Removal of Air Pollutants in the Make-Up Air of a Semiconductor Plant by Fine Water Spray

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Published online: 15 Feb 2007.

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ABSTRACT

This study presented a fine water spray technique to remove acidic/basic gas/particulate from the make-up air of a semiconductor manufacturing plant. The concentration of air pollutants was measured at the inlet and the outlet of the device with the help of HDS (Honeycomb Denuder System) samplers followed by sample analysis. Results show that the removal efficiency of the fine water spray device for gas pollutants, i.e., HF, HCl, HNO_2, HNO_3, SO_2, and NH_3 was higher than that for ionic species, i.e., Cl^-, NO_3^-, SO_4^{2-} and NH_4^+ in fine particulates. For example, the removal efficiency was 85.7 ± 7.8 and $66.2 \pm 22.3\%$ for SO_2 and NH_3, respectively, whose inlet concentration ranged from 3.1 ± 1.4 and 13.2 ± 6.9 ppb, respectively. When gas pollutant

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concentration was at low level, it was observed that the concentration at the outlet was close to that at the inlet of the fine water spray device. This is due to the reduction of the concentration gradient at the gas–liquid interface, which lowers the absorption efficiency.

Key Words: Air pollutants; Fine water supply; Removal efficiency.

INTRODUCTION

The air quality of a clean room becomes more demanding as the trend toward smaller feature size in semiconductor manufacturing continues. To prevent device failure and increase the product yield, it is important to manufacture the products in a "contaminant-free" environment. Many production engineers have made a great effort to remove particulate contaminations from the manufacturing environment. Besides that, the particulates are harmful to the products, and gas species are also increasingly critical to the product yield. Muller et al.[1] reported that the surface arrival rates of gas species are several times higher than that of particulates due to their greater diffusivities and deposition velocities. Saiki et al.[2] reported that inorganic gases are believed to react with ammonia in the clean room air and cause irreversible damage to process wafers. Ye et al.[3] investigated the formation of particles and showed that stable aerosol of the order of $0.2 \, \mu m$ is formed during vacuum pump down process. The traces of gases, i.e., SO_2 and H_2O_2 react in the liquid phase during water vapor condensation to form stable sulfuric acid droplets.

In semiconductor industry, the white smoke emitting from the stacks of wet scrubbers is a common problem of the plant.[4] The results showed that the main chemical species in the waste stream of the wet scrubbers are particulate silica, ammonium and chloride, and gaseous ammonia and chloric acid. It is believed that the emission has an influence on the air quality in the clean room. Removing pollutants at the intake of the make up air to the clean room is important to the increase of product yield. In this study, we have investigated fine water spray technique to remove air pollutants at the make up air supply of the clean room.

In this study, a honeycomb denuder/filter pack system (HDS, Ogawa & Co. Inc.) was used to measure the concentrations of the acidic/basic air pollutants at the inlet and the outlet of the proposed fine water spray device inside the air conditioning system, which was installed at the inlet of the make-up air system of the clean room. The removal efficiency of the device for both acidic/basic gases and particles was studied.

METHODS AND MATERIALS

Diffusion denuder was used in this study. It is a sampler that removes gases from an aerosol stream to measure their concentrations separately. Gas or vapor molecules diffuse rapidly to the wall of a diffusion sampler and adsorb onto the wall coated with a suitable material. The gas concentration can be determined by extracting the coated substrates and analyzing the samples.[5–9] Various denuders were designed and reported in the last 20 years. Pui et al.[6] designed a compact coiled denuder and compared the performance with an annular denuder.[5] Koutrakis et al.[8] and Sioutas et al.[9] developed a glass honeycomb denuder/filter pack system (HDS) to collect atmospheric gases and particles. The system is considerably smaller than the annular denuder system and can be easily used for large field studies. The field studies indicated an excellent agreement between the HDS and HEADS (Harvard-EPA Annular Denuder System) for determining the concentrations of particulate SO_4^{2-}, NH_4^+, and gas-phase HNO_3 and NH_3. Tsai et al.[10] compared the acidic aerosol concentrations measured by an annular denuder system (ADS) and a HDS in a field study. The results showed that the HDS performed equally well as the ADS for atmospheric aerosol sampling, except for the measurement of the gas-phase HNO_3.

Figure 1 shows the schematic representation of the air-conditioning system in the make-up air inlet. The system includes a prefilter, a fan, a fine water spray device, a heating equipment, and a HEPA filter. The fan draws outside air through the prefilter, before entering the fine water spray device. The scrubbed air then enters the heating equipment and passes through the HEPA filter and clean room. The fine water spray device (section III, in Fig. 1) contains 1200 nozzles and a demister. The demister was installed with a steeper flute angle to direct the water toward the air-entering side. The nozzles were capable for spraying small size droplets with an accurate flow rate. Totally, there were 1200 nozzles made of 416 stainless steels in the 2.5×2.5 m duct. The DI (deionized) water flow rate was 6 tons/hr, including 1.5 tons/hr overflow. The ion concentrations of DI at the inlet and the outlet of the fine water spray device were measured. The results showed that the quality of the DI water was quite good, and most of the data were below the method detection limit. The method detection limit (MDL) was 0.039, 0.041, 0.014, 0.067, 0.093, and 0.023 μg/mL in terms of liquid phase concentration, for F^-, Cl^-, NO_2^-, NO_3^-, SO_4^{2-}, and NH_4^+, respectively.

The HDS (honeycomb denuder/filter pack system) samplers were placed at the inlet (mark 1, in Fig. 1) and outlet (mark 2, in Fig. 1) of the fine water spray device for simultaneous sampling of 24 hr. The six samples were taken from January 2000 to January 2001. The air sampling flow rate was 10 L/min.
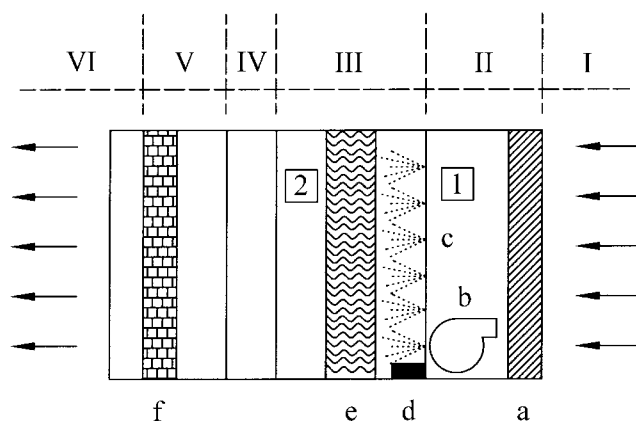
Figure 1. Schematic representation of the air-conditioning system for the make-up air: a, prefilter; b, fan; c, spray nozzle; d, DI water container; e, demister; f, HEPA filter. 1, inlet sampling point; 2, outlet sampling point. I, incoming air; II, prefiltration section; III, fine water spray section; IV, heating section; V, high-efficiency filtration section; VI, to clean room.

The components of the HDS included an impactor with the 50% cut size of 2.5 μm, a glass-transition section, two honeycomb denuders, a spacer, and a filter pack.[8,9] The gas collection section of the HDS was a glass honeycomb consisting of 212 cylindrical tubes, and the inner diameter and length of each tube was 0.2 and 3.8 cm, respectively. The first denuder was coated by using 1% (w/v) sodium carbonate, 1% (w/v) glycerol in 1:1 (v/v) water/methanol solution to collect acid gases, such as SO_2, HNO_3, and HNO_2. To collect ammonia gas, 2% (w/v) citric acid/1% (w/v) glycerol solution was used for the second denuder. A three-stage filter pack was placed downstream of the denuders. The filter pack consisted of a Teflon filter (Gelman Science, 2-μm pore size) to collect fine particles, a nylon filter (Gelman Science, 1-μm pore size) to collect HNO_3 and HCl, and a glass fiber filter (AP40, Millipore Inc.) coated with citric acid to collect NH_3. Particulate NO_3^- and Cl^- concentrations were determined as the sum of those collected on the Teflon filter and the nylon filter.

After sampling, the denuders and the glass fiber filters were extracted with 10 mL of deionized water. The Teflon filters were cut into six pieces and placed inside the extracting bottle. The filters were wetted by adding 0.2 mL of ethanol and then extracted with 10 mL of deionized water. The filters within extracting bottle were kept for 20 min with rotating at an angle of 90° in every 5 min. The nylon filters were extracted with 10 mL of anion eluent

(1.8 mM Na_2CO_3 and 1.7 mM $NaHCO_3$) to extract Cl^- and NO_3^- efficiently. The extracts were kept in a refrigerator at 4°C for further analysis. Finally, soluble ions such as F^-, NO_3^-, NO_2^-, Cl^-, SO_4^{2-}, and NH_4^+ were analyzed by ion chromatography (Model 4500i, Dionex Corp., CA, USA) following the method described in Sioutas et al.[9] In this study, QA/QC procedure includes establishment of the calibration curve using standard solutions and method detection limit (MDL), blank analysis, and repeated analysis. The method detection limit was 0.036, 0.021, 0.005, 0.018, 0.017, and 0.021 ppb for HF, HCl, HNO_2, HNO_3, SO_2, and NH_3 gases, respectively, based on 24-hr sampling at 10 L/min. The blank values of HDS samples for all ion species were non-detectable. The results of precision analysis showed that the relative bias of detected concentrations was below 5%.

RESULTS AND DISCUSSION

The concentrations of HF, HCl, HNO_2, HNO_3, SO_2, and NH_3 at the inlet and at the outlet of the fine water spray device were found to be 0.9 ± 0.5, 1.0 ± 0.6, 2.1 ± 0.9, 0.3 ± 0.2, 3.1 ± 1.4, and 13.2 ± 6.9, and 0.3 ± 0.1, 1.0 ± 0.6, 0.4 ± 0.3, 0.2 ± 0.1, 0.4 ± 0.2, and 3.6 ± 2.5 (average $\pm$ standard deviation) ppb, respectively. The scattering of results might be due to the variations of external parameters including weather conditions, volatilization from the particulates, and operating parameters of the make-up air system. The removal efficiencies of the fine water spray device were found to be not good for the low concentration pollutants existing in the ambient air. It is probably due to the volatilization of HCl, HNO_3, and NH_3 gas-phase pollutants from NH_4NO_3 and NH_4Cl particulate in the intake air resulting in the appearance of gas pollutants in the outlet.

The inorganic particulate concentrations of air pollutants, i.e., Cl^-, NO_3^-, SO_4^{2-}, and NH_4^+ at the inlet and at the outlet of the equipment were found to be 1.5 ± 1.8, 1.1 ± 1.3, 5.4 ± 4.9, and 3.5 ± 1.8, and 0.9 ± 0.8, 0.9 ± 1.3, 3.4 ± 3.0, and 2.4 ± 1.3 (average $\pm$ standard deviation) $\mu g/m^3$, respectively.

Figure 2(a)–(f) show the removal efficiency of the fine water spray equipment for gas and particulate pollutants vs. sampling number. The removal efficiency of the device for gas pollutants, i.e., HF, HCl, HNO_2, HNO_3, SO_2, and NH_3 is calculated to be 61.4 ± 23.4, 18.1 ± 17.4, 76.4 ± 18.5, 48.9 ± 18.3, 85.7 ± 7.8, and $66.2 \pm 22.3\%$, respectively. The more soluble gases, such as HNO_3, SO_2, and NH_3, have higher removal efficiency than less soluble gases. The efficiency for inorganic particulates (aerodynamic diameters $< 2.5 \, \mu m$) is considerably lower than that of

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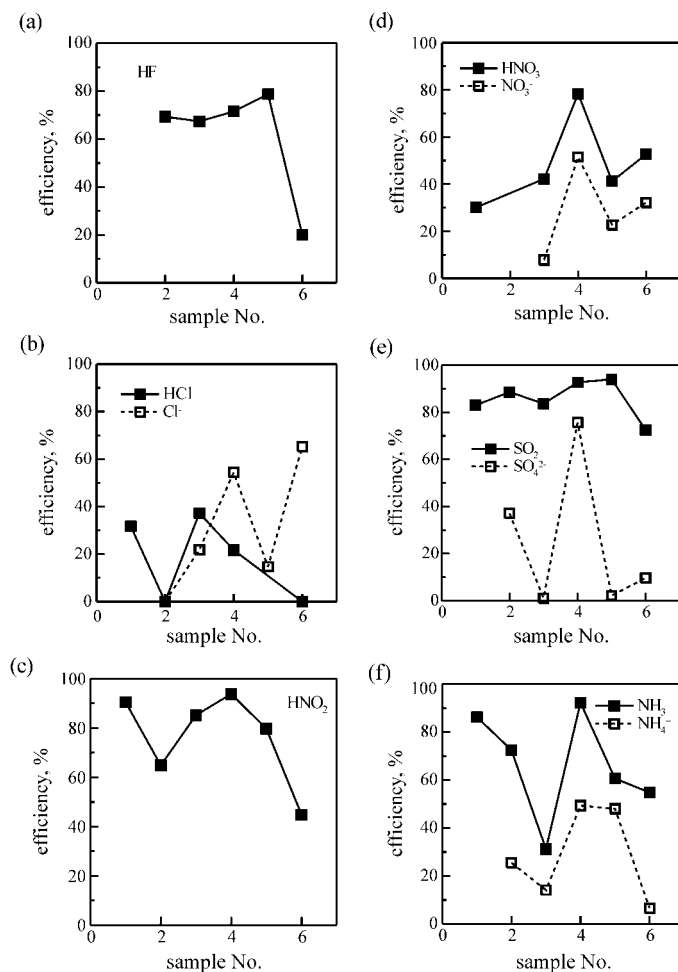
Figure 2. Removal efficiency of the device for the gas and inorganic particulate pollutants vs. sampling number.

inorganic gases. For particulate Cl^-, NO_3^-, SO_4^{2-}, and NH_4^+, the efficiency is 31.2 ± 27.5, 22.8 ± 20.3, 25.1 ± 31.7, and $28.7 \pm 19.4\%$, respectively. The results indicate that the removal efficiency of the fine water spray device for the gas pollutants is higher than that for the fine particulates due to more soluble nature of acidic/basic gases in water than particulates. The fine spray water cannot remove efficiently fine particulate due to their small inertia.

However, these fine particles can be further removed efficiently by the following HEPA filter in the air conditioning system (section V, Fig. 1).

CONCLUSION

Gas and particulates presented in the ambient air of a semiconductor plant can be removed with moderate efficiency by the low-cost fine water spray device. In this study, the concentrations of pollutants were measured at the inlet and the outlet of the fine water spray device by using HDS samplers. The removal efficiency of the device was found to be higher for gas pollutants than for fine particulates. However, the fine particulates can be controlled by the HEPA filter before entering the clean room. More soluble gases were found to have higher removal efficiency than less soluble gases.

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