ABSTRACT: The semiconductor industry continues to shrink the device sizes while applying more complex shapes and using diverse materials, which requires parallel improvements in the quality of ultrapure reagents. The need for ultrapure reagents has led to ever-higher demands for the performance of analytical instruments used to detect ultratrace impurities. In this study, nonvolatile impurities in ultrapure reagents were quantified using a scanning mobility particle sizer (SMPS). The performances of three different sample introduction systems, i.e., an electrospray (ES), an aerosol generator with a heating chamber and a Nafion desolvation membrane (NB-II), and a MicroMist nebulizer with a heated cyclonic spray chamber and a three-stage Peltier-cooled desolvation system (MM-APEX), were evaluated for the lower limit of detection of a SMPS. The MM-APEX equipped with the SMPS was able to detect NaCl additives at a concentration of 100 parts per trillion (ppt, ng/L) in ultrapure water, which was approximately 104- and 102-fold lower than those of ES and NB-II, respectively. The practical application of MM-APEX with the SMPS for commercial isopropanol samples was also studied. The results clearly demonstrate that the impurity concentrations presented by the NaCl-equivalent concentrations among different sources of isopropanol were at the ppt to parts-to-billion (ppb) scale. The SMPS system equipped with MM-APEX is capable of recognizing impurities with concentrations ranging from tens ppt to thousands of parts per million (ppm), which is beneficial for an ultratrace analysis of nonvolatile impurities in semiconductor process chemicals.

INTRODUCTION

The global semiconductor market is projected to reach US$ 726.73 billion by 2027. A faster, miniaturized, and advanced memory chip is in high demand owing to the emergence of artificial intelligence, the Internet of Things, and machine learning, among other areas. With the reduction in electronic devices, the effect of defects, which frequently emerge during the manufacturing process, is significantly amplified and influences the performance of the product. Defects can be caused by impurities in ultrapure reagents during processing. The types of impurities include organic compounds, metals, ionic species, and nanoparticles, which are detected by qualitative and quantitative measurements conducted using several methods, including total organic carbon (TOC) monitoring, ion chromatography (IC), inductively coupled plasma mass spectrometry (ICP-MS), and liquid particle counters. However, these metrology tools are limited because they provide limited information on specific impurity items. For example, IC and ICP-MS measure the ionic species and metal ions in ultrapure reagents, respectively. The liquid particle counter measures the number of particles and provides the size distribution of the particle impurity in an aqueous sample, whereas the major restriction is its resolution in terms of the particle size, from which particles with diameters of <20 nm cannot be recognized. A method that satisfies the industrial requirements, including fast measurements, high sensitivity to impurities, and suitability to a wide range of impurity types, is required for in-line and real-time monitoring.

Nonvolatile residues (NVRs) can be used as an alternative for the fast identification of contamination in ultrapure reagents. NVRs consist of dissolved inorganic materials (cations and anions), particles, and high-boiling-point organics, other than the solvent itself. NVR detection provides an immediate indicator of contamination. Blackford et al.
developed a system for monitoring the NVR concentration in ultrapure water using a nebulizer and a condensation particle counter (CPC). The signal-to-noise ratio was approximately 10:1 when measuring KCl at 9.8 parts per trillion (ppt) in ultrapure water. Allen et al. used a concentric pneumatic nebulizer with CPC as a detector for liquid chromatography. They concluded that the specific limits of detection (LODs) obtained are strongly dependent on the aerosol source employed, and values as low as 10–100 fg/mL for 200 μL of flow injected samples of NaNO₃ in ultrapure water could be obtained in their study. Although an extremely low LOD could be achieved using a nebulizer coupled with CPC, the dry residue size distribution could not be obtained.

In addition to nebulizers and CPC systems, a scanning mobility particle sizer (SMPS) has been adopted for NVR measurements. A SMPS has been well developed for the characterization of the nanoparticle size and is typically equipped with a nebulizer that aerosolizes the nanoparticles in an aqueous solution and a CPC for quantification of the nanoparticle number. Rather than measuring the size of the nanoparticles, this technique can also be used for impurity detection, from which the nonvolatile, suspended impurity forms residual particles from the dried aerosols. The NVR measurement through the SMPS works by forming an aerosol of the liquid through a nebulizer, evaporating the liquid in the aerosol, and measuring the size and number of particles in the aerosol. As the total NVR increases, the mode of the residual particle size distribution shifts to higher diameters and number concentrations. The SMPS provides the total impurity instead of a single impurity type, indicating that it is a universal rather than a selective detector for the determination of nonvolatile impurities.

Several studies have demonstrated the capability of SMPSs to characterize impurities in a solvent or those released from storage bottles. In addition, residual particle monitors based on SMPS have previously been proposed and adopted as an ASTM standard. These approaches shed light on the potential application of the SMPS for the detection of impurities in suspended reagents. The limit of detection for NVR measurements using an SMPS is dependent on several experiment parameters, including the solution uptake rate, carrier gas flow rate, nebulization efficiency, and composition of the NVR. Knight et al. used a SMPS with an ultrasonic nebulizer for residue particle measurements of deionized water stored in a variety of bottles. They found that the absolute residual particle concentration obtained using a Meinhard concentric pneumatic nebulizer is approximately 10 times lower than that of an ultrasonic nebulizer, clearly demonstrating the influence of the sample introduction system on the detection limit. In addition, Stabile et al. studied the effects of the salt type and molar concentration on the nebulization. Higher particle number concentrations and distributions measured using an SMPS were observed for more concentrated solutions (2 mM), whereas distributions toward smaller diameters were observed for the diluted solution (2 μM). The differences among the various solutions are nearly negligible for the lowest molar concentration (2 μM), indicating the NVR-concentration-dependent nebulization property.

In this study, three different sample introduction systems, namely, an electrospray (ES), an aerosol generator with a heating chamber and a Nafion desolvation membrane (hereafter referred to as NB-II), and a MicroMist nebulizer with a heated cyclonic spray chamber and a three-stage Peltier-cooled desolvation system (MM-APEX), were selected for a performance evaluation of an impurity detection using a SMPS. ES and NB-II are the most popular sample introduction systems adopted by a SMPS for nanoparticle characterization; however, they have rarely been studied for NVR measurements. A modern pneumatic nebulizer coupled with a desolvation system (MM-APEX) was also evaluated as an alternative sample introduction system for the lower limit of detection of impurities when applying a SMPS. To demonstrate the capability of the measurement system, both ultrapure water (UPW) and isopropyl alcohol (IPA) were
chosen because they are employed during the final steps of the wet cleaning process in the semiconductor industry. Any nonvolatile residue present in the UPW and IPA can remain on the wafer surface after the solvent has evaporated, possibly causing defects in the resulting semiconductor device. Therefore, there is a need to monitor the UPW and IPA in such processes to ensure that the concentration of the NVR remains at or below an acceptable level. The concentration of NVR in the UPW and IPA solvents was determined by the equivalent sodium chloride (NaCl) concentration calculated from the correlation curve between the spiked NaCl concentration and the volume concentration from the SMPS. In addition, sodium chloride (NaCl) was added to the UPW as an impurity to assess the LOD of different sample introduction systems. Commercially available ultrapure isopropanol of different qualities was tested, and impurity concentrations at the ppt scale were recognized by an MM-APEX coupled with the SMPS system. The capability of the SMPS for impurities with ultralow concentrations was approved, and its promising application in in-line impurity monitoring was demonstrated.

**RESULTS AND DISCUSSION**

**Evaluation of Three Sample Introduction Systems.** The measurement principle of a SMPS with the sample introduction system is shown in Figure 1. The aqueous solution in the bottle was self-aspirated using a nebulizer and then transformed into small droplets. The gas flow carries nebulized droplets into a heater and becomes a dried particle that moves to the SMPS for particle distribution measurements. The residual particle diameter depends on the total dissolved impurity concentration in the solvent and the nebulized droplet diameter. A higher dissolved impurity concentration and larger droplet size led to a larger residue particle size. Prior to evaluating their performance in an impurity analysis, the performances of the different sample introduction systems were tested.

An ES is known to generate monodisperse droplets, that is, droplets with a narrow size distribution, and the sucrose method has been commonly used for the calculation of the droplet size produced by the ES. The diameter of the residual particles was measured using an SMPS and correlated with the concentration of the sucrose solution, allowing the droplet size to be derived. The particle size distribution of the residue at different sucrose concentrations is shown in Figure 2a. Here, five concentrations of sucrose solutions were prepared and the detected diameters were 18.8 nm for 3000 ppm, 13.5 nm for 1000 ppm, 10.9 nm for 500 ppm, 8.8 nm for 300 ppm, and 6.8 nm for 100 ppm sucrose. The mean particle diameter increased with increases in the sucrose concentrations. Similar to the observations of Park et al., the diameters of the dominant residue particles increased linearly with the cubed root of the concentration. Figure 2b shows the measured residue size as a function of the sucrose concentration. The experimental data were fitted using a power form equation, and the power law of the fitting curve was 0.323. This result is consistent with the theoretical value, and the calculated diameter of the droplet generated by the ES in this study was 145 nm.

The droplet sizes of the other two nebulizers (NB-II and MM) were measured using a laser diffraction system. Figure 3 shows the primary aerosol diameter distribution obtained using the NB-II and MicroMist nebulizers at self-aspirated flow rates of 227 and 154 μL/min, respectively. Both distributions have two peaks, with one centered at 1 μm and the other at 8.6 μm for NB-II, and one centered at 2.5 μm and the other at 10 μm for a MicroMist. The Sauter mean diameter ($D_{3,2}$) obtained using NB-II was 1.1 μm, which is lower than that obtained using a MicroMist (2.0 μm).

Figure 2. (a) Residue particle size distribution at different sucrose concentrations. (b) Correlation between the measured residue diameter and the concentrations of the sucrose solution.

Figure 3. Droplet diameter distribution of the primary aerosols generated using two different nebulizers (NB-II and MM).
In addition to the droplet size, the nebulization efficiency of the three sample introduction system affects the amount of droplet transfer to the SMPS. The nebulization efficiency was determined based on the nebulization of NaCl at known concentrations, accompanied by knowledge of the aerosol flow rate, liquid flow rate, and total mass concentration measured from the SMPS. The nebulization efficiencies of ES, NB-II, and MM-APEX were 0.0015, 0.393, and 68.8, respectively (Table 1). The lower nebulization efficiency of the ES and NB-II sample introduction systems can be attributed to the loss of particles during their transportation to the SMPS. In particular, for NB-II, the Nafion membrane led to a loss of particles at a ratio of 13.1%, as shown in Table S1. By contrast, the nebulization efficiency of the MM-APEX systems is approximately 10^{-5} and 10^{-6}-fold those of the ES and NB-II, respectively, mostly because of the larger aerosol Sauter mean diameter obtained using the nebulizer, which reduces the diffusion loss of particles inside the tube. MM-APEX has a much higher amount of sample loading because of the higher sample flow rate and nebulization efficiency and is more effective for a diluted sample measurement.

**Nebulizer-Dependent Limit of Detection.** To determine the concentration in an unknown sample using a SMPS, a calibration curve consisting of an additive concentration of the detected volume concentration of the residue was constructed. In this study, the equivalent impurity concentration was obtained in terms of the [NaCl] concentration from the volume concentrations of the nanoparticles through the calibration curve. The selected range of NaCl concentrations was 100 to 3000 ppm for an ES, 500 ppt to 100 ppm for NB-II, and 100 ppt to 1 ppm for MM-APEX. The size distributions of the residual NaCl particles at different NaCl concentrations were measured, and a reduction in both the diameter and the number concentration of residue particles corresponding to the decreasing additive concentration was observed (Figure S1). By integrating the total detected NaCl volume concentration and corresponding to the concentration of the NaCl solution, calibration curves of NaCl for various nebulizers were obtained, as shown in Figure 4. The detection limits are 105, 7.21, and 0.07 ppb for the ES, NB-II, and MM-APEX, respectively, which were calculated by dividing 3σ by the slope. Among the three sample introduction systems, MM-APEX showed the best performance because of its lower detection limit, which could be attributed to the dry aerosol conditions, higher nebulization efficiency, and a moderate flow rate. The higher nebulization efficiency of MM-APEX may be attributed to the larger aerosol Sauter mean diameter, which reduces the diffusion loss of the particles inside the tube. Our previous study compared three sample introduction systems: a high-performance concentric nebulizer with a heated cyclonic spray chamber, a three-stage Peltier-cooled desolvation system (HPCN-APEX), a conventional sample introduction system (i.e., a commercially available MicroMist nebulizer with a cyclonic spray chamber), and a total consumption (TC) system for single-particle inductively coupled plasma mass spectrometry. Our previous results showed that the size and number detection limits of HPCN-APEX were 1.6- and 10-fold lower than those of the TC and conventional sample introduction systems, respectively, which are similar to the current results, indicating that a pneumatic concentric nebulizer coupled with a desolvation system is a suitable sample introduction system for an impurity analysis of an ultrapurereagent.

![Figure 4](https://doi.org/10.1021/acsomega.1c07168)  
**Figure 4.** Calibration curve of different sample introduction systems.

Detection of the Equivalent Impurity Concentration in Isopropanol. The applicability of the MM-APEX system to a SMPS was examined by determining the impurity concentration in various types of IPA. The calibration curve built using spike/IPA is necessary because IPA presents different properties from water, such as viscosity, and because the residue volume can differ. The NaCl-equivalent impurity concentration was calculated by fitting the detected volume concentration of the impurity from the IPA samples to the NaCl/IPA calibration curve. Figure 5a shows the calibration curve of the NaCl spike in IPA. The detection limit is 10.5 ppt as calculated by dividing 3σ by the slope, which was lower than the NaCl spike in water. One potential cause of the lower detection limit of IPA is the change in the initial wet aerosol size distribution. Browner et al. reported that the initial wet aerosol size distribution produced from a 100% organic solvent is narrower and smaller than that from water. The impurity concentration of the seven IPA samples is shown in Figure 5b. The calculated equivalent NaCl concentration was 109 ppt for IPA-1, 1.31 ppb for IPA-2, 0.58 ppb for IPA-3, 135 ppt for IPA-4, 185 ppt for IPA-5, 5.25 ppb for IPA-6, and 3.82 ppb for IPA-7. The equivalent NaCl concentration provides a conceptual quantitation of the nonvolatile residue in an IPA. However, NaCl is not the only component, and the main composition exists in IPA batches. To validate the accuracy of this method, a 307 ppt NaCl spike IPA was used as an unknown sample, and its concentration was determined by applying both a SMPS and an ICP-MS. Figure S2 shows the calibration curve of the volume concentration measured using the SMPS and the intensity measured by the ICP-MS in

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**Table 1. Parameter Values Associated with the Determination of Nebulization Efficiency Using the Three Sample Introduction Systems**

| Nebulizer      | ES     | NB-II | MM-APEX |
|----------------|--------|-------|---------|
| droplet size   | 145 nm | 1.1 μm| 2.0 μm  |
| [NaCl] (g/L)   | 0.1    | 0.001 | 0.001   |
| drying temperate (°C) | 20    | 140   | 140     |
| carrier gas flow rate (L/min) | 1.5  | 3     | 1       |
| mass detected by SMPS (g/L) | 6.41 \times 10^{-12} | 2.98 \times 10^{-8} | 1.06 \times 10^{-5} |
| sample flow rate (μL/min) | 0.066 | 227   | 154     |
| nebulization efficiency (ε) | 1.5 \times 10^{-3} | 3.93 \times 10^{-1} | 6.88 \times 10^{1} |
understanding of the reagent purity. A correlation between these instruments and SMPS for a better adoption of di-ppt-scale impurities from a reagent. The results also suggest the capability of the system built in this study for the detection of concentrations in several IPA batches were within the range of concentrations on atomization. Higher particle number effects of salts and molar concentrations measured using a SMPS. The detection limit for MM-APEX was 0.07 ppb for NaCl spiked in water, which is lower than those for the other two systems. In addition, MM-APEX equipped with a SMPS shows promise for application in the impurity concentration analyses of ultrapure reagents. A systematic evaluation of the sample introduction systems presented in this study is crucial for SMPS applications. Users can choose a suitable instrument configuration for improved detection efficiency and lower detection limits for an impurity analysis using a SMPS.

CONCLUSIONS

The performances of three different sample introduction systems, i.e., an ES, an aerosol generator with a heating chamber and a Nafion desolvation membrane (NB-II), and a MicroMist nebulizer with a heated cyclonic spray chamber and a three-stage Peltier-cooled desolvation system (MM-APEX), were systematically evaluated in terms of their residue analysis using a SMPS. The nebulization efficiencies of the ES, NB-II, and MM-APEX were 0.0015, 0.393, and 68.8, respectively, indicating that MM-APEX was more advantageous for dilute samples. The detection limit for MM-APEX was 0.07 ppb for NaCl spiked in water, which is lower than those for the other two systems. In addition, MM-APEX equipped with a SMPS shows promise for application in the impurity concentration analyses of ultrapure reagents. A systematic evaluation of the sample introduction systems presented in this study is crucial for SMPS applications. Users can choose a suitable instrument configuration for improved detection efficiency and lower detection limits for an impurity analysis using a SMPS.

EXPERIMENTAL SECTION

Chemicals and Sample Preparation. Sodium chloride (NaCl) was dissolved in ultrapure water and isopropanol (IPA) at different concentrations ranging from ppt (ng/L) to parts per million (ppm) (μg/L) to model the impurities in the reagent. Isopropanol of different purities was then labeled and tested. The amount of impurity in the IPA is presented as an equivalent concentration toward the NaCl calibration curve.

Sample Introduction System. Three sample introduction systems, including an electrospray (ES, TSI-3480), an aerosol generator (TSI 3076) with a heating chamber and a Nafion membrane (NB-II), and a MicroMist nebulizer with a heated cyclonic spray chamber and a three-stage Peltier-cooled desolvation system (MM-APEX) (hereinafter referred to as MM-APEX) (Elemental Scientific, Omaha, NE) were used in this study. The drying temperatures of the ES, NB-II, and MM-APEX were 20, 140, and 140 °C, respectively. The droplet size generated by ES was calculated as 145 nm using the sucrose method reported in the literature, and the droplet sizes generated by nebulizer II and MM-APEX were characterized using a laser diffraction system (Spraytec, Malvern Panalytical). The calculation of the nebulizer efficiency (ε), as shown in eq 1, follows that of a previous study, which considered the flow rates of the carrier gas (Qc) and aqueous sample (Qs), the concentration of the NaCl solution (C), and the total detected mass concentration of NaCl in the aerosol (C).
In this study, $Q_s$ was a high-velocity gas jet to blast the solution into an aerosol and was set as 1.5, 3, and 1 L/min for ES, NB-II, and MM-APEX, respectively. In addition, $Q_s$ was 65.6 mL/min for ES. The sample flow rates ($Q_s$) for both NB-II and MM-APEX have a strong dependence on the carrier gas flow rates and the dimensions of the sample uptake tubing owing to the nature of a pneumatic nebulizer. The devices were operated according to the manufacturer's specifications; therefore, the sample uptake was 227 µL/min for NB-II and 154 µL/min for MM-APEX. The total detected mass concentration derived from the SMPS assumes that all of the particles are perfect spheres with a known density, the calculation of which is shown in eq 2.

$$C_s = \frac{N_i \pi}{6} D_{pi}^3 \rho$$  \hspace{1cm} (2)

where $N_i$ is the number concentration at each channel size, $D_{pi}$ is the geometric midpoint of each particle size channel, and $\rho$ is the density (2.17 g/cm³).

**Instrument.** All measurements were conducted using a differential mobility analyzer (TSI-3080) connected to a nanoDMA column (TSI-3085) and a condensed particle counter (TSI-3776). The sheath flow was 15 L/min and the aerosol flow rate entering the CPC was 1.5 L/min. The size of each sample was measured five times and presented as the mean ± standard deviation.

**ASSOCIATED CONTENT**

Supporting Information
The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.1c07168.

Particle size distribution of different NaCl concentrations spikd in water with different sample introduction systems and particle size distribution of different batches of IPA (PDF)

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**Author Contributions**
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**Notes**
The authors declare no competing financial interest.

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