# Simultaneous removal of nano-sized contaminant particles with multi-modal size distribution using hybrid particle beams

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## ABSTRACT

The manufacturing yield of submicron-scale devises can be reduced by particulate contamination of their surfaces. Cryogenic aerosol beam using micron-sized aerosol particles has long been successfully used to remove contaminant particles down to 50 nm, and supersonic particle beam using particles smaller than 100nm lowered the limit of cleaning down to 20 nm size.

In this study, an improved nano particle beam technique was developed, where a mixture of bullet particles of two different sizes were used instead of mono-disperse bullet particles. Cleaning performance was sensitive to the bullet size and velocity. Small bullet particles selectively removed the 10 ~ 20 nm contaminant particles, and big bullet particles worked on the contaminant particles in the range of 30 nm or larger. When two different bullet particles were used sequentially, we obtained cleaning efficiency about 99 % for 10 ~ 100 nm $Al_2O_3$, which was the first ever report of a successful removal of contaminant particles in the 10 nm range with a wide size distribution.

***Keywords***: hybrid particle beam, $CO_2$, supersonic nozzle, gas-phase nucleation, cleaning efficiency

## 1 INTRODUCTION

The manufacturing yield of submicron-scale devices can be reduced by particulate contamination on their surfaces. Size of semiconductor device features are expected to decrease continuously, reaching 25 nm by 2015 for DRAM/FLASH memory device, and together with this decrease, critical killer particle size is expected to decrease to 12.5 nm [1]. In the nanometer range, the adhesion force per contact area between a particle and surface increases linearly as particle size decreases, but the fluid drag force changes in proportion to the cross-sectional area of a particle. Thus the used of drag force to remove contaminant particles (CPs) becomes less efficient as CP size decreases [2, 3].

One promising method of removing very small CPs is the cryogenic aerosol technique, in which the contaminated surface is bombarded by fine, high-velocity bullet particles (BPs) made of volatile material. CPs on the surface can be removed if the energy transferred from BPs to the CP is sufficient to overcome the adhesion energy between the CP and the substrate. In the conventional form of this technology, condensable gas is pre-cooled close to liquid nitrogen temperature and then expanded through a simple nozzle such as a cylindrical hole. Cooling liquefies some of the condensable gas, expansion of the gas through the nozzle atomizes the liquid into fine droplets, and adiabatic cooling causes solid particles to form during further expansion. Typical particle size is a few microns and typical velocity is 100 m/s. The minimum size of CP that can be removed efficiently using the micro-sized particle is 50 nm [4, 5].

Lee et al. showed that extremely tiny Ar bullet particles generated by gas phase nucleation could remove contaminant particles effectively, and other studies reported the applicability of Ar aerosol technique to nano contaminant cleaning [6-9]. One problem with the technique is that removing performance is very sensitive to the size of BPs and CPs with a broad size distribution cannot be removed with BPs of a single size.

This study thus aims to explore the possibility of using the hybrid bullet particles smaller than 100nm for removing nano-sized contaminants down to 10nm. An improved $CO_2$ particle beam technique was developed, where a mixture of BPs of different sizes were used instead of mono-disperse BPs. The nano-sized CPs were selectively removed by using BPs of two different sizes. BP size and velocity were varied by varying the nozzle contour, gas composition, and stagnation pressure. Cleaning experiments were done for a flat surface contaminated with a variety of particles of size down to 10 nm.

## 2 EXPERIMENT

The experimental system is schematically shown in Fig. 1. At room temperature and atmospheric pressure, the gas phase is the only stable phase, implying that the end product is always $CO_2$ gas, regardless of the initial phase. A condensable gas at room temperature was expanded directly to a vacuum through a supersonic Laval nozzle. During supersonic expansion through the nozzle, tiny condensation nuclei form and grow; their final size can be easily controlled by adjusting the stagnation pressure, the back pressure of the vacuum chamber, and the nozzle geometry. The BPs are formed entirely by homogeneous nucleation and growth downstream of the nozzle throat.

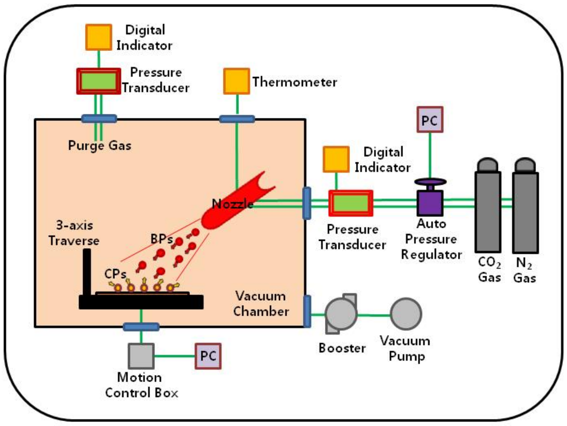



Figure 1: Schematic of the experimental setup.

To generate BPs travelling at the greatest possible supersonic velocity, $CO_2$/He mixture gas was used. Stagnation pressure was varied and stagnation temperature was kept at room temperature. The shape and size of the nozzle were adjusted to make the Mach number at the nozzle exit > 5.0 over the whole range of experimental conditions. The nozzle is nearly conical at the throat but rapidly expands near the exit to compensate for boundary layer growth and to prevent shock-wave formation.

In all trials, the CPs were 10-nm $Al_2O_3$ particles; they were generated by spin-coating a drop of $Al_2O_3$ solution onto a wafer. Cleaning was completed within 1~2 seconds of exposure in all the cases shown. Distance between the nozzle and the wafer and the angle of the particle beam were variable. The chamber pressure was also varied to guarantee a supersonic flow in the range of 1~20 torr.

## 3 RESULTS AND DISCUSSION

### 3.1 Characteristics of Ar and $CO_2$ BPs

Previously, it has been proven that 10 nm CP can be removed with 100% efficiency by using Ar particle beam [9]. Despite the above success, two practical difficulties with using Ar BPs remain: (1) BP size is very sensitive to temperature, so it must be controlled very accurately to achieve BP size uniformity. The dent of Ar BPs is shown in Fig. 2. The liquid atomization gives a much wider size distribution of Ar BPs. In low temperature technology, such temperature should be controlled accurately, which is difficult; (2) BP velocity is limited by low temperature. If the BP can be made at high temperature (room temperature), it would eliminate the need to control temperature and increase BP velocity. The resulting uniform, fast BPs should improve removal of nanometer CPs.

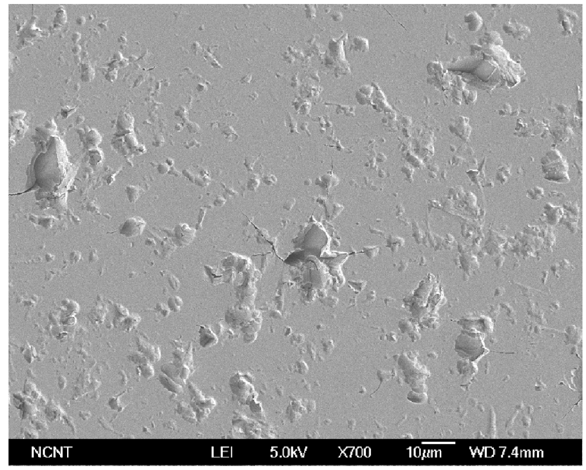



Figure 2: SEM image of dents on a photo resist film made by the micron-sized Ar BPs generated by atomization.

Carbon dioxide ($CO_2$) BP has above advantage. $CO_2$ gas of room temperature is used, then the temperature can be maintained constant easily, which facilitates the generation of uniform-sized BPs. Compared to previous studies based on atomization technique, nano-bullets generated in this study were smaller and had a narrow size distribution (Fig. 3). Size of the nano-bullets used in this study is on the same order of magnitude as that of the CPs.

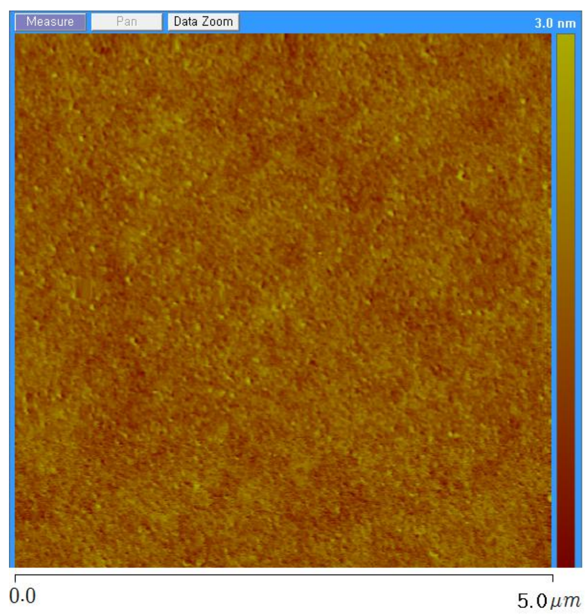
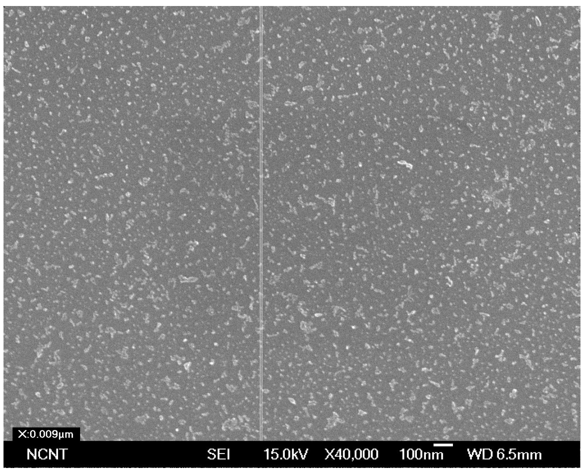



Figure 3: AFM image of dents on a photo resist film made by nano-particles generated by nucleation and growth with pure $CO_2$.

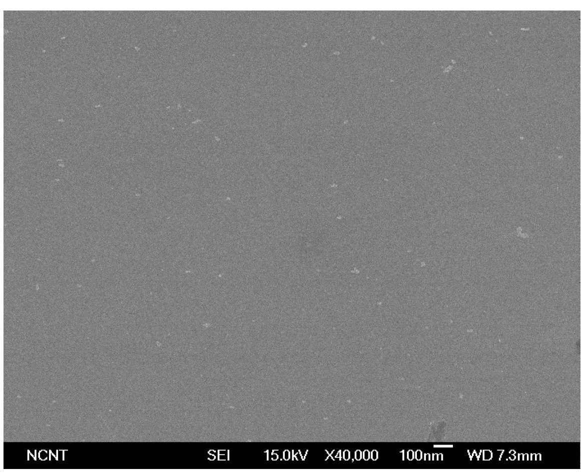
### 3.2 Cleaning with hybrid $CO_2$ particle beam

Removal was tried for 10 ~ 100 nm $Al_2O_3$ contaminant particle on a flat silicon wafer (Fig. 4a), where nano-bullets were generated with $CO_2$/He mixture gas at 40 bar. And BPs of different sizes were generated using nozzles of different contours in such a way that one nozzle generates big BPs in the range of 40~50 nm and the other nozzle small BPs in the range of 10 ~ 15 nm.

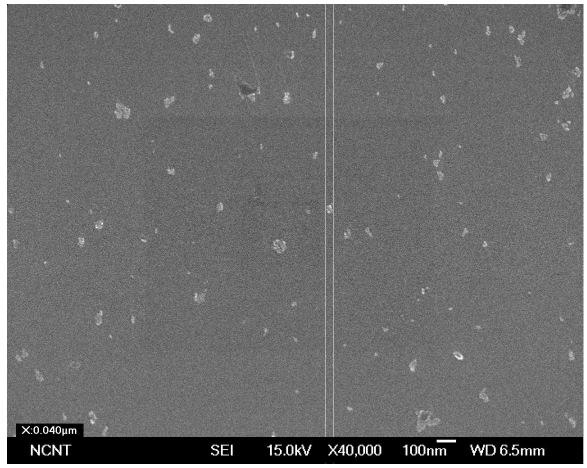
When the CPs were exposure to big BPs, 30 nm or lager CPs were removed but most 10-nm CPs remained (Fig. 4b). The small BPs removed 10-nm CPs almost completely, but most 30-nm or lager CPs remained (Fig. 4c). When we used the two different BPs sequentially, we obtained cleaning efficiency about 99 % for 10 ~ 100 nm $Al_2O_3$ CPs (Fig. 4d). These results imply that the size of the nano-bullets is the most important factor contributing to removal of nanometer-sized CPs from a substrate by collision with volatile nano-bullet. When we used appropriately uniform size and velocity of the $CO_2$ BPs that were obtained by optimizing the nozzle contours, the nano-sized CPs were perfectly controlled. And we can simultaneously remove CPs in the 10-nm range with a wide size distribution.



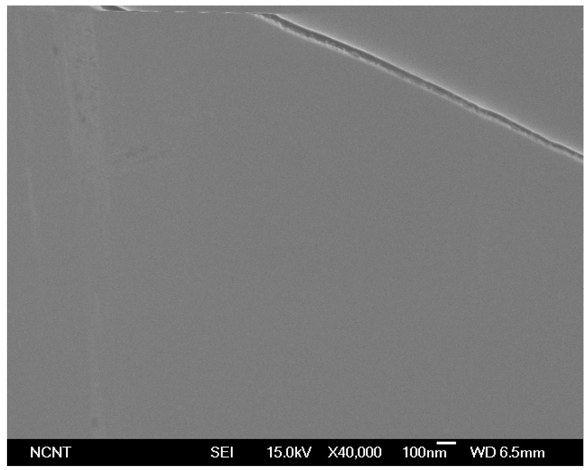

(a)




(b)




(c)




(d)

Figure 4: SEM images before (a) and after cleaning of $Al_2O_3$ particles on Si surface using (b) big $CO_2$ BP, (c) small $CO_2$ BP, and (d) hybrid $CO_2$ BP.

## 4 CONCLUSIONS

Nano-particles on a flat surface were cleaned by bombardment with supersonic $CO_2$ particle beam. Particle beams were generated by supersonic expansion through contoured Laval nozzles, such that the final particle velocity was in the supersonic range. By controlling the nozzle contour, $CO_2$ bullets of 1 ~ 50 nm size were generated by gas-phase nucleation and growth. Compared to Ar BPs generated by liquid atomization, $CO_2$ BPs were more uniform size and had higher velocity. It can be simultaneously remove for a various distribution of ceramic CPs down to 10 nm using the $CO_2$ hybrid particle beam.

## 5 ACKNOWLEDGMENT

This work was supported by the Korea Science and Engineering Foundation (KOSEF) grants funded by the Korea government (MOST) (No.ROA-2008-000-20045-0).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