Article

Differential Removal of Nanoparticles on the Surface of a Thin Film Substrate

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3D AFM image of an acidic particle

Mechanism of differential removal of nanoparticles
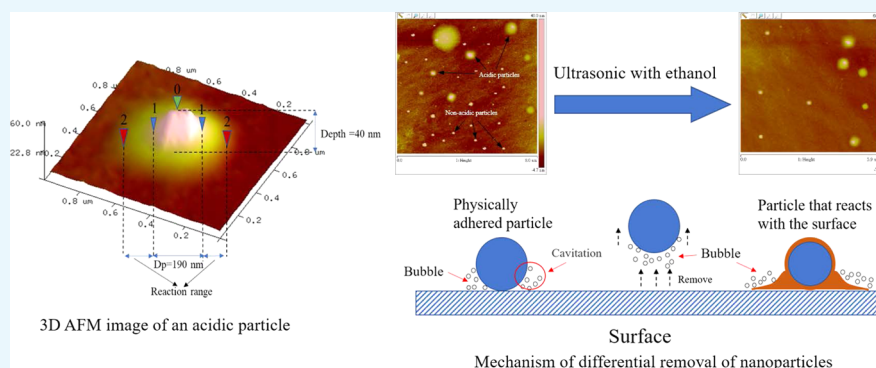
ABSTRACT: Purposeful identification, selection, and collection of particles are of great significance in environmental research. Microscopy is the common technique used in previous studies of particle identification. However, the microscopic technique was intricate and time-consuming. To conduct an intensive analysis of targeted particles, there is a need for the development of a simple method that can differentially abandon the nontargeted particles and only retain the targeted particles on the surface of a substrate. In the study, three methods were attempted for differential removal of nontargeted nanoparticles on the surface, including air jet, nanobubble, and ultrasonic methods. Acidic particles were taken as the targeted particles, while nonacidic particles were regarded as nontargeted particles. The results showed that regardless of methods, acidic particles were retained on the surface due to the strong particle−surface interaction. As for nonacidic particles, air jet treatment and nanobubble treatment were not able to completely remove nonacidic particles from the surface with the removal efficiencies of 5.1 ± 3.4 and $89.3 \pm 4.1\%$, respectively, while the nonacidic particles were entirely removed in the ultrasonic treatment. Ethanol rather than deionized (DI) water was the proper solution in the ultrasonic treatment to avoid contamination. In conclusion, ultrasonic by ethanol was fully efficient for differential removal of nonacidic particles on the surface. The principle of differential removal of particles is the differences in the particle−surface interaction force between nonacidic particles (i.e., physically attached particles) and acidic particles (i.e., chemically formed particles). Nonacidic particles are removed from the surface through cavitation to form bubbles in the gap between a nonacidic particle and the surface in the ultrasonic treatment. In contrast, the space between an acidic particle and the surface is filled by the reaction, and thus bubbles cannot enter the crevice to remove the acidic particle. The developed method is useful for aerosol research.

1. INTRODUCTION

Purposeful identification, selection, and collection of particles are essential and common in environmental studies.[1] The analysis of targeted particles can meaningfully minimize the downstream efforts on preparation for environmental monitoring and/or analysis and improve the signal quality. Previous particle identification and selection were mainly accomplished with the aid of the microscopic technique. Wang et al.[2] used an atomic force microscope (AFM) to identify and quantify acidic ultrafine particles in the atmosphere by scanning the surface of a detector that collected ambient particles. Kessler et al.[3] conducted a selective collection of iron-rich dust particles through natural trichodesmium colonies and examined the collected particles using a scanning electron microscope

(SEM). Furthermore, through an electron microscope and mass spectrometry, nine kinds of particles in East Asia were classified according to their elemental and morphological spectra, including mineral dust, K-rich, sea salt, metal, fly ash, sulfate, nitrate, soot, and organic particles.[4] Although the identification and selection of particles could be achieved by

the microscopic technique, studies proved that the technique was complex and time-consuming. The properties of each particle were manually identified and/or confirmed based on the morphology and spectroscopy. Multiple scans of a sample are required to obtain reliable results, which wastes a lot of resources and leads to high costs. The microscopic technique is also unable to scan the nanoparticles on the entire surface in an acceptable period. In addition, it is impractical to use the microscopic technique for analysis if the sample size is huge. Therefore, there is a need to develop a simple method, which can differentially abandon the nontargeted particles and only detect the targeted particles. Compared to the microscopic technique, the developed method should be highly time-saving and cheap. By differentially removing the nontargeted particles on the entire surface, targeted particles can be reserved for further investigation (e.g., numeration and composition analysis). Concentrations of targeted particles can be obtained by considering collection efficiency, number of targeted particles, and sample volume, and the impact of targeted particles on human health and the environment can be further evaluated by measuring the chemical components and compositions in the targeted particles with the aid of other instruments (i.e., inductively coupled plasma-mass spectrometry and gas chromatography-mass spectrometry). For example, studies indicated that acidic particles play key roles in new particle formation (NPF).[5−7] Two methods were developed by our group for the measurement of acidic particles in the atmosphere. Nevertheless, the methods are neither convenient nor simple.[2,8] More importantly, these methods are time-consuming, so intensive detection of acidic particles cannot be achieved. By differentially removing nonacidic particles and purposefully retaining acidic particles to quantify acidic particles in the atmosphere, the key mechanisms of NPF may be explored and explained, and intensive detection of acidic particles can also be achieved. Thus, in aerosol study, the differential removal of nonacidic particles (nontargeted particles) and the intentional retention of acidic particles (targeted particles) on a substrate are both scientifically and practically meaningful.

Removal of nanoparticles from the surface of a substrate is an imperative but challenging issue in micrometer-scale manufacturing and research.[9,10] As the particle size decreases, the average adhesion stress, defined as the adhesion force per unit adhesive contact area between the particle and the substrate, increases according to the fractional power law. The primary force of adhesion of nanoparticles on a dry surface is mainly controlled by the particle−surface interaction and slightly influenced by the particle−particle interaction.[11,12] Thus, to remove the nanoparticles from the surface, the principle is to overcome the adhesion between the particles and the surface. The adhesion/interaction between the particles and the surface is related to several different mechanisms, including van der Waals (VdW) interaction, deformation of particles and the substrate, and chemical/ hydrogen bonding. Each of these mechanisms is dependent on the interfacial chemical and physical properties of the contact area established between the particles and the surface. Thus, the morphology of the interaction surface plays a controlling role in particle adhesion.[13] To change the interaction between the particles and the surface, a thin film coating is commonly adopted to alter the properties of the adhesive surface. A thin film is a layer of material with a thickness ranging from a few nanometers (single layer) to several microns.

Traditional particle removal methods can be mainly divided into two types, i.e., wet cleaning and dry cleaning. Liquid acoustic cleaning is a common wet-cleaning method.[14] This method is thorough because ultrasound can penetrate anywhere the liquid enters. This method can be further categorized into ultrasonic cleaning (less than 100 kHz) and megasonic cleaning (0.8 to 1.2 MHz).[15] Brems et al.[16] reviewed the removal techniques of acoustics and pointed out the high effectiveness of the technique in removing nanoparticles. Bakhtari et al.[17] removed 63 nm polystyrene latex (PSL) particles from bare silicon wafers and wafers with 4 nm Si cap films by acoustic streaming, and the removal efficiency on both surfaces was ∼99% in less than 10 min of the processing time. However, the damage of surface/materials appeared as the frequency increased, especially in the megasonic range.[16] Furthermore, wet cleaning may have other disadvantages, such as being recontaminated by dissolved chemicals, watermarks, etc.[18] Dry-cleaning methods are also proposed to remove nanoparticles from the surface. The most common dry-cleaning method is spraying the gas (i.e., air, CO_2, and argon). That is, high-pressure gas passes through the nozzle to generate airflow to sweep particles on the surface.[19−21] Xu et al.[22] removed 60−80% of SiO_2 particles from the surface with a jet spray nozzle, accelerated by a N_2 gas flow. In addition to the above traditional methods, several new methods have been proposed to remove particles from the surface, such as gas bullets,[23] plasma,[24] and electrostatics.[25]

Although the methods were developed to remove nanoparticles from the surface, no approach was reported to differentially remove nanoparticles from a surface based on the property of particles. Previous methods removed all of the particles on a surface regardless of particle properties because these methods assumed that the removed particles will not react with the surface, which is not always the case. Thus, this study aims to develop a method for differential removal of nanoparticles on the surface. To overcome the defects of the microscopic technique, the developed method should be simple and effective. More importantly, the effectiveness of the methods should be able to be controlled. Both wet-cleaning and dry-cleaning methods were attempted. The blowing/air jet method is typically simple and frequently used among all dry-cleaning methods and the effect of this method is related to the pressure of air supply and time interval.[22] As for the wet-cleaning process, generation of bubbles in the solution is an important mechanism for particle removal on the surface through the cavitation effect. Nanobubble and ultrasonic are both simple but efficient methods for generating abundant bubbles in the solution. The former one is novel and its effect is controlled by the time of treatment, while the latter one is traditional and the effect is related to the frequency and processing time. Therefore, these three methods were selected and tested. Although all these removal techniques have already existed, previous studies used them to remove all nanoparticles from a surface regardless of their properties. No studies were reported for differential removal of nanoparticles. The novelty of the method introduced in this study is the surface coating that changes the particle−surface interaction, and thus nontargeted nanoparticles could be differentially removed, while targeted nanoparticles are retained on the surface through this traditional removal technique, which is only a component/tool in the developed method. In this study, acidic particles were regarded as targeted particles that should be collected and reserved on the surface, while nonacidic particles
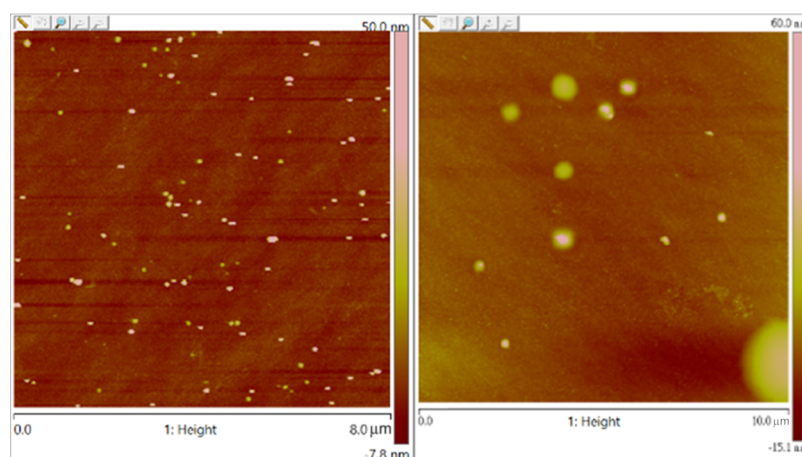
Figure 1. AFM images of the collected standard nonacidic particles (left) and acidic particles (right).
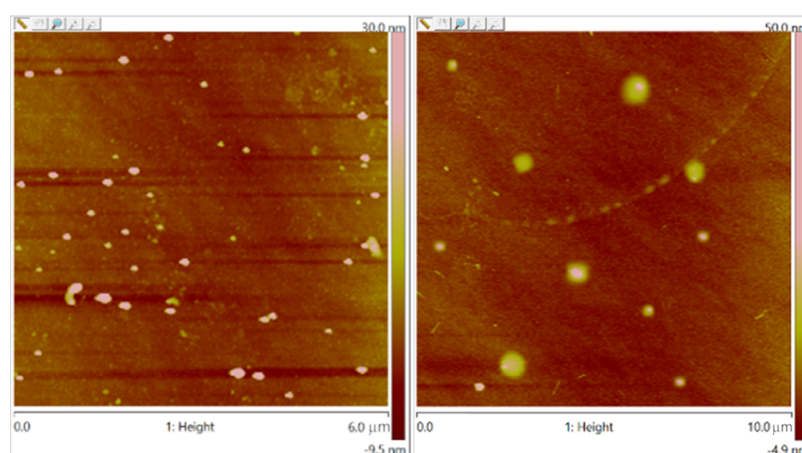

Figure 2. AFM images of nonacidic (left) and acidic particles (right) after air jet treatment.
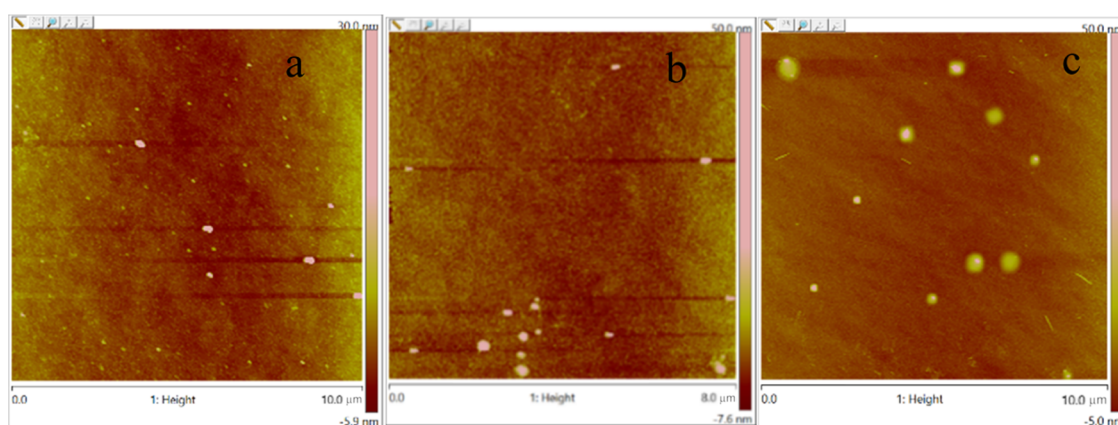

Figure 3. AFM images of nonacidic ((a) 5 treatments and (b) 10 treatments) and acidic particles ((c) 10 treatments) after nanobubble treatment.

were nontargeted particles that would be removed from the surface. The surface was first coated with a thin film to alter the particle–surface interaction for potential differential removal of different particles. AFM was used to verify the particle removal efficiency of the developed method based on the differences in the particle number before and after the application of the developed method. This is the first attempt to differentially remove unwanted particles and intentionally retain wanted particles on a surface. The method is expected to be targeted, simple, effective, low cost, and time-saving.

2. RESULTS AND DISCUSSION

2.1. Air Jet Method. The AFM images of collected nonacidic and acidic particles on the coated detectors are shown in Figure 1. The acidic particles deposited on the detector coated with a nanofilm metal had a clearly distinguishable and unique reaction spot that formed a central elevation with a surrounding yellow halo, visualized through the AFM, while no such reaction spot was observed in the image of nonacidic particles.[2,8,26] Noteworthily, there should not be any overlapping particles on the surface. Moreover, due
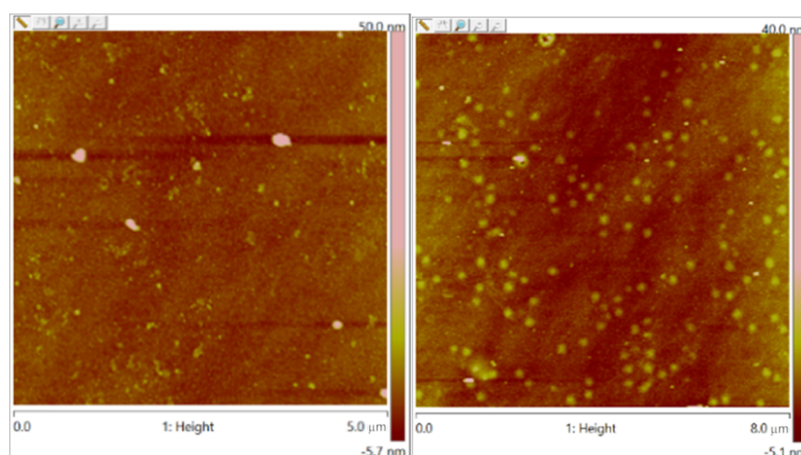
Figure 4. AFM images of nonacidic (left) and acidic particles (right) after ultrasonic treatment with DI water.
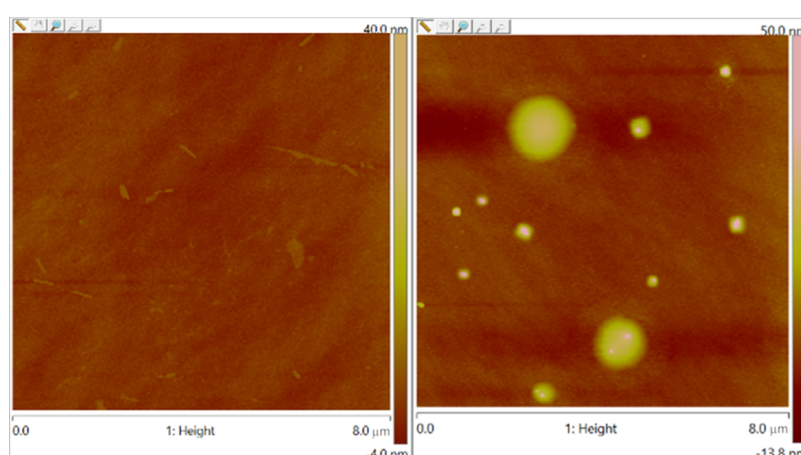

Figure 5. AFM images of nonacidic particles (left) and acidic particles (right) after ultrasonic treatment with ethanol for 30 min.

to the difficulty of the standard acidic particle generation (SAPG) system to control the particle size, the sizes of the acidic particles (0.2−2 μm) generated were usually larger than those of the nonacidic particles (0.03−0.1 μm). In principle, the smaller the particles, the more difficult they are to remove.[11] That is, if the larger acidic particles cannot be removed, the smaller acidic particles will also remain. Therefore, it is not necessary to generate smaller acidic particles (i.e., < 0.2 μm). If the nonacidic particles are removed while the acidic particles remain on the surface using a method, it can prove that this method can be used to differentially remove particles.

The AFM images of detectors containing acidic and nonacidic particles after air jet treatment are shown in Figure 2. Both acidic and nonacidic particles still remained on the surfaces. The removal efficiency of nonacidic particles was as low as 5.1 ± 3.4%. Moreover, a similar number of acidic particles were counted before and after the particle removal treatment. The results indicated that the air jet treatment was ineffective to remove nanoparticles from the surface, regardless of acidic or nonacidic particles. Indeed, the air jet method was commonly effective to remove large particles above 10 μm.[21] For nanoparticles (<1 μm), they were effectively removed from the surface using some unique gases (e.g., CO_2 or Ar) to generate particle beams.[19,20] However, the process/system is complicated. On the one hand, the treated gas (e.g., CO_2 or Ar) needs to be recooled to its triple point of about −170 °C.

On the other hand, the process must be conducted in an ultrahigh vacuum environment (e.g., 10 Torr). Thus, the method is too complicated to be widely used.

2.2. Nanobubble Treatment. Figure 3 shows the AFM images of nonacidic and acidic particles after nanobubble treatment for different times. Similar to the air jet treatment, after 10 nanobubble treatments, acidic particles remained on the surface, and the number of particles was the same as before the treatment (Figures 1 and 3c). In addition, fewer nonacidic particles were observed on the surface, suggesting that the nanobubbles generated by the alcohol−water exchange process had a certain efficiency in removing nonacidic particles from the surface. By increasing the number of nanobubble treatments from 5 to 10, the removal efficiency of nonacidic particles insignificantly increased from 80.8 ± 8.5 to 89.3 ± 4.1% (p > 0.05). Nevertheless, this method still could not completely remove nonacidic particles from the surface. The failure of this method might be caused by the incomplete coverage of the nanobubbles on the entire surface, so the cleaning effect is not complete.

2.3. Ultrasonic Treatment. *2.3.1. Ultrasonic Treatment with DI Water.* Figure 4 presents the AFM images of nonacidic and acidic particles after ultrasonic treatment with DI water. On the one hand, both nonacidic and acidic particles were not completely removed from the surface. On the other hand, after the ultrasonic treatment with DI water, although no visible damage to the surface was noticed, the surfaces seemed to be
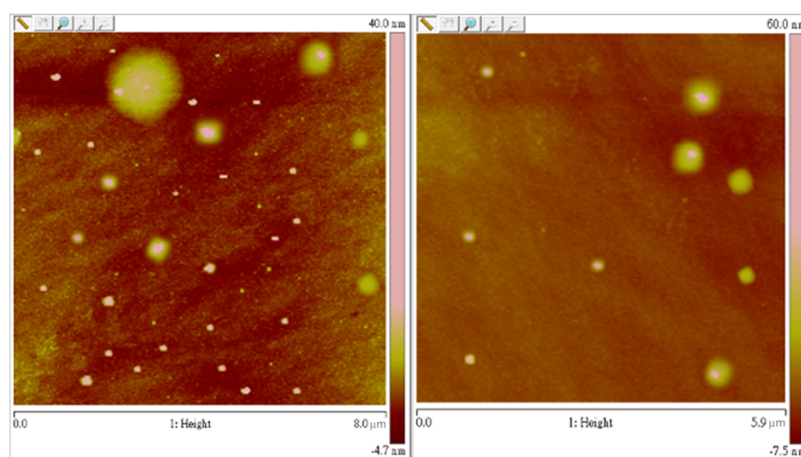
Figure 6. AFM images of a detector surface containing acidic and nonacidic particles before (left) and after (right) ultrasonic treatment with ethanol for 30 min.

contaminated because many nanoimpurities were observed. The contamination on the surface with acidic particles was much more serious than that with nonacidic particles, which was mainly due to the fact that (1) for the surfaces containing nonacidic particles, the long drying process might cause slight contamination, owing to the reception of impurities from the external environment (e.g., ambient air) and (2) for the surfaces with acidic particles, the dissolved acidic particles in DI water could be responsible for the severer contamination because sulfuric acid is soluble in DI water.[27] Eventually, the impurities and the dissolved components resulted in the contamination of the surface.

2.3.2. Ultrasonic Treatment with Ethanol. Figure 5 shows the AMF images of nonacidic and acidic particles after ultrasonic treatment with ethanol for 30 min. For nonacidic particles, a few particles were still on the surface after the ultrasonic treatment for 10 and 20 min (Figure S3), while the removal efficiency reached 100% after 30 min, i.e., all of the nonacidic particles were removed. As for acidic particles, a similar number of acidic particles were counted before and after the treatment, revealing that the ultrasonic treatment with ethanol was unable to remove the acidic particles from the surface of the detectors. Therefore, the ultrasonic treatment with ethanol was likely an effective method for differential removal of particles on the surface. To validate the efficiency of this method, Figure 6 presents the AFM images of a detector surface containing both acidic and nonacidic particles before and after the ultrasonic treatment with ethanol for 30 min. Clearly, all of the nonacidic particles were removed through the treatment, while acidic particles were still present on the surface. It was proved that although the processing time of the ultrasonic method (30 min) was comparable to that of the nanobubble method (~30 min) and less than that of air jet treatment (60 min), the ultrasonic treatment with ethanol was more effective with a higher removal efficiency (ultrasonic: ~100% vs nanobubble: ~90% and air jet: ~5%). Noteworthily, the ultrasonic treatment with ethanol did not cause any visible damage and/or contamination to the thin film. Moreover, the intactness of the acidic particles (with a central elevation and a surrounding yellow halo) was maintained during the treatment (Figures S4 and S5). Unlike the nanobubble method that could not provide full coverage of nanobubbles on the surface, the ultrasonic treatment with ethanol constantly generated bubbles where there is liquid by cavitation, leading to complete removal of nonacidic particles from the surface. However, since the interaction force between acidic particles and the surface is stronger, the acidic particles were not removed. In the next section, the potential mechanism was further discussed.

2.4. Mechanism and Implication. Figure 7 shows the mechanism of differential removal of particles in ultrasonic
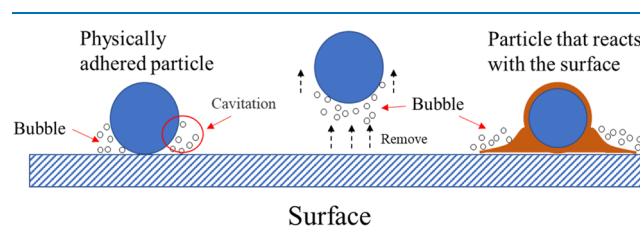

Figure 7. Mechanism of differential removal of particles in ultrasonic treatment.

treatment. The key to ultrasonic cleaning is the bubbles. These bubbles are created by sound waves when they move through water. This is known as cavitation, which is simply the formation of bubbles (cavities) in the solution. Generally, particles collected on the surface of a substrate are either physically adhered particles (e.g., nonacidic particles in this study) or particles that react with the surface (e.g., acidic particles). In the process of differential removal of particles, the physically adhered particles are removed from the surface due to cavitation collapse pressure. The bonding neck between the physically adhered particles and the surface acts as a crevice, which entraps gas and improves cavitation erosion. The generated bubbles can enter the gaps between the physically adhered particles and the surface. In contrast, for the particles that react with the surface, the space between the particles and the surface is filled and no crevice exists (Figures S4 and S5). Under such circumstances, no cavitation occurs, and the bubbles cannot enter the gap to remove the particles.

Based on the results obtained from this study, there are several principles that need to be followed to differentiate the removal of nanoparticles on the surface. First, the targeted particles should react with the surface, while the nontargeted particles only adhere to the surface. To react, the surface can be coated with a thin film of a special material, which can react with the targeted particles. This process ensures the difference in the particle−surface interaction between targeted particles

and nontargeted particles. Second, the components of the targeted particles should not be dissolved in the solution used for sonication. Otherwise, the surface will be contaminated and even the targeted particles will be damaged. Lastly, the solution selected for ultrasonic treatment should be volatile to expedite the drying after treatment and thus avoid contamination during the drying process.

3. SUMMARY AND CONCLUSIONS

In the study, methods were proposed and trialed for differential removal of nanoparticles on the surface. The aim was to remove nontargeted particles but retain targeted particles on the surface. Acidic particles were treated as the targeted particles. A thin metal film was coated on the surface of a substrate so that the acidic particles can react with the surface, and subsequently, the interaction between the particles and the surface alters. Three methods were attempted for differential removal of nanoparticles, including air jet, nano-bubble, and ultrasonic methods. An AFM was used to determine the particle removal efficiency by comparing the difference in the particle number before and after the treatment. Due to the strong particle–surface interaction between the acidic particles and the coated surface, the acidic particles adhered to the surface regardless of before or after the treatment. For the nonacidic particles, the air jet method and the nanobubble method (10 treatments) were not able to completely remove them from the surface with the particle removal efficiencies of 5.1 ± 3.4 and $89.3 \pm 4.1\%$, respectively. In contrast, the particle removal efficiency of the nonacidic particles reached 100% in ultrasonic treatment. Noteworthily, ethanol was a better solution than DI water in ultrasonic treatment, which avoided contamination. At last, the mechanism of differential removal of nanoparticles from the surface was discussed. In ultrasonic treatment, the nonacidic particles are removed from the surface by cavitation, which creates bubbles in the gaps between the nonacidic particles and the surface. In contrast, the space between the acidic particles and the surface is filled, so the bubbles cannot enter the crevice to remove the particles.

4. METHODOLOGY

4.1. Surface Preparation. To change the interaction between the particles and the surface, the surface coating was carried out before the collection of particles to allow the acidic particles to react with the surface. A silicon wafer was chosen as the substrate, and a magnetron sputtering system (MSS) was adopted to coat a nanofilm of metal on the surface of the silicon wafer to form a nanofilm metal detector. Before sputtering, the silicon wafer was first cut into silicon chips with a size of $5 \text{ cm} \times 5 \text{ cm}$ using a laser. Each chip was ultrasonically cleaned with the sulfuric acid solvent (1 mol/L) and ethanol, respectively, to remove the impurities present on its surface. In the MSS process, the base pressure of the chamber was lower than 4×10^{-5} Torr before MS deposition, and the total pressure for sputtering was maintained at 1.3×10^{-2} Torr. In an ultrahigh vacuum environment, the metal target was activated at a high voltage of ~400 V to generate plasma, and then metallic atoms were sputtered onto the surface of the chip. To obtain a metal film thickness of ~25 nm, a sputtering time of 2.5 min was used. After sputtering, the nanofilm metal detectors were stored in a nitrogen atmosphere to avoid oxidation of the nanofilm metal.

4.2. Particle Generation and Collection. The system for generation and collection of nonacidic particles is shown in Figure S1. Nonacidic aerosols were generated using a particle generator (Model 7.811, GRIMM, Germany). One to two drops of three different PSL (Thermo Scientific) microspheres with sizes of 32, 52, and 102 nm were mixed into Milli-Q water to generate one PSL solution, which was then added into the particle generator. The PSL aerosols produced by an atomizer and buffered in a 1.5 L bottle were dried with a silica gel dryer (40 cm long × 5 cm diameter) and then collected onto the nanofilm metal detectors using an electrostatic precipitator (ESP).

The system used to generate and collect acidic particles is shown in Figure S2. A system was established, namely, the SAPG system, which included a nanocarbon particle generation system, a H_2SO_4 aerosol generation system, and a sample collection system. In the nanocarbon particle generation system, a high temperature of 700 °C was controlled in a silicon tube furnace. A large amount of glucose aerosols generated by the ultrasonic atomizer was introduced into a quartz tube in the furnace. At a high temperature, without sufficient oxygen supply for complete combustion, the glucose aerosols underwent thermal decomposition to produce ultrafine carbon particles. The carbon particles were then buffered and cooled in two conical flasks and passed through the headspace of a high-purity H_2SO_4 solution heated on a coil heater at ~400 °C. A filter was placed before the carbon particles passed through the acidic vapor to remove the large particles (i.e., >1 μm). Finally, the mixture of nanocarbon and sulfuric acid vapor was passed through a water-cooled condenser. Sulfuric acid was coated onto the carbon particles during the condensation process to form standard acidic particles, which were then collected on the nanofilm metal detectors using the ESP.

In the study, acidic particles and nonacidic particles were first collected on separated detectors for identification of particles. After that, the detectors were treated with different methods to determine the removal efficiency. Once an effective method was found for differential removal of nonacidic particles, both acidic and nonacidic particles were collected on a detector and proceeded with the treatment to validate the method.

4.3. Methods for Particle Removal. *4.3.1. Air Jet Method.* The schematic of the air jet treatment system in the study is shown in Figure 8. Compressed air was used to provide high-pressure airflow. The air pressure was controlled by a valve and then led into a high-efficiency particulate air (HEPA) filter to remove all of the particles in the compressed air. At the outlet of the HEPA filter, a nozzle was connected to
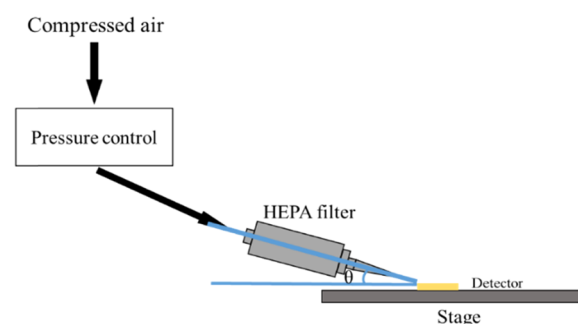

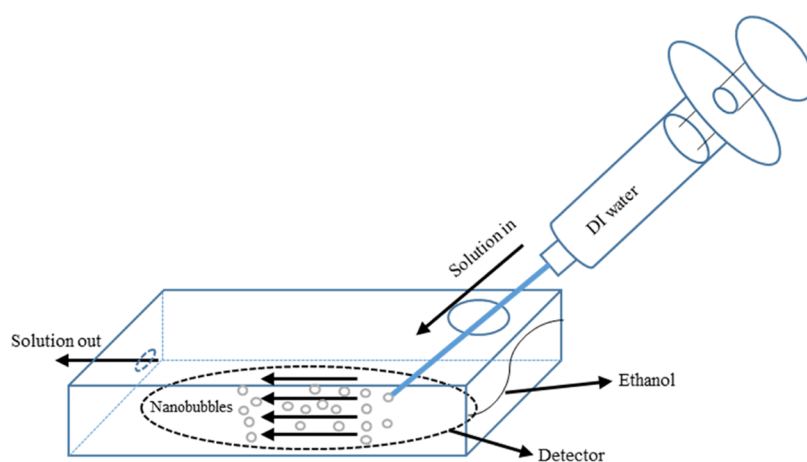**Figure 8.** Schematic of the air jet treatment.

Figure 9. Schematic of the nanobubble process.

generate an air jet. The particle-free air ejected from the nozzle flushed the surface of a substrate at a high pressure of 0.5 MPa. The angle between the nozzle and the surface of the detector was set to ~30°.[20] Air jet treatment was conducted for 60 min.

4.3.2. Nanobubble Method. Nanobubble is a method that can effectively remove nanoparticles from the surface. In the nanobubble process, the surface of a substrate is first covered with ethanol and then rinsed with deionized water (DI water). A large number of nanobubbles were generated through the ethanol−water exchange process to remove nanoparticles, and the coverage rate of nanobubbles on the surface was remarkably high.[19] To adopt this method, a small device (i.e., a diminutive chamber) was developed to provide a suitable space for the ethanol−water exchange process. The schematic of the nanobubble process is shown in Figure 9. The coated detector was first placed in the middle of the diminutive chamber. Ethanol was then poured into the chamber until the surface of the detector was completely covered. A hole was drilled at the top of the diminutive chamber for injection of DI water. The pinhead of the needle was as close as possible to the surface of the detector to flush the surface. The extra ethanol−water solution was discharged from the bypass hole on the left side of the chamber. The duration of one nanobubble treatment was ~3 min. The removal efficiency of the method is related to the treatment time. A previous study claimed that ~80% of physically adhered nanoparticles were removed from a plain silicon wafer after one nanobubble treatment and the removal efficiency was further enhanced to ~90% after three nanobubble treatments.[28] Since the study aimed to completely remove all of the nontargeted particles (i.e., physically adhered particles) on the surface, detectors containing particles were processed five times (~15 min) and 10 times (~30 min) with the nanobubble method, respectively, to take full advantage of this method.

4.3.3. Ultrasonic Method. In this study, ultrasonic treatment was conducted with ethanol and DI water as a solution. Particle-containing detectors were put into a beaker and immersed in ethanol and DI water, respectively. The beaker with the immersed detectors was then agitated in an ultrasonic bath (Crest Ultrasonic, model 275HTAE) at an ultrasonic frequency of ~40 kHz for 30 min. The frequency was commonly used for ultrasonic equipment in previous studies.[29,30]

4.4. AFM Imaging. AFM is a recognized and reliable tool that can be used to assess the cleaning efficiency of a method.[16]

The tapping mode of an AFM (NanoScope, Version 5.31R1, Veeco Instrument Inc.) was used to evaluate the particle removal efficiency of the different methods in the study. AFM images were scanned with the parameter settings as follows: scan rate: 0.6−1 Hz; amplitude setpoint: 0.65−0.80 V; integral gain: 0.2; and proportional gain: 0.5. The scanning areas were from 5×5 μm to 10×10 μm. To minimize the uncertainty in evaluating the particle removal efficiency, each detector was randomly scanned in 16 different areas, following the procedure of the previous study.[31] The particle removal efficiency was determined using the following equation (eq 1)

$$\eta = 1 - \frac{n_a \cdot S_i}{n_i \cdot S_a} \quad (1)$$

where η is the particle removal efficiency, n_i is the average number of particles on the surface before the particle removal treatment, n_a is the average number of particles on the surface after the particle removal treatment, S_i is the AFM scanning area before the particle removal treatment, and S_a is the AFM scanning area after the particle removal treatment.

■ ASSOCIATED CONTENT

ⓢ Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.1c00334.

> Generation and collection systems for nonacidic and acidic particles; 2D AFM images of nonacidic particles; and 3D images of acidic particles before and after ultrasonic treatment (PDF)

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Notes

The authors declare no competing financial interest.

■ REFERENCES

(1) Cheng, M. D. Selective collection of airborne particulate matter. *Aerosol Air Qual. Res.* **2018**, *18*, 1361−1365.

(2) Wang, D. W.; Guo, H.; Chan, C. K. Measuring ambient acidic ultrafine particles using iron nanofilm detectors: method development. *Aerosol Sci. Technol.* **2012**, *46*, 521−532.

(3) Kessler, N.; Armoza-Zvuloni, R.; Wang, S.; Basu, S.; Weber, P. K.; Stuart, R. K.; Shaked, Y. Selective collection of iron-rich dust particles by natural Trichodesmium colonies. *ISME J.* **2020**, *14*, 91−103.

(4) Li, W.; Shao, L.; Zhang, D.; Ro, C. U.; Hu, M.; Bi, X.; Geng, H.; Matsuki, A.; Niu, H.; Chen, J. A review of single aerosol particle studies in the atmosphere of East Asia: morphology, mixing state, source, and heterogeneous reactions. *J. Cleaner Prod.* **2016**, *112*, 1330−1349.

(5) Fiedler, V.; Maso, M. D.; Boy, M.; Aufmhoff, H.; Hoffmann, J.; Schuck, T.; Birmili, W.; Hanke, W.; Uecker, J.; Arnold, F.; Kulmala, M. The contribution of sulphuric acid to atmospheric particle formation and growth: a comparison between boundary layers in Northern and Central Europe. *Atmos. Chem. Phys.* **2005**, *5*, 1773−1785.

(6) Kulmala, M.; Lehtinen, K. E. J.; Laaksonen, A. Cluster activation theory as an explanation of the linear dependence between formation rate of 3nm particles and sulphuric acid concentration. *Atmos. Chem. Phys.* **2006**, *6*, 787−793.

(7) Yue, D. L.; Hu, M.; Zhang, R. Y.; Wang, Z. B.; Zheng, J.; Wu, Z. J.; Wiedensohler, A.; He, L. Y.; Huang, X. F.; Zhu, T. The roles of sulfuric acid in new particle formation and growth in the mega-city of Beijing. *Atmos. Chem. Phys.* **2010**, *10*, 4953−4960.

(8) Wang, D. W.; Guo, H.; Chan, C. K. Diffusion sampler for measurement of acidic ultrafine particles in the atmosphere. *Aerosol Sci. Technol.* **2014**, *48*, 1236−1246.

(9) Salih, H. H.; El Badawy, A. M.; Tolaymat, T. M.; Patterson, C. L. Removal of stabilized silver nanoparticles from surface water by conventional treatment processes. *J. Nanopart. Res.* **2019**, *08*, 21.

(10) Kohli, R. Electrostatic Removal and Manipulation of Small Particles and Surface Cleaning Applications. In *Developments in Surface Contamination and Cleaning: Applications of Cleaning Techniques*; Elsevier, 2019; pp 391−421.

(11) Bowling, R. A. A Theoretical Review of Particle Adhesion. In *Particles on Surfaces 1*; Springer: Boston, MA,1988; pp 129−142.

(12) Busnaina, A.; Bakhtari, K.; Park, J. G. Particle Deposition and Adhesion. In *Handbook of Silicon Wafer Cleaning Technology*; William Andrew Publishing, 2018; pp 153−184.

(13) Cooper, K.; Gupta, A.; Beaudoin, S. Substrate morphology and particle adhesion in reacting systems. *J. Colloid Interface Sci.* **2000**, *228*, 213−219.

(14) Gale, G.; Busnaina, A. Removal of particulate contaminants using ultrasonics and megasonics: a review. *Part. Sci. Technol.* **1995**, *13*, 197−211.

(15) Busnaina, A. A.; Gale, G. W. Ultrasonic and megasonic particle removal. *Proc. Precis. Clean.* **1995**, *15*, 347−359.

(16) Brems, S.; Hauptmann, M.; Camerotto, E.; Pacco, A.; Kim, T. G.; Xu, X.; Wostyn, K.; Mertens, P.; De Gendt, S. Nanoparticle removal with megasonics: a review. *ECS J. Solid State Sci. Technol.* **2014**, *3*, N3010.

(17) Bakhtari, K.; Guldiken, R. O.; Makaram, P.; Busnaina, A. A.; Park, J. G. Experimental and numerical investigation of nanoparticle removal using acoustic streaming and the effect of time. *J. Electrochem. Soc.* **2006**, *153*, G846.

(18) Otani, Y.; Namiki, N.; Emi, H. Removal of fine particles from smooth flat surfaces by consecutive pulse air jets. *Aerosol Sci. Technol.* **1995**, *23*, 665−673.

(19) Kim, I.; Hwang, K.; Lee, J. Removal of 10-nm contaminant particles from Si wafers using CO_2 bullet particles. *Nanoscale Res. Lett.* **2012**, *7*, 211.

(20) Lee, J. W.; Hwang, K. S.; Lee, K. H.; Yi, M. Y.; Lee, M. J. Removing 20 nm particles using a supersonic argon particle beam generated with a contoured Laval nozzle. *J. Adhes. Sci. Technol.* **2009**, *23*, 769−777.

(21) Donovan, R. P. *Particle Control for Semiconductor Manufacturing*; Marcel Dekker: New York, 1990; pp 1−480.

(22) Xu, K.; Pichler, S.; Wostyn, K.; Cado, G.; Springer, C.; Gale, G. W.; Dalmer, M.; Mertens, P. W.; Bearda, T.; Gaulhofer, E.; Podlesnik, D. Removal of nano-particles by aerosol spray: effect of droplet size and velocity on cleaning performance. *Solid State Phenom.* **2009**, *145−146*, 31−34.

(23) Kim, I.; Lee, J. The removal of 10-nm contaminant particles from micron-scale trenches using CO_2 nano bullets. *J. Nanopart. Res.* **2013**, *15*, No. 1579.

(24) Park, J. K.; Yoon, J. W.; Whang, K. H.; Cho, S. H. Removal of nanoparticles on silicon wafer using a self-channeled plasma filament. *Appl. Phys. A* **2012**, *108*, 269−274.

(25) Kawamoto, H.; Guo, B. Improvement of an electrostatic cleaning system for removal of dust from solar panels. *J. Electrost.* **2018**, *91*, 28−33.

(26) Lu, H.; Lyu, X.; Guo, H. A novel semi-automatic method for measuring acidic ultrafine particles in the atmosphere. *Atmos. Environ.* **2020**, *245*, No. 118044.

(27) Margarella, A. M.; Perrine, K. A.; Lewis, T.; Faubel, M.; Winter, B.; Hemminger, J. C. Dissociation of sulfuric acid in aqueous solution: determination of the photoelectron spectral fingerprints of H_2SO_4, HSO_4^-, and SO_4^{2-} in water. *J. Phys. Chem. C* **2013**, *117*, 8131−8137.

(28) Yang, S.; Duisterwinkel, A. Removal of nanoparticles from plain and patterned surfaces using nanobubbles. *Langmuir* **2011**, *27*, 11430−11435.

(29) Charasseangpaisarn, T.; Wiwatwarrapan, C. The effect of various frequencies of ultrasonic cleaner in reducing residual monomer in acrylic resin. *Ultrasonics* **2015**, *63*, 163−167.

(30) Paniwnyk, L.; Alarcon-Rojo, A.; Rodriguez-Figueroa, J. C.; Toma, M. The use of ultrasound as an enhancement aid to food extraction. In *Ingredients Extraction by Physicochemical Methods in Food Handbook of Food Bioengineering*; Grumezescu, A. M.; Maria, A., Eds.; Elsevier, 2017; pp 399−440.

(31) Cohen, B. S.; Heikkinen, M. S.; Hazi, Y.; Guo, H.; Peters, P.; Lippmann, M. Field evaluation of nanofilm detectors for measuring acidic particles in indoor and outdoor air. *Res. Rep. - Health Eff. Inst.* **2004**, *121*, 1−35.