



# Area-selective deposition of charged particles derived from colloidal aerosol droplets on a surface with different hydrophilic levels

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## ABSTRACT

The effect of chemical treatment of a metallic substrate on the deposition behavior of charged aerosol particles derived from spraying droplets was investigated. A single substrate with areas having different hydrophilic levels was prepared as target surface. The treated (i.e., higher hydrophilic level) area, measured using a surface potential meter, showed a higher negative potential. A numerical simulation predicted that positively charged aerosol particles tended to approach and were subsequently immobilized on the high hydrophilic area. The area with higher hydrophilicity could collect particles with higher number concentration (density) than the other areas. The relationships were demonstrated in (i) the electrostatic surface potential, (ii) the hydrophilicity of surface, and (iii) the enhancement of adhesive force between the deposited particles and the surface.

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## 1. Introduction

Particles suspended in the gas phase as charged aerosols have been regarded as an effective route for transferring nanoparticles from the source (e.g., colloidal suspension) to a solid substrate at high deposition efficiency, without coagulation of particles (Jaworek & Sobczyk, 2008). Deposition of aerosol particles for producing nano-structured materials has been widely used in several aspects, such as biology (Kim et al., 2008), microelectronics (Deng & Gomez, 2011), biomolecular technology (Ranganath et al., 2011), environmental health applications (Jung et al., 2011), and pharmaceutical particle fabrication (Rezvanpour et al., 2012). Even though the relationship between the conditions of the substrate and the deposition mechanism of the charged aerosol particles is also important, there are few uncertainties about this topic. The surface conditions of a substrate can be modified by changing its physical and chemical properties. Barry et al. (2003) reported the electrostatic driven self-assembly of charged nanoparticles onto the charged surface, where it offered a more efficient method compared with other techniques. The charged areas were generated by a technique which had used

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a flexible and conductive electrode to pattern electrons and holes in a thin-film electret. Xie et al. (2010) investigated a single step to generate polymeric particles with patterns of dots, circles, squares, and bands on the substrate using deposition of charged aerosol particles through a mask.

The physical effect of electrically patterned surface on the deposition of aerosol particles had been investigated by Krinke et al. (2001) and Lenggono et al. (2006). The particles were attracted to a desired position due to the electrostatic force of particles and the 'patterned' area. The charged particles could also be deposited onto a surface patterned with a photoresist controlled by the introduction of ions having the same polarity together with the charged aerosol particles (Kim et al., 2006). The ions accumulated on the photoresist modified the applied field, in order to focus the nanoparticles onto the exposed parts of the patterned surface. Furthermore, Lee et al. (2011) studied patterning films of metal nanoparticles based on plasma electrochemical reduction. This approach provided fabrication for transparent, flexible and patterned films of metal nanoparticles. Our group reported for the first time the relationship between the chemical condition of the surface and the efficiency of aerosol deposition, using an aluminum substrate treated by sodium hydroxide and phosphoric acid (Naim et al., 2010). In our previous studies, the whole substrate was chemically treated. Even though an untreated substrate had a lower number concentration of the deposited particle than that of the treated one, the deposition mechanism was still uncertain. Meanwhile, the present study used phosphoric acid as an agent to create an area of the substrate which is chemically different. To understand better about particle deposition, a half area of the substrate was treated, whereas the other area was left untreated. The research hypothesis is that the chemical treatment by phosphoric acid will alter the surface at the molecular level, under the condition that enables the formation of the surface with different hydrophilicity. Furthermore, the hydrophilicity may also change the surface electric potential. In this case, we assume relationship between chemical treatment, hydrophilicity, and surface electric potential.

Jacobs and Stemmer (1999) measured the surface electric potential distribution using atomic force microscopy (AFM) and Kelvin probe force microscopy (KFM). They investigated the potential maps of composite metal and semiconductor films, by which they differentiated the materials with a resolution of a few nm. The surface charges were detected as an image of electric surface potential. Meanwhile, Ozasa et al. (2008) made direct measurement using KFM to observe the negative values of surface potential patterns on tris(8-hydroxyquinolino)aluminum(III) ( $\text{AlQ}_3$ ) thin film. The surface charges on a photoresist surface generated by corona discharge of  $\text{N}_2$  positive ions were also measured using KFM (Lee et al., 2009). In the measurements of KFM, a conductive probe is used to detect the electrostatic force generated by the potential difference between the sample surface and the probe. However, the surface electric potentials investigated in the previous studies were limited to the nanometer or micrometer scale.

Our present study uses the same principle of surface electric measurement in the KFM, but we have measured the surface electric potential in a larger area with a probe diameter of about 10 mm. The 'bulk' measurement using this technique has enabled measurement of the surface electric potential of the target (i.e., half area of the substrate). We have investigated the relationship between chemical treatment and the corresponding surface electric potential. The objective of our study is to investigate the relationship between the hydrophilic surfaces and the efficiency (or potential) of the surface to capture and immobilize the particles transported from the gas phase.

## 2. Experimental methods

### 2.1. Preparation of materials and substrates

Charged particles were suspended in the gas phase by the aerosolization of a suspension. The suspension was composed of a mixture of  $\text{SiO}_2$  (Snowtex, Nissan Chemical Industries Co. Ltd., Tokyo) and  $\text{AgNO}_3$  salt (Wako Pure Chemical Industries Co. Ltd., Tokyo). Firstly,  $\text{SiO}_2$  40 wt% with a mean diameter of 70–100 nm (primary particle, the value given by the maker) was dispersed in a solvent mixture of water:ethanol (75:25wt/wt) in order to make  $\text{SiO}_2$  of 0.1 wt%. A salt  $\text{AgNO}_3$  0.1 M, 0.3 wt% of total weight solution, was added into the  $\text{SiO}_2$  suspension in order to amplify the possibility of detection using the X-ray fluorescence method (XRF, JSX 3100 RII, JEOL, Tokyo), because the heavier element (Ag) is more easily detectable than Si.  $\text{H}_3\text{PO}_4$  (Wako Pure Chemical Industries) was used for substrate treatment by dilution into a ultra-pure water sample to obtain 0.1 M in concentration. All chemicals were used as received without further purification. Particle-size distributions in the liquid phase were measured using an analysis of dynamic light scattering (DLS) (HPPS 5001, Malvern Instrument, Worcestershire), and the average particle diameter was 623 nm. The conductivity and pH of the suspension were measured by a conductivity meter (CM-21P, TOA-DKK, Tokyo) and a pH meter (Orion 920Aplus, Thermo Electron Corporation, Tokyo) at 25 °C and the results were  $32.3 \times 10^{-3}$  S/m and 2.96, respectively. The suspension was ultrasonicated (35 kHz) within 15 min in order to prevent fouling on the wall surface of the container and further aggregation during storage.

An aluminum sheet with 25 mm diameter and 0.02 mm thickness was used as a model substrate. The substrates were treated in the following sequence steps: (i) submerged about half of the substrate (12.5 mm of height) into  $\text{H}_3\text{PO}_4$  solution for 10 min, (ii) submerged with ultra-pure water for 10 min, and (iii) dried in an oven at 120 °C for 1 h. The treated and dried substrates were kept in a desiccator before the deposition experiments. It was important to use a single aerosol generator for a single experiment in order to investigate the other parameters of a substrate, such as a treated or untreated area. The surface electric potential values of the treated and untreated areas (before the deposition process) were measured by a non-contact surface potential meter (Model 323, Trek, New York) as  $-590$  and  $-260$  mV, respectively. The size (diameter) of the probe was 10 mm, which covered the measured surface. The chemical elements of the substrate, analyzed before the

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