

Charge Distribution of Fine and Ultrafine NaCl Aerosol Particles by Tandem DMA Technique

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ABSTRACT: In this paper, charge distribution of NaCl aerosol particles having a diameter of 54-292 nm has been obtained using Tandem Differential Mobility Analyser (TDMA) technique. Because of experimental constraints, the studies were limited to estimation of the negatively charged fraction up to three electronic units. The non-neutralised charged fractions for different sizes were compared with that of neutralised NaCl aerosol particles. It was observed that 19-21% particles are singly charged, 15-18% particles are doubly charged and 7-13% particles are triply charged. The charged fractions for other than singly charged particles were higher for non-neutralised particles as compared to that of neutralised particles.

KEYWORDS: Aerosol, Electrical mobility, Differential Mobility Analyser (DMA), Neutralisation, Nebulisation.

I. INTRODUCTION

Electrically charged aerosol particles have significant implications in the field of aerosol science and technology. Electrostatic properties of these particles affect aerosol sampling and transport [1], deposition of respirable particles in the human respiratory system [2], and particle collection characteristics of air filters [3]. To investigate the electrical effects in aerosol studies, information is needed about the magnitude and distribution of particle charge. Researchers have developed several approaches and charge analysers (integral, differential etc.) to measure the particle charge distribution. Integral mobility techniques have also been developed by Hurd [4]; Adachi et al. [5], to provide the magnitude of the charge of solid and liquid particles. However, a disadvantage of the integral mobility technique is its inability to provide the polarity of the particle charge. Buckley et al. [6] presented a technique for rapid estimation of the charge distribution of submicron aerosols under atmospheric conditions, using two DMA systems in parallel. This method involves measurement of aerosol size and mobility distribution in independent manner. Forsyth et al. [7] developed a charge analyser system to measure absolute charge distribution of commonly generated laboratory aerosols. Their study presents the absolute average charge of NaCl aerosols of more than 150 nm in diameter. By using a relationship of aerosol charge and diameter [8], a predicted mobility distribution was obtained from the size distribution measurement. The charge distribution was estimated by altering the parameters in the expression until the fit between the predicted and observed mobility distribution was optimized. However, limitation of a-prior knowledge of charging model restricts usage of this technique for several practical cases.

Another approach involves the use of a Differential Mobility Analyser (DMA) coupled to an Optical Aerosol Spectrometer to measure the charge distribution of particles [9]. However, the size detection limit of the optical aerosol

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spectrometer limited the range of measurement to relatively larger particle sizes (0.5-20 μ m). Moon [10] applied the tandem differential mobility analyser (TDMA) technique to monitor the evolution of charge distribution of combustion particles. However, because of rather low resolution of electrical aerosol analyzer used in their study, doubly and more highly charged particles were not accurately detected. Kim et al. [11] presented a method to measure the positive as well as negative charge distribution of nano-sized soot aerosol particles using a tandem differential mobility analyser system.

With increasing concern being shown for the toxicity associated with nano particles, emphasis has shifted towards the determination of particle size distributions in lower size ranges and hence analyzing their size related dynamics. Simultaneously growth of nano-aerosol technology requires generation of small particles of special materials using aerosol route. Nebulization is most commonly preferred technique for the generation of aerosol particles in fine and ultrafine size ranges. Significant amount charges are carried by aerosols generated due to nebulization [7]. This charge plays a crucial role in determining dynamics and depositional characteristics of nebulized aerosols.

In spite of, lot of available studies on integral charge measurements, a limited work has been carried out for polarity specific charge distribution on aerosol particles. Additionally, charge size distribution of nebulized aerosol particles in fine as well as ultrafine size ranges has not been studied in detail. In this work, controlled experiments have been performed using TDMA system. Measurement of singly, doubly and triply negative charged particles for broad size range (50-300 nm) has been performed under this work. The obtained results are then compared with standard Boltzmann charge distribution theory.

II. MATERIALS AND METHOD

A DMA operates by balancing the electrical force on a charged particle in an applied electric field, with the drag force on the particle as it moves through the fluid. DMA consists of a cylindrical central electrode and coaxially aligned cylindrical stainless steel housing. Filtered, dry air is supplied in the annular region around the central electrode, and an annular flow of sampled aerosol is introduced from the top of the DMA column. Aerosol is normally passed through a radioactive ionizing source, to give a known charge level to the particles. Aerosol particles enter the DMA in a thin annular ring adjacent to the outer cylinder, and the charged particles are attracted or repelled by the potential on the central rod. Near the end of the central rod is a slit through which particles of the desired electric mobility pass, while particles of higher electric mobility (smaller and/or more highly charged) hit the central rod upstream of the sample slit and particles of lower electric mobility (larger and/or uncharged) pass out of the DMA through the excess output air flow.

The TDMA consists of two differential mobility analysers (DMAs) and a condensation nuclei counter (CNC). Path (B) in Figure 1 shows the schematic of the TDMA system. In TDMA technique, DMA-1 is simply used as an electrical field classifying system, which is operated at a chosen fixed positive voltage to extract particles of equivalent electrical mobility without employing a radioactive ionizing source prior to the analyser. DMA-2 with CNC (SMPS) measures the size distribution of particles extracted from DMA-1. In our experiment, GRIMM DMA (Vienna type, Model-55-U, size range 10.25-1024 nm) without charge neutraliser has been used as DMA-1, While GRIMM DMA, Model 5.5-900 has been used for size classification (size range 9.8-878 nm). Due to non-provision of polarity switching in GRIMM SMPS (+ve voltage on central electrode), we could only measure negative charge distribution of aerosol particles. However the same technique can be applied for dual polarity cases.

The particle electrical mobility is defined as [12],

$$Z_p = \frac{neC_c(d_p)}{3\pi\mu d_p} \quad (1)$$

Where, n is the number of elementary units of charge = electronic charge, $C_c(d_p)$ is the Cunningham slip correction factor ($=1 + 2.514 \lambda/d_p + 0.8 \lambda/d_{ep} - 0.55 d_p/\lambda$), d_p = particle diameter, λ is the mean free path of air, and μ is the viscosity of air.

The centroid of the mobility band of the particles exited through central rod of DMA-1 at a fixed voltage is given by Knutson [13],

$$Z_p^* = \frac{(q_c + q_m)}{4\pi AV} \quad (2)$$

Where, q_c and q_m are sheath air and excess air flow rates respectively and $\Lambda=L/\ln(b/a)$, where L is the length of the central rod and a and b are the radius of the inner and outer cylinder respectively. From Equation (1) and (2) one can write

$$\frac{neC_c(d_p)}{d_p} = \frac{3\mu(q_c+q_m)}{4\Lambda} \frac{1}{V} \quad (3)$$

For a specific voltage applied to DMA-1, one can find out all the possible charge (n) and particle diameter (d_p) combinations of the extracted particles. When these selected particles of specific mobility are scanned through SMPS we get size spectrum showing peaks at those size channels corresponding to $n=-1,-2,-3$ etc. These size channels can be denoted by d_p^n and the number concentration of these particles are denoted by $N(d_p^n)$. If $N'(d_p)$ is the number concentration of generated aerosols as directly measured by SMPS, the fraction of particles carrying n unit of charge is estimated by,

$$f(d_p, n) = \frac{N(d_p^n)}{N'(d_p)} \quad (4)$$

III. EXPERIMENTAL PROCEDURE

The schematic of the experimental arrangement is shown in Figure 1. NaCl solution (0.1 % concentration) was used to produce the aerosol particles using BARC nebuliser [14]. These aerosols were dried using Nafion dryer and then passed through a Mixing Chamber to homogenize the aerosol concentration. Temperature and Relative Humidity (RH) level of the chamber have been monitored continuously and found to be in the range of 24-26 °C and 50-55 % respectively.

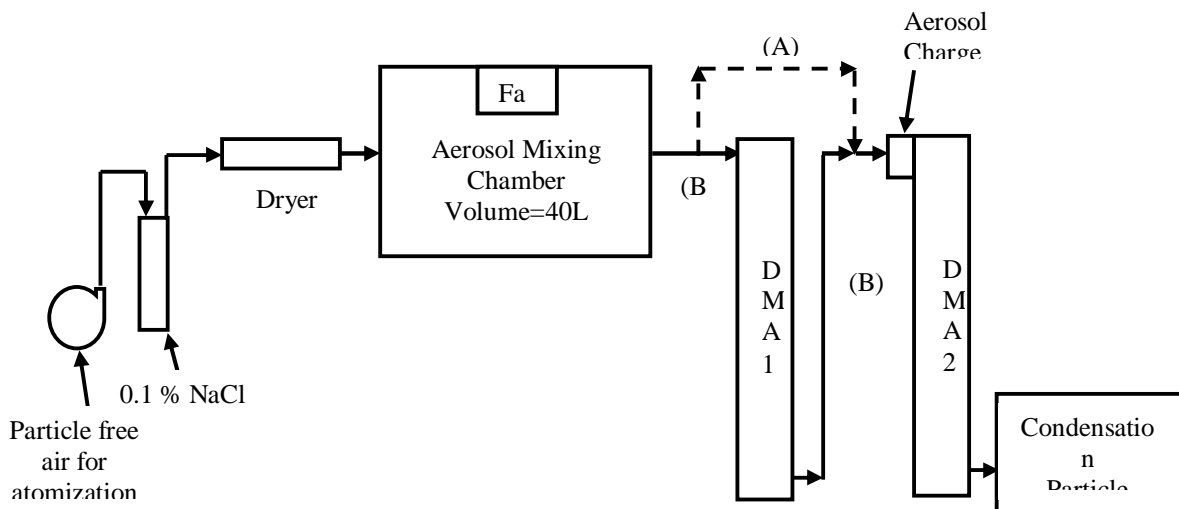


Fig.1. Experimental set up for measuring particle charge distribution

Path (A) in Figure 1 has been utilised to determine the size distribution of the generated aerosols and typical size distributions for 3 experiments are shown in Figure 2.

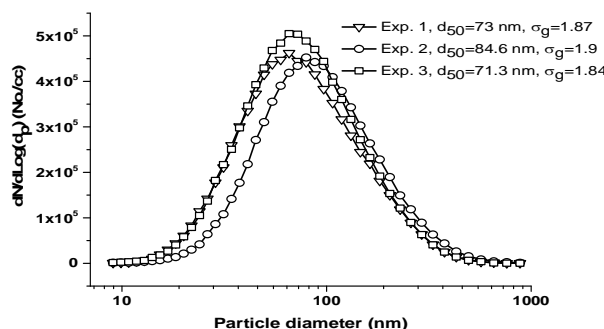


Fig. 2. Average number-Size distribution of nebulized NaCl aerosol particles

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These size distributions were obtained at different point of time during the experiment. The total particle number concentration was $\sim 10^5/cc$ having average median diameter (d_{50}) of 76.3 nm with an average Geometric Standard Deviation (σ_g) of 1.87. From Figure 2, it is observed that the size distributions are quite steady which is very essential for estimating charged fraction from the observed data.

The experimental set up was then switched to Path (B) of Figure1 for measuring charge distribution by Tandem DMA technique. Appropriate voltages were applied to DMA-1 and the size distribution of the selected mobility aerosols was measured by SMPS. Typical voltages applied to DMA-1, corresponding mobility and size-charge combinations of the particles which will be exiting from DMA-1 are given in Table1. As can be seen from the table at a fixed applied voltage to the DMA-1, the exit particles will have same electrical mobility corresponding to different size and charge combinations. As the applied voltage is increased the central mobility decreases which correspond to larger size particles will be exiting from DMA-1.

Table 1: Applied voltage to DMA-1 and corresponding mobility and size-charge combinations of transmitted particles

Applied voltage (-ve V)	Particle mobility ($m^2s^{-1}V^{-1}$)	Particle charge (electronic unit)	Particle diameter (nm)
120	8.16E-8	-1	54
		-2	79.2
		-3	99.9
238.9	4.1E-8	-1	79
		-2	118
		-3	151
404.8	2.42E-8	-1	107.04
		-2	163
		-3	212
1145	8.55E-9	-1	203.84
		-2	330
		-3	448.1
1364	7.18E-9	-1	228.98
		-2	375.5
		-3	514.5
1624	6.03E-9	-1	258.03
		-2	429
		-3	592.8
1932	5.07E-9	-1	291.75
		-2	491.5
		-3	684

Size distributions of transmitted particles at different voltages as measured by DMA-2 in combination with CPC are shown in Figure3. Diameter corresponding to singly charged particle (-ve)(d_p^{-1}) is also mentioned in this Figure. For a fixed applied voltage, the different peaks correspond to number concentration of singly, doubly and triply charged particles of respective size as shown in Table 1. First peak gives the singly charged particle concentration of particular mobility whereas later peaks represent multiply charged particle of higher sizes as compared to singly charged particle size. At lower applied voltages (e.g., 120 and 238.9 Volt) up to three peaks could be resolved, whereas at

higher applied voltages (1624 and 1932 Volt) peaks due to multiply charged particles could not be resolved as these peaks correspond to those particle sizes for which particle number concentration was statistically insignificant.

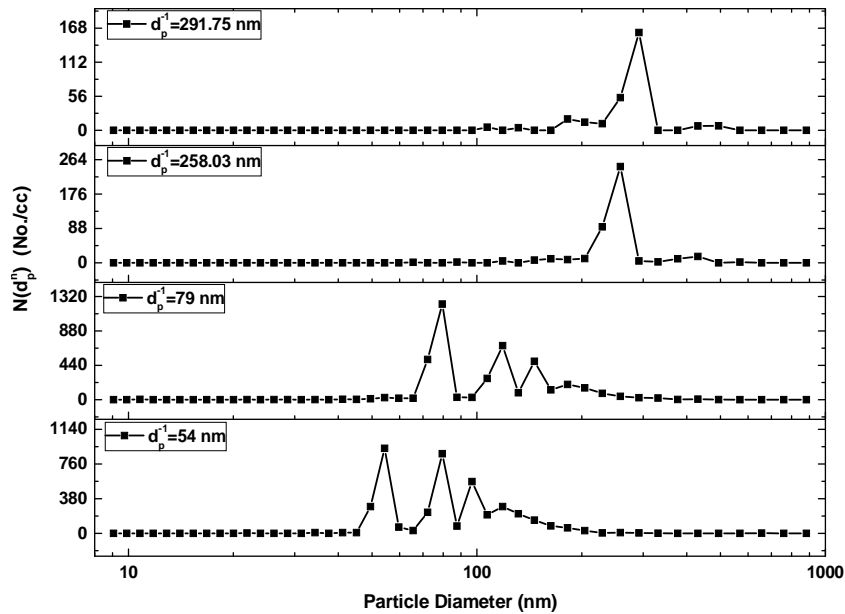


Fig.3. Size spectrum of transmitted particles through DMA 1 at different applied voltage

The size spectrum contains particle diameter for singly, doubly, triply and higher order charges as shown in Table 1. These higher order charges were not included in the calculation due to too low (statistically insignificant) particle concentration in those size ranges. The applied voltages were selected in such a way that we get singly, doubly and triply charged particle of all selected diameters. Now $\frac{N(d_p^n)}{N(d_p)}$ was calculated to obtain the negatively singly charged fraction for particle diameter d_p . There could be losses of particles in the pipes (polyurethane) used for connections. These could be diffusional sampling losses and increased depositional losses due to charge. As the experimental set-up involved different paths (path A and path B) for size distribution measurement and charge distribution estimation, accounting of loss fraction in longer path is important. To estimate the loss fraction, separate experiment was carried out by measuring the charged fraction of NaCl aerosol particles when they are passed through radioactive source before sampling through DMA-1(path B of Fig. 1).

For neutralized aerosol particles the fraction of particles carrying up to two elementary charges was estimated by,

$$f(d_p, n) = 10^{\left[\sum_{i=0}^5 a_i(n) \cdot \left(\log \frac{d_p}{nm} \right)^i \right]} \quad (5)$$

The approximation co-efficient $a_i(n)$ was taken from the work of Widensohler[15]. The fraction of particles with three or more elementary charge units is given by Gunn [16],

$$f(d_p, n) = \frac{e}{\sqrt{4\pi^2 \epsilon_0 d_p}} \exp \left[- \frac{\left[n - \frac{2\pi \epsilon_0 d_p k T}{e^2} \ln \left(\frac{c_{nl+} Z_{l+}}{c_{nl-} Z_{l-}} \right) \right]^2}{2 \frac{2\pi \epsilon_0 d_p k T}{e^2}} \right] \quad (6)$$

Where ϵ_0 = dielectric constant, k = Boltzmann's constant, T =temperature, $c_{nl\pm}$ =ion concentration and $Z_{l\pm}$ =ion mobility. Charged fraction for different sizes of neutralised NaCl aerosol particles was determined experimentally as explained above and was compared with theoretical calculation obtained from Equation (5) and (6). The results are shown in

Figure 4. The difference in the obtained fraction could be due to losses in pipes and systems when we switch over from Path (A) to Path (B). The percentage loss is also shown in Figure 4.

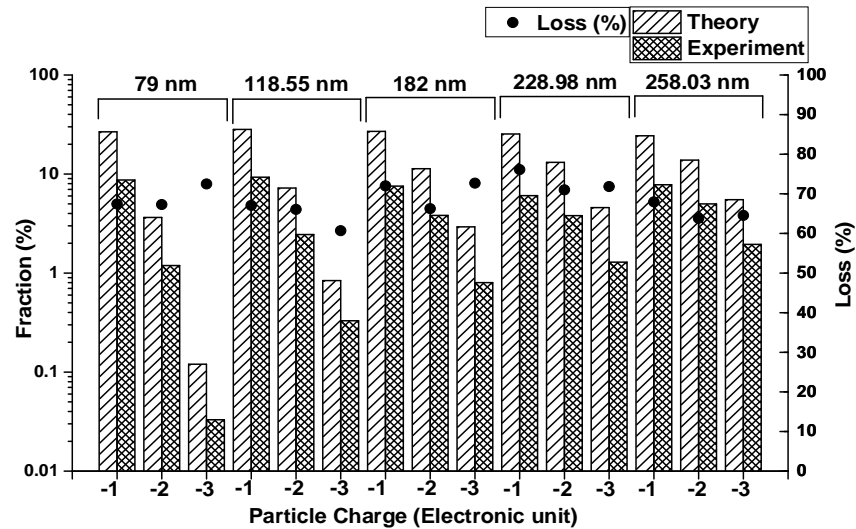


Fig.4. Theoretical and experimental charge fraction of Neutralised NaCl aerosol particles

Although the trend of charge distribution matched very well with the theoretical calculations, we observed that experimental fractions are less as compared to theoretical fractions indicating an average ‘loss factor’ of 0.70. This was attributed to the losses in the systems and piping used for experiments. Losses due to diffusion were estimated to be a very small (1-5%). Major losses were expected due to deposition of charged particles as evident from the work of Chen [9]. His work reported that the deposition in the pipe will increase as charge to mass ratio increases. In the present study the charge to mass ratio was in the range of 10^{-3} to 10^{-5} C/gm which will result in high deposition in the pipes. This loss factor was considered for obtaining the charge distribution of non-neutralised NaCl aerosol particles.

IV. RESULTS AND DISCUSSION

Taking into consideration of the ‘loss factor’, the charge distribution of non-neutralised NaCl particles for few sizes were measured and shown in Table 2.

Table 2. Comparison of charged fraction of neutralised (theoretical) and non-neutralised NaCl aerosol particles

Particle diameter (nm)	Fraction (%)					
	-1 unit charge		-2 unit charge		-3 unit charge	
	Non-neutralised (Experimental)	Neutralised (Theoretical)	Non-neutralised (Experimental)	Neutralised (Theoretical)	Non-neutralised (Experimental)	Neutralised (Theoretical)
54	18.99	23.18	14.60	1.43	6.78	0.01
79	20.64	26.73	14.73	3.64	6.82	0.12
107.04	20.87	28.11	15.08	6.24	8.72	0.56
203.84	20.09	26.28	15.79	12.26	12.24	3.72
228.98	19.38	25.39	14.89	13.11	7.75	4.58

258.03	21.90	24.36	17.41	13.82	13.24	5.50
291.75	21.74	23.24	17.64	14.40	9.75	6.46

The fraction for neutralised aerosols has also been shown for comparison purpose. As can be seen from Table 2, the non-neutralised fractions are less for singly charged particles whereas for doubly and triply charged particles, the non-neutral fractions are more as compared to neutral fractions.

Figure5 gives the graphical representation of obtained charge distribution along with theoretical Boltzmann’s charge distribution as obtained using Eq.5 and 6.

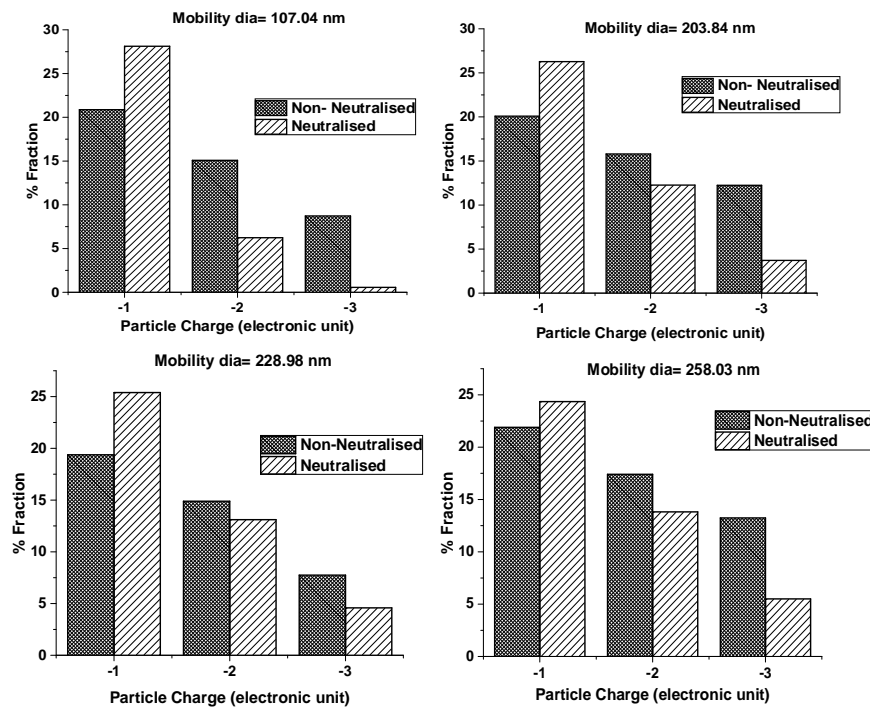


Fig.5. Comparison of charge distribution of neutralised (theoretical) and non-neutralised NaCl aerosol particles

It could be seen from Figure 5, as the number of charges increases (more than 1), ratio of charged fraction of non-neutralised aerosols to that of neutralised fraction also increases. As the amount of charge increases the fraction decreases for all the sizes. It is observed that around 20% particles are singly charged, 15-17 % particles are doubly charged and 7-13 % particles are triply charged.

V. CONCLUSIONS

The charged fraction of neutralised aerosols was computed and the same was experimentally determined to calibrate the TDMA measurement technique. Using this technique Charge distribution of non-neutralised NaCl aerosol particles was determined. Experimentally obtained charge distribution of non-neutralised aerosol was compared with that of neutralised aerosol particles. For the studied range of size it was observed that 19-21% particles are singly charged, 15-18% particles are doubly charged and 7-13% particles are triply charged. The data could be resolved for up to 3 units of charges because of the aerosol size distribution characteristics wherein the particle number concentrations were sufficient (~ 5000 No./cc) for particle size range of 50-300 nm. This polarity specific charge size fraction of nebulised aerosols will play vital role in understanding the aerosol aerodynamic behaviour e.g. deposition in the human airways system.

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