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An Efficient Venturi Scrubber System to Remove Submicron Particles in Exhaust Gas

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ABSTRACT

An efficient venturi scrubber system making use of heterogeneous nucleation and condensational growth of particles was designed and tested to remove fine particles from the exhaust of a local scrubber where residual $\mathrm{SiH_4}$ gas was abated and lots of fine $\mathrm{SiO_2}$ particles were generated. In front of the venturi scrubber, normal-temperature fine-water mist mixes with high-temperature exhaust gas to cool it to the saturation temperature, allowing submicron particles to grow into micron sizes. The grown particles are then scrubbed efficiently in the venturi scrubber. Test results show that the present venturi scrubber system is effective for removing submicron particles. For $\mathrm{SiO_2}$

IMPLICATIONS

Many air pollution control devices for gas pollutant treatment do not remove fine particles efficiently. In this study, an efficient venturi scrubber with the help of fine-water mist was used to remove fine particles from the exhaust gas of a local scrubber which is mainly used to abate residual SiH₄ gas in the semiconductor industry. The study shows that nucleation and condensation growth of fine particles indeed increase the removal efficiency of the venturi scrubber considerably with only a moderate pressure drop. Same level of particle removal efficiency can't be achieved with the traditional venturi scrubber without the expense of a very large pressure drop. Hence this highly efficient venturi scrubber system is very useful for fine particle control in the semiconductor industry.

particles greater than 0.1 μ m, the removal efficiency is greater than 80–90%, depending on particle concentration. The corresponding pressure drop is relatively low. For example, the pressure drop of the venturi scrubber is $\sim 15.4 \pm 2.4$ cm H_2O when the liquid-to-gas ratio is 1.50 L/m³. A theoretical calculation has been conducted to simulate particle growth process and the removal efficiency of the venturi scrubber. The theoretical results agree with the experimental data reasonably well when SiO_2 particle diameter is greater than 0.1 μ m.

INTRODUCTION

The separation of fine particles from exhaust gas is an important gas cleaning process. Venturi scrubbers are efficient wet collectors for particle control in which the particle collection efficiency depends largely on pressure drop. In the literature, extensive researches have been made on the separation efficiency and the pressure drop of venturi scrubbers. $^{1-4}$ To remove particles greater than 1 μ m, the pressure drop of the scrubber is still reasonable. However, to remove submicron particles, the required pressure drop (or running cost) is excessively high. For example, to achieve the cutoff aerodynamic diameter as low as 0.5 μ m, the pressure drop must be greater than 200 cm H_2O when the liquid-to-gas ratio is 1 L/m 3 .5

In the semiconductor industry, local scrubbers (point-of-use abatement devices) are normally installed right after the manufacturing processes such as dry etching, ion implantation, thermal diffusion, and chemical vapor deposition to treat residual gases.^{6–8} But fine particles generated in the process chambers or high-temperature reaction chambers of the local scrubbers are usually not treated well. Fine particles exist the local scrubbers will enter the central scrubbers and emit from the stacks. Sometimes, these particles may clog the exhaust tubes after the local scrubbers. Not only these fine particles pollute the environment, they are also the sources of white smoke.⁹ Therefore, it is necessary to remove fine particle from the exhaust gas of the manufacturing processes.

In this study, a venturi scrubber system to treat fine particle exhaust of a local scrubber was designed and tested. The local scrubber has an electrical heater working at 850° C for SiH₄ gas abatement where residual SiH₄ gas is oxidized at high temperature while a large number of SiO₂ particles are generated. A three-stage packed tower after the reaction chamber of the local scrubber is originally designed to remove particles but was found to have very low collection efficiency, <10%, in this study. In the present efficient venturi scrubber system, a fine-water mist was added to quench high-temperature exhaust gas to a temperature low enough for the gas to reach supersaturation condition. Submicron particles will grow because of heterogeneous nucleation and condensation to micron size particles, and are removed efficiently by the venturi scrubber.

In the literature, the rate of fine particle growth in a super-saturated atmosphere (atm) of water vapor was studied from both theoretical and experimental pointsof-view.¹⁰ Their results showed that the rate of particles growth undergoing condensation was very rapid, and that the volume-mean diameter of grown particles was determined by the number concentration of particles and the initial state of super-saturation in the surrounding gas. An efficient multistage process that removed a wide range of fine particle assisted by the nucleation method had been investigated and discussed. 11,12 In their methods, exhaust gas stream was mixed with super-saturated steam in a chamber, water vapor condensed on the particles that would then grow to bigger sizes in a subsequence cooling chamber. The grown particles were removed by a cyclone after the gas flow was accelerated through a nozzle. Test results showed that steam mixing and subsequent cooling were useful to increase fine particle removal efficiency.

In this study, the submicron particle collection efficiency of the current efficient venturi scrubber system was also compared with the traditional venturi scrubber. The influences of the liquid-to-gas ratio and the amount of residual ${\rm SiH_4}$ gas (or generated ${\rm SiO_2}$ concentration) on the fine particle removal efficiency were also investigated. A theoretical calculation was also conducted to simulate the

particle removal efficiency by the venturi scrubber. The theoretical results were then compared with the experimental data of present study.

EXPERIMENTAL METHODS

Figure 1 illustrates the schematic diagram of the experimental system. The waste SiH4 gas and dilution air are injected into a reaction chamber containing an electrical heater operating at 850° C. Residual SiH₄ gas is abated because of high-temperature oxidation, in which a lot of SiO₂ particles are generated. In the experiment, SiH₄ gas was supplied by a stainless steel bottle and the flow rate was controlled by a MFC (Mass Flow Controller, AE Corp., FC-770AC; Fort Collins, CO). For the test, 1% SiH₄ of 0.5 L/min and 1 L/min were used. Figure 2 illustrates the schematic diagram of the venturi used in this study. The diameter of the throat is 1 cm and its length is 3 cm. The total flow rate passing through the venturi scrubber was fixed at 180 L/min by using an orifice meter, and the velocity at the throat was calculated to be 38.22 m/s. The ratio of the steam to dry air mass flow rate (or mixing ratio) was fixed at 0.09, and the scrubber liquid-to-gas ratio was 1.17, 1.50, 1.80 L/m³, respectively. The scrubber liquid was injected tangentially at the entry of the venturi throat and the flow rate was controlled by a rotameter. Exhaust gas exiting the reaction chamber was measured to be $250 \pm 20^{\circ}$ C.

A SMPS (Scanning Mobility Particle Sizer, TSI, Model 3934) system was used to measure the particle removal efficiency of the venturi scrubber. It consists of an Electrostatic Classifier (TSI, Model 3071; St. Paul, MN), a CPC (Condensation Particle Counter, TSI, Model 3022), and computer software. In this study, the sampling tube of the SMPS was placed at the inlet and the outlet of the venturi scrubber for the size distribution measurement of fine particles ranging from 50–478 nm in diameter. The removal efficiency of a certain particle size was calculated from the difference between the outlet and inlet number concentration divided by the inlet number concentration of the corresponding particle size. The fine-water mist

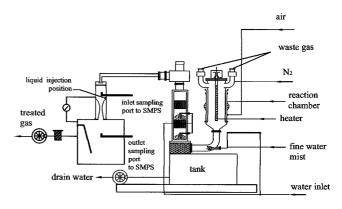


Figure 1. Schematic diagram of the experimental system.

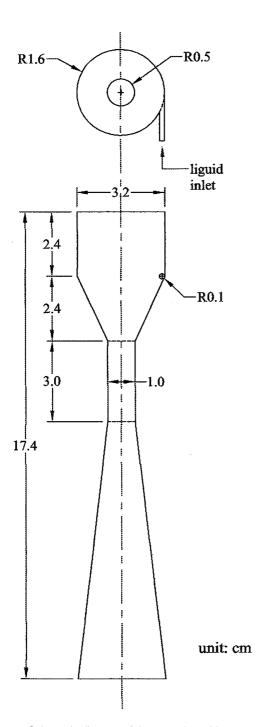


Figure 2. Schematic diagram of the venturi scrubber.

was sprayed from a nozzle (Type M10 nozzle, Hago Manufacturing Corp., Mountainside, NJ) to quench the exhaust gas to $\sim 48 \pm 2^{\circ}$ C. The particle removal efficiency of the venturi scrubber was also determined when the fine mist spray was turned off, simulating the case of the traditional venturi scrubber. A differential pressure gauge was also set up to measure the pressure drop of the venturi scrubber.

THEORY OF PARTICLE REMOVAL EFFICIENCY

Theoretical calculation was conducted to investigate the effect of mixing high temperature exhaust with fine-water

mist on particle growth and particle removal efficiency of the venturi scrubber. When the exhaust gas is mixed with fine-water mist that is assumed to evaporate completely, the mixing ratio (or the ratio of the steam to dry air mass flow rate) can be expressed as follows:

$$mixing \ ratio = \frac{M_{w2}}{M_{a1}(1+w_1)} \tag{1} \label{eq:mixing}$$

where M_{a1} (Kg/sec) represents the mass flow rate of dry air in the exhaust gas; Mw2 (Kg/sec) is the mass flow rate of water vapor; and w is the absolute humidity of the exhaust gas (subscript "1").

After mixing, the absolute humidity of the mixed gas, w₃, can be expressed as follows:

$$w_{3} = \frac{M_{a1}}{M_{a3}} w_{1} + \frac{M_{a2}}{M_{a3}} w_{2} = \frac{M_{a1}}{M_{a1} + M_{a2}} \cdot w_{1} + \frac{M_{a2}}{M_{a1} + M_{a2}} \cdot w_{2}$$

$$(2)$$

where w_2 is the absolute humidity of water vapor, M_{a2} (Kg/sec) and M_{a3} (Kg/sec) are the mass flow rate of dry air in the water vapor and in the mixed gas, respectively.

The temperature of the mixed gas can be derived from the energy balance equation, and can be expressed as:

$$T_{3} = \frac{M_{a1} \cdot (C_{pa} + w_{1} \cdot C_{pg}) \cdot T_{1} + M_{w2} \cdot C_{pg} \cdot T_{2} - M_{w2} \cdot h_{fg2}}{M_{a1} \cdot (C_{pa} + w_{1} \cdot C_{pg}) + M_{w2} \cdot C_{pg}}$$

$$(3)$$

where $C_{\rm pa}~(1002.1 \cdot J/kg \cdot K)$ and $C_{\rm pg}~(1762.1~J/kg \cdot K)$ are specific heat of air and water vapor, respectively; h_{fg2} (J/Kg) is the evaporation latent heat of water.

After mixing, the saturation ratio, S, can be derived from the following equation:

$$S = \frac{W_3 \cdot P_m V_g}{R_a \cdot T_3 + W_3 P_w V_\sigma} = \frac{W_3 \cdot P_m \cdot V_g}{(R_a + W_3 \cdot R_\sigma) \cdot T_3}$$
(4)

where R_a (286.9865J/kg·K) is the gas constant of air; and $R_{\rm g}~(461.5043J/kg{\cdot}K)$ is the gas constant of water vapor. $P_{\rm m}$ (Pa) is the total pressure; T_3 (K) is the temperature; P_w (Pa) is the saturation pressure of water vapor at temperature T_3 ; and V_g (m³/kg) is the specific volume of water vapor at temperature T_3 .

The variation of particle diameter with time can be described by the following equation:13

$$\begin{split} \frac{dD_p}{dt} &= \frac{4 \cdot D \cdot M_w}{R \cdot \rho_w \cdot D_p} \left(\frac{P_s}{T} - \frac{P_d}{T_d} \right) \\ &\times \left(\frac{0.75 \cdot \alpha \cdot (1 + Kn)}{0.75 \cdot \alpha + 0.283 \cdot Kn \cdot \alpha + Kn + Kn^2} \right) \ (5) \end{split}$$

where $D_{\rm p}$ (m) is the particle diameter; t (sec) is time; and $\rho_{\rm w}$ (1000 kg/m³) is the density of water; D (m²/sec) is the air-water vapor diffusion coefficient at temperature T; $M_{\rm w}$ (18.015 g/mole) is the molecular weight of water; R (8.314 N·m/mole·K) is the ideal gas constant; $P_{\rm s}$ (N/m²) is the partial pressure of water vapor in the mixed gas; $P_{\rm d}$ (N/m²) is the water vapor pressure on the particle surface; T (K) and $T_{\rm d}$ (K) are the mixed gas temperature and particle surface temperature; Kn is the Knudsen number and α (between 0 and 1) is the accommodation coefficient.

The fourth-order Runge-Kutta method was used to integrate the particle growth equation, eq 5. In a time step, Δt , the particle grows from the initial diameter $D_{\rm p1,i}$ to $D_{\rm p2,i}$, and the mass of water condensed per unit gas volume on the particle surface equals the reduced mass of water vapor in the unit gas volume of the mixed gas, and can be shown to be:

wloss =
$$\sum_{i} \mathbf{N} \cdot \mathbf{f}_{i} \cdot \frac{\pi}{6} \cdot \rho_{w} \cdot (D_{p2,i}^{3} - D_{p1,i}^{3})$$
 (6)

where wloss (kg/m^3) is the reduced mass of water vapor per unit gas volume; f_i is the fraction of particles with the diameter of $D_{p1,i}$; N (particles/m³) is the total number concentration.

After a time step, the absolute humidity and temperature of the mixed gas was reduced to w_4 and T_4 as:

$$w_4 = \frac{M_{a3} \cdot w_3 - wloss \cdot Q}{M_{a3}} = w_3 - \frac{wloss \cdot Q}{M_{a3}}$$
 (7)

$$T_4 = T_3 + \frac{wloss \cdot Q \cdot h_{fg3}}{M_{a3} \cdot C_{pa} + M_{a3} \cdot w_4 \cdot C_{pg} + wloss \cdot Q \cdot C_{pw}}$$
 (8)

where Q ($\rm m^3/sec$) is the volumetric flow rate after mixing process, $\rm h_{fg3}$ (J/Kg) is the condensation latent heat of water vapor.

In the next time step, we use w_4 and T_4 as the initial conditions to repeat the above calculation (eqs 5 and 6) until the saturation ratio (S) is less than 1.

The particle removal efficiency of the venturi scrubber was calculated following Calvert's theory (1970)³ as:

$$P = \exp\left[\frac{2}{55} \frac{Q_{\ell}}{Q_{g}} \frac{\rho_{D} D}{\mu_{a}} v_{t} F(K_{pt}, f)\right]$$
 (9)

$$K_{pt} = \frac{\rho_p C_{d_p} d_p^2 v_t}{9\mu_a D} = \frac{2Stk}{1 - u_t}$$
 (10)

$$Stk = \frac{\rho_p C_{d_p} d_p^{\ 2}(v_t - v_D)}{18\mu_a D} \eqno(11)$$

$$\begin{split} F(K_{pt},f) &= \frac{1}{K_{pt}} \bigg[-0.7 - K_{pt}f + 1.4 ln \bigg(\frac{K_{pt}f + 0.7}{0.7} \bigg) \\ &+ \frac{0.49}{0.7 + K_{pt}f} \bigg] \end{split} \tag{12}$$

where P is the penetration of particles with the diameter d_p ; f is an empirical factor; Q_l and Q_g (m³/sec) are volumetric flow rates of liquid and gas, respectively; μ_a (N·S/ M^2) is the dynamic viscosity of air; V_t and V_D (m/sec) are the velocities of gas and droplet at the venturi throat, respectively; K_{pt} is a dimensionless parameter; Stk is the Stokes number; d_p (m) is the diameter of the particle; C_{dp} is the slip correction factor; u_t is the velocity ratio of the droplet and gas at the throat; ρ_D and ρ_p (kg/m³) are the densities of the droplets and particles, respectively; D (μ m) is the Sauter mean diameter of droplets, which can be derived from Nukiyama-Tanasawa (1938) equation as:14

$$D = \frac{585}{v_{rel}} \sqrt{\frac{\gamma}{\rho_D}} + 597 \left[\frac{\mu_D}{\sqrt{\gamma \rho_D}} \right]^{0.45} \left[\frac{1000Q_\ell}{Q_g} \right]^{1.5}$$
 (13)

where $V_{\rm rel}$ (m/sec) is the relative velocity of gas and liquid at the nozzle; γ (dyne/cm) is the surface tension of liquid; $\mu_{\rm D}$ (dyne-sec/cm²) is the dynamic viscosity of liquid.

RESULTS AND DISCUSSION Pressure Drop

Pressure drop is one of the important operation parameters for the venturi scrubbers. Figure 3 shows the experimental data for the relationship between the pressure drop and the liquid-to-gas ratio, which is 1.17, 1.50, and

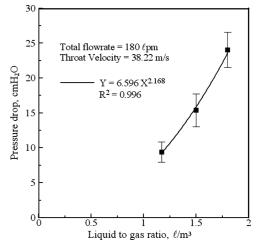


Figure 3. The relationship between pressure drop and liquid-to-gas ratio.

1.80 L/m³. The gas velocity is 38.22 m/sec at the throat corresponding to a fixed total flow rate of 180 L/min. It is seen that as the liquid-to-gas ratio is increased, the pressure drop also increases. The pressure drop is 9.4 ± 1.4 , 15.4 ± 2.4 , 24 ± 2.5 cm H₂O for the Q₁/Q_g = 1.17, 1.50, and 1.80 L/m³, respectively.

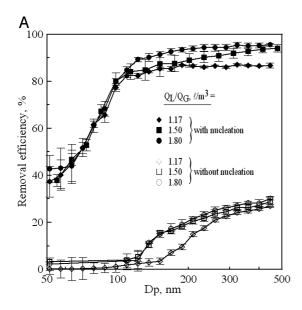
Fine Particle Removal Efficiency with or without Nucleation and Growth

When SiH₄ gas of different flow rates is abated in the high temperature reaction chamber, a lot of SiO₂ particles are produced that are to be removed by the venturi scrubber. The removal efficiency of fine particles was measured at the total flow rate of 180 L/min. Figure 4a, b show the fine particle removal efficiencies at different liquid-to-gas ratios for 1% SiH₄ flow rate of 0.5 L/min and 1 L/min, respectively. The removal efficiency without fine-water mist (or without nucleation and condensational growth) simulating the case of a traditional venturi scrubber is also shown for comparison. It is seen that the particle removal efficiency by the venturi scrubber system with fine-water mist is much greater than that without fine-water mist. The results indicate that fine-water mist is effective to enhance fine particle removal efficiency. That is, finewater mist is able to quench high temperature exhaust gas to reach the super-saturation condition so that fine particles grow to bigger sizes thus the efficiency is improved. The influence of the liquid-to-gas ratio on the particle removal efficiency is not obvious for the present efficient venturi scrubber system. In comparison, the particle removal efficiency is increased slightly as the liquid-to-gas ratio is increased for the case without nucleation and growth.

The particle removal efficiency by the venturi scrubber without nucleation is found to be low, typically below 30%. In this case, particle removal efficiency increases with an increasing particle diameter. For the case with fine-water mist and low SiH₄ mass flow rate (Figure 4a), the increase in efficiency with particle diameter is more obvious. For example, Figure 4a shows that the removal efficiency increases from \sim 40% at dp = 50 nm to 80% at dp = 100 nm monotonically. Then the efficiency gradually increases to $84 \sim 95\%$ at dp = 478 nm depending on the liquid-to-gas ratio. The figures also show that the removal efficiency of the larger SiH₄ flow rate (Figure 4b) is higher than those of the smaller SiH₄ flow rate (Figure 4a). For example, the max particle removal efficiency with nucleation is 84% and 96%, respectively, for 1% SiH₄ of 0.5 and 1 L/min flow rates, at the liquid-to-gas ratio of 1.17 L/m³. At the inlet of the venturi scrubber, the total number concentration (TNC), number median diameter (NMD), and geometric SD (GSD) were measured by the SMPS to be: TNC = 3.22×10^5 #/cm³, NMD = 271 nm, GSD = 2.54 for 1% SiH₄ of 0.5 L/min; TNC = 3.15×10^6 $\#/\text{cm}^3$, NMD = 263 nm, GSD = 2.85 for 1% SiH₄ of 1 L/min.

Comparison of Theoretical Results with Experimental Data

The experimental data suggest that heterogeneous nucleation and condensational particle growth by fine-water mist in front of the venturi scrubber is effective for fine particle removal. Theoretical calculation was conducted to show how the mixing process affects particle growth and removal efficiency of the venturi scrubber. The calculation assumed that fine-water mist would evaporate completely and became available for condensation on the



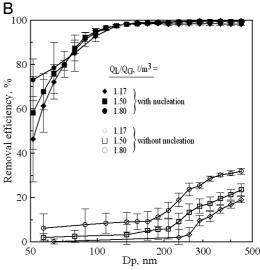


Figure 4. The relationship between particle removal efficiency and particle diameter, (a) 1% SiH₄ of 0.5 L/min, and (b) 1% SiH₄ of 1 L/min.

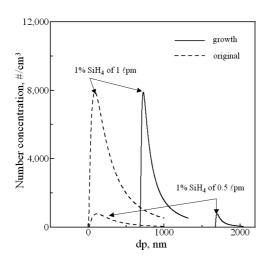
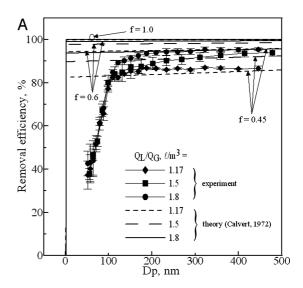


Figure 5. The particle size distribution after particle condensation growth at the exhaust gas temperature of 250° C.

fine particle surfaces, and there was no water vapor loss on the wall. The accommodation coefficient was assumed to be 1. Figure 5 shows that the original particle size distribution and the calculated particle size distribution at the exhaust gas temperature of 250° C for 1% SiH_4 flow rate of 0.5 L/min and 1 L/min. It is seen that the mixing process will help the particle growth. The figure also shows that as the initial particle concentration decreases, the final particle diameter increases.

Figure 6a, b show the comparison between the theoretical results and the experimental data of the particle removal efficiency for the scrubber with condensation growth. The figures show that the theoretical particle removal efficiencies increase with an increasing empirical factor f. The theoretical particle removal efficiencies for particles greater than 100 nm calculated by Calvert's theory with the empirical factor of 0.45 agree best with the experimental data for 1%, 0.5 L/min of SiH₄, as shown in Figure 6a. For particles less than 100 nm, the theoretical results over-predict the collection efficiency. For a larger amount of SiH₄ gas such as 1%, 1 L/min of SiH₄ (shown in Figure 6b), the empirical factor in Calvert's theory must be assumed to be 1 for the best fit of the experimental data. In this study, the empirical factor f was used to adjust the effects of particle's hygroscopic behavior, particle concentration, and coagulation occurred between droplets and fine particles on the theoretical particle removal efficiency. As stated before, the number concentration of SiO₂ particles and initial particle diameter increase with an increasing SiH₄ gas concentration in the exhaust gas. For a fixed amount of water mist, theoretically the saturation ratio will reduce with the elapse of time as water vapor condenses on growing particles. The decrease in the saturation ratio is more obvious for particles with high concentration, resulting in a smaller final particle size. Consequently, the theoretical particle removal efficiency for high concentration particles should decrease. But on the



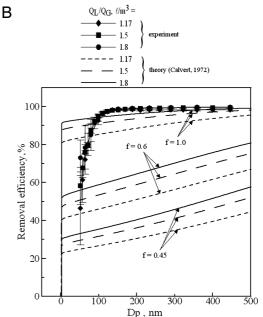


Figure 6. The comparison of the theoretical results of particle removal efficiency with experimental data with nucleation by water mist. (a) One percent SiH_4 of 0.5 L/min, and (b) 1% SiH_4 of 1 L/min.

contrary, the experimental particle removal efficiency does not change too much for higher SiO₂ concentration as shown in Figure 6b. This may be attributed to particle coagulation effect at high SiO₂ particle concentration, which increases the particle removal efficiency. In this study, the empirical factor f in Calvert's theory was used to adjust this effect and it is 0.45 and 1, for lower and higher SiO₂ particle concentration, respectively.

CONCLUSIONS

This study has investigated the fine particle removal efficiency of an efficient venturi scrubber system incorporating fine-water mist to quench high temperature exhaust gas so that heterogeneous nucleation and condensational growth occur for submicron particles. The grown particles

are then scrubbed efficiently by a venturi scrubber at high collection efficiency and low-pressure drop. When SiO₂ particles are greater than 0.1 µm, the removal efficiency is greater than 80–90% at the pressure drop of 15.4 ± 2.4 cm H₂O when the liquid-to-gas ratio is 1.50 L/m³. The influences of the pressure drop, the liquid-to-gas ratio, and the amount of SiH₄ gas on fine particle removal efficiency by the venturi scrubber system have been investigated. With fine-water mist for nucleation and growth, test results show that the max removal efficiency achieved for particles at 478 nm in diameter is much greater than the traditional venturi scrubber. It is 96% for 1% SiH₄ of 1 L/min at the liquid-to-gas ratio of 1.17 L/m³. The influence of the liquid-to-gas ratio and the amount of SiH4 gas on the removal efficiency is shown to be not significant. In comparison, the removal efficiency of the traditional venturi scrubber is typically less than 30% in this study. A theoretical study has been conducted to simulate particle growth and removal efficiency. For lower particle concentration and when particle diameter is larger than 100 nm, the agreement between theory with the empirical factor of 0.45 and experiment is good. For higher number concentrations of SiO₂ particles, the empirical factor in Calvert's theory was assumed to be 1 for the best fit of the experimental data.

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