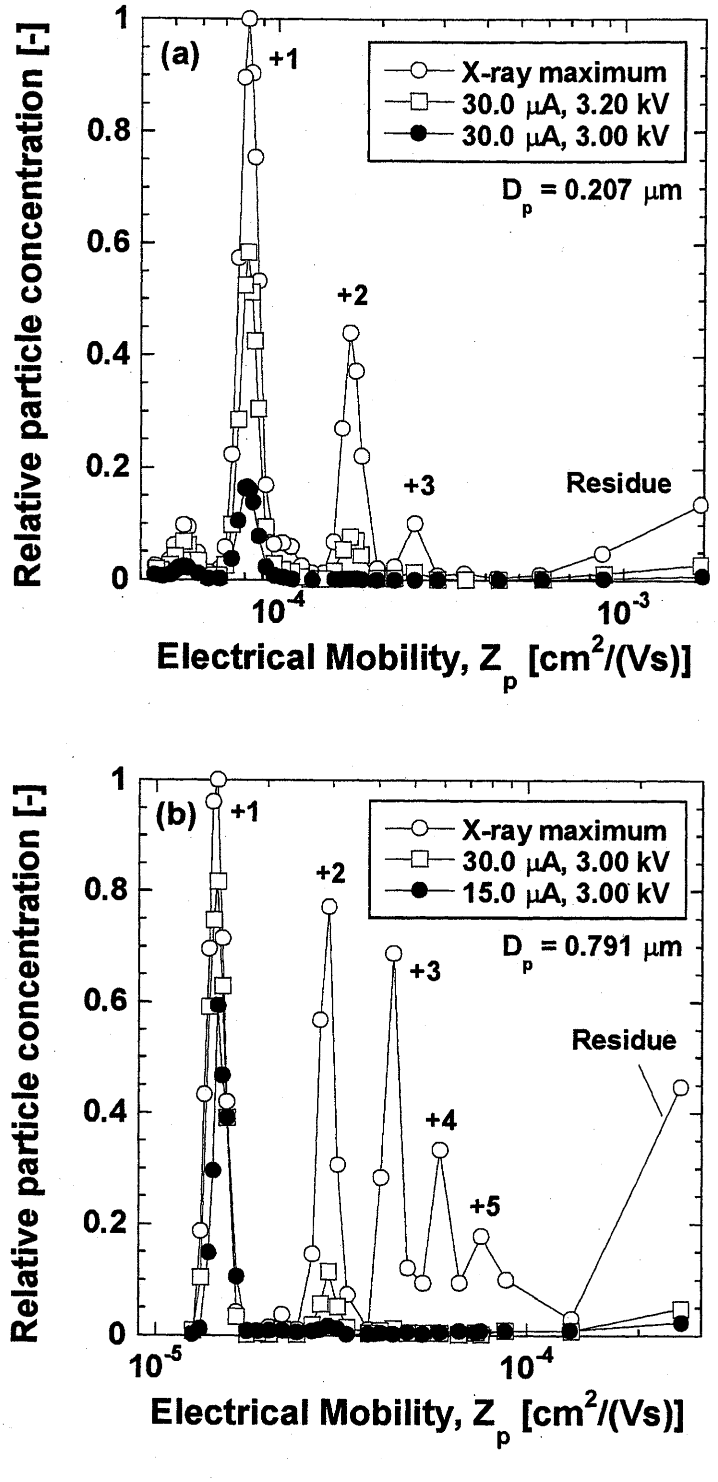
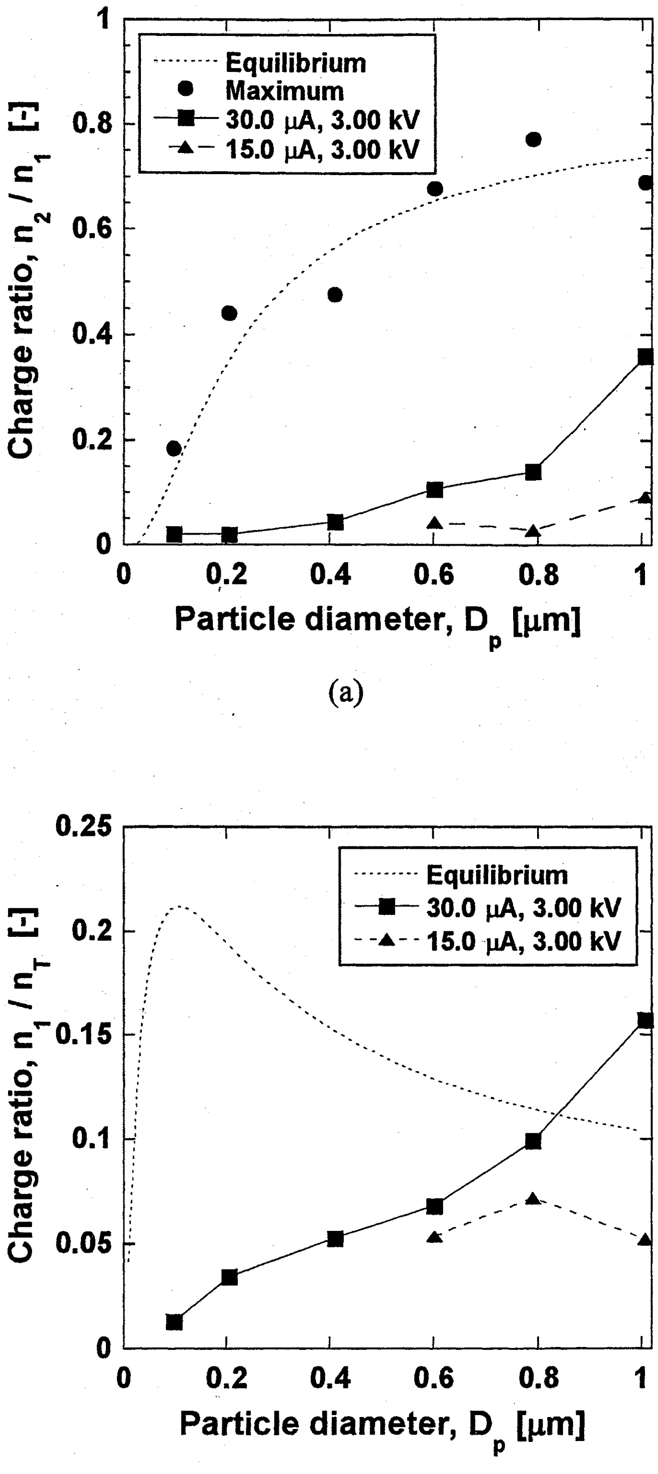


Figure 5. Han et al.



(a)

(b)

Figure. 6 Han et al.

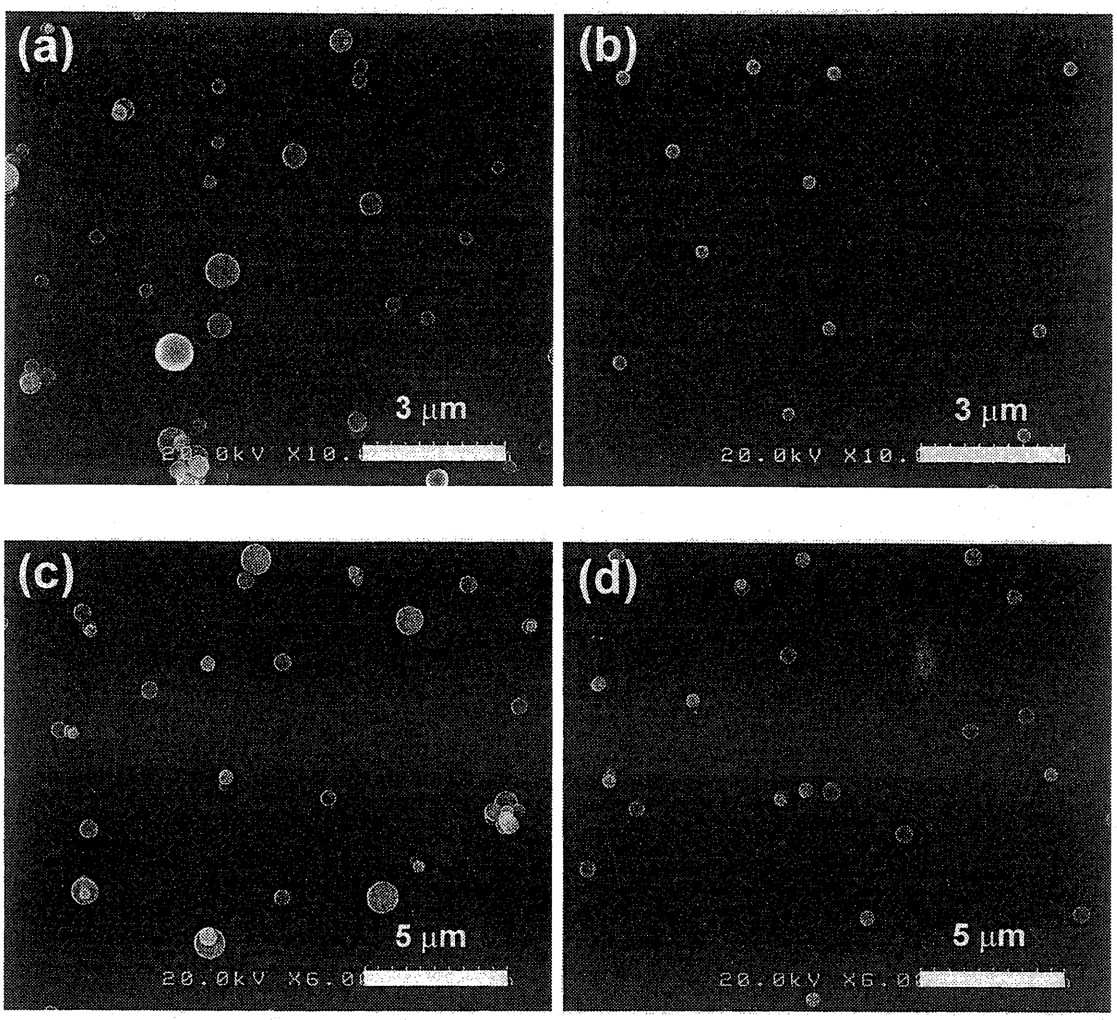


Figure. 7 Han et al.

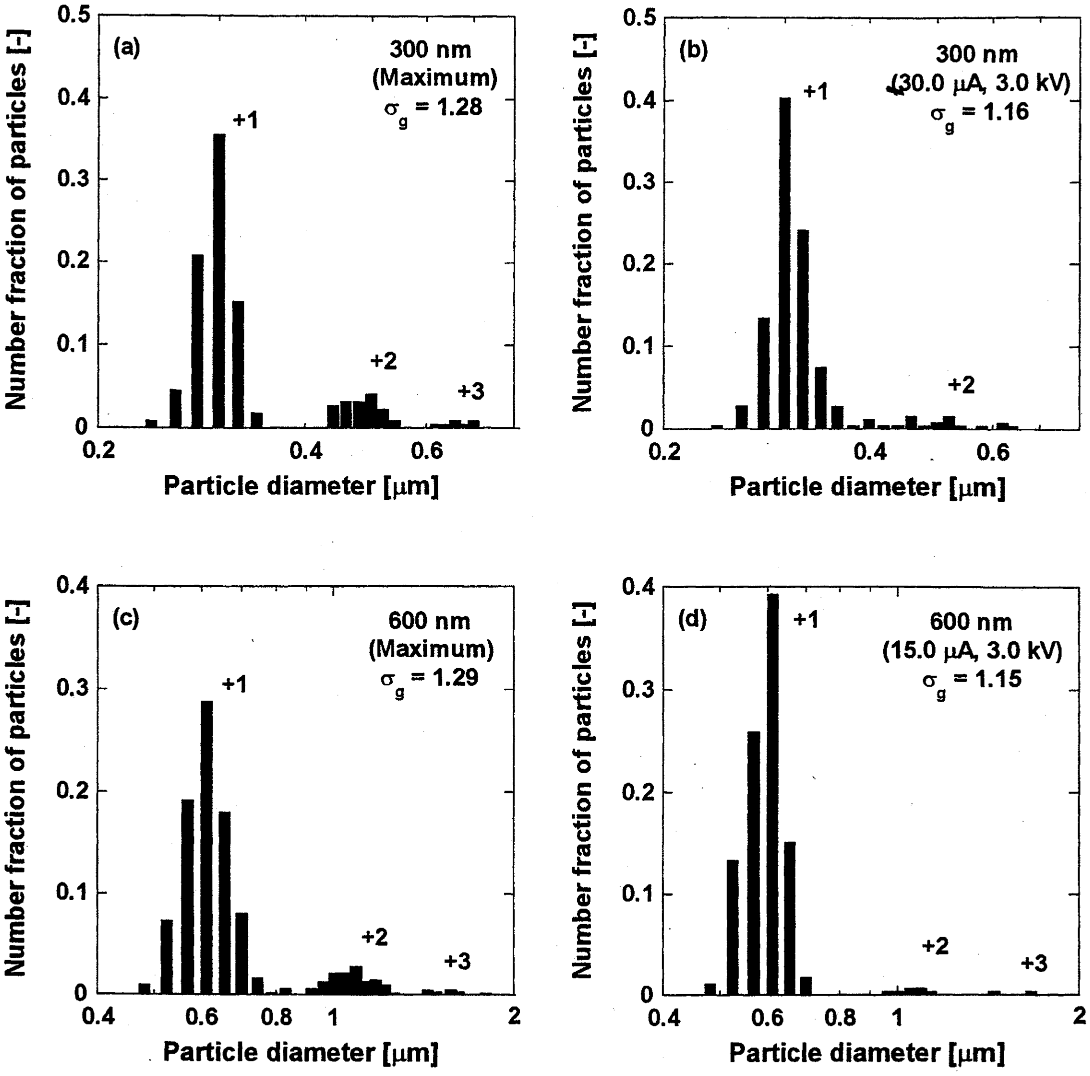


Figure. 8 Han et al.