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Honeycomb wet scrubber for acidic gas control: modeling and long-term test results

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Abstract

A laboratory scale, 1.0 CMM ($\text{m}^3 \text{min}^{-1}$) wet scrubber packed with water-absorbing honeycomb material (HWS) with a very large geometric surface area of $480 \text{ m}^2 \text{m}^{-3}$ and a low pressure drop developed in our previous study was shown to achieve a very high removal efficiency for acidic gases but there were no long-term test data. In this study, the HWS scaled up to operate at a 100 CMM flow rate was tested for removing mixed acidic gases at a semiconductor fab for a very long period of 3.5 yr. Results showed that the removal efficiency for the mixed gases emitted from the fab always maintained as high as $> 95\%$ for HF, CH_3COOH , HCl, HNO_3 , HNO_2 , and H_2SO_4 with the inlet concentrations ranging from supper-ppmv to sub-ppmv, during a 3.5-yr period. With water jet cleaning of the honeycomb modules once per year, the pressure drop of the HWS remained to be low at 0.5–0.8 cm H_2O , indicating minimal scaling in the HWS. Additionally, the predicted height and removal efficiencies of the HWS were very close to the experimental data. The excellent long-term performance of the HWS warrants its potential applications in many areas in which liquid absorption is the preferred treatment method and the theoretical equations can facilitate the design of the HWS.

Keywords: Honeycomb, Wet scrubber, Acidic gas, Long-term operation, Pressure drop

Introduction

Inorganic acids such as HF, HCl, HNO_3 , H_2SO_4 , and CH_3COOH are widely used in semiconductor or photo-electric manufacturing factories (or fabs) for wafer cleaning and wet-etching processes [1–5] resulting in the emission of gaseous inorganic acidic pollutants from the stacks, which need to be treated to meet the emission standard of Taiwan Environment Protection Agency. The standard stipulates that the total mass emission rate from all stacks must be $< 0.6 \text{ kg h}^{-1}$ for HF, HCl, and HNO_3 , and $< 0.1 \text{ kg h}^{-1}$ for H_2SO_4 , for a semiconductor fab, and the removal efficiency (RE) for each species

must be $\geq 95\%$ [1]. Among these gaseous pollutants, HF and HCl are considered as hazardous air pollutants [6]. These gases are also precursors for secondary inorganic components in $\text{PM}_{2.5}$ [7] which is considered as a major health-risk concern in many countries [8]. Wet scrubbers (WSs) are widely used to control the emission of acidic gases [9–11] due to their small footprint, simple structure, and low capital cost [12]. However, the measured removal efficiency (RE) of most traditional WSs cannot meet the required value of greater than 95% [1]. For instance, the RE of a vertical packed WS with a 1500 CMM ($\text{m}^3 \text{min}^{-1}$) airflow rate were 71–93% for HF, HCl, HNO_2 , HNO_3 , and CH_3COOH at a high-tech factory while the RE of a lab-scale packed WS with a 0.6–1.8 CMM airflow rate was found to be as low as 77–86% for H_2SO_4 mist [13]. When the RE of the WSs cannot meet the standard value, they must follow the design

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criteria and operating conditions in which the specific surface area of packing materials must be $\geq 90 \text{ m}^2 \text{ m}^{-3}$, the residence time through the scrubbing section must be $\geq 0.5 \text{ s}$, a wetting factor must be $\geq 0.1 \text{ m}^3 \text{ h}^{-1}$, and the pH of scrubbing liquid must be ≥ 7 as shown in Table 1 [1].

Among all operating parameters, the inlet concentration was found to be the most important parameter affecting the RE of the packed WS. The RE of a conventional packed WS increased with increasing inlet concentration at low inlet concentration (C_{in}) and approached a constant value until C_{in} reached a certain value for HF, HCl, HNO_3 , HNO_2 , and CH_3COOH [14]. For example, the RE of a multistage dual-flow sieve plate WS was 93.8% as C_{in} was as high as 100–300 ppmv for HCl [15] while a packed tower was found to have a RE of less than 90% when C_{in} was lower than 1.0 ppmv for HF and HCl [9, 14]. The WSs normally have high pressure drop resulting in high power consumption and operation cost [13, 16]. For example, a WS installed mesh MV filters operated at a 90 CMM flow rate achieved > 95% RE for NH_3 (L/G of 2.0 L m^{-3}) at a high pressure drop of > 1411 Pa [17]. However, there were no long-term test data provided. Therefore, it is a challenge to design a WS with a high RE and low pressure drop which lasts for a long time in an actual plant.

The WSs are normally packed with random packing materials to enhance RE. The packing materials, which can be in the form of a saddle, ring, or tellerette and made of ceramics, glass, metal, porcelain, steel, or plastics [18, 19], should have a high specific surface area to maximize the gas-liquid contact surface for mass transfer [20] and liquid holdup. The REs of acidic gases were found to increase when the packing material size decreased [21]. For instance, the RE of HCl was increased by 2.0–3.5% when the specific surface area of packing balls was increased from 150 to $224 \text{ m}^2 \text{ m}^{-3}$ [22]. The packing materials also need to resist fouling and corrosion, avoid particle clogging, and enhance water

retention to minimize the pressure drop and maintenance cost for long-term use. In addition, they need to provide good air distribution for uniform absorption [13] and low pressure drop in the WSs. The pressure drop of a WS with conventional plastic packing materials is about 413–830 Pa (4.2–8.5 cm H_2O) per meter of the packing column [23–25].

The RE can also be improved by increasing the L/G ratio due to the increased gas-liquid contact surface area. The RE could be increased from 83 to 98% as the L/G ratio was increased from 1.5 to 3.0 L m^{-3} [26]. However, a higher L/G ratio will cause a higher pressure drop [11, 23]. Adding surfactants in the scrubbing liquid can help enhance the RE of the WSs due to the charge generation on the water surface by surfactants but the surfactants are costly [10]. The overall efficiency of a packed WS for NH_3 , HF, and HCl ($C_{\text{in}} = 0.2\text{--}3 \text{ ppmv}$) was reported to increase from 56 to 83% at a 1.1 s residence time and 10 CMM flow rate when surfactants were added [9, 10]. The pH of the scrubbing solution is another factor that affects the RE of the WSs for some weak acidic gases such as CH_3COOH but not for strong acidic gases such as HCl [11]. There is no significant effect on the RE of CH_3COOH when the pH is higher than 7.5 due to the gas dissolution limit while the RE of HCl and HNO_3 is nearly constant for the pH ranging from 7 to 9 [11, 27]. To achieve good efficiency for mixed acidic gases and save the operating cost, the pH of scrubbing liquid should be maintained > 7.5.

Most of the previous researches have focused on enhancing the RE of WSs by determining the optimal operating conditions [16, 26, 28], attempting to improve the performance of the existing WSs operating in high-tech industries [9, 10, 22], and designing lab-scale WSs for short-term study only [13, 19, 29]. A WS packed with a multi-parallel-plate (MPP) module (PPWS) developed by our group can achieve very high REs for HCl (> 99%), HNO_3 (> 98%), and CH_3COOH (> 99%), and very low pressure drop (23.5 Pa) at a regulated residence time of 0.5 s, a low inlet concentration of < 3 ppmv, and a high L/G ratio of 18.5 L m^{-3} [11]. It is because that the PPWS uses polypropylene plates coated with nano- TiO_2 packing material with a $327 \text{ m}^2 \text{ m}^{-3}$ specific surface area to enhance the hydrophilicity for scrubbing liquid to form a uniform liquid film. However, this packing material requires a time-consuming and meticulous procedure for preparation and assembly with a precise gap of 3 mm. The pilot-scale 1.0 CMM WS with the honeycomb module (HWS) was further developed with the RE as high as the PPWS and the packing module is easy to fabricate [14]. The HWS showed nearly 100% for HF ($C_{\text{in}} = 0.1\text{--}0.4 \text{ ppmv}$) and 99% for HCl ($C_{\text{in}} = 3.4\text{--}10.8 \text{ ppmv}$) and CH_3COOH ($C_{\text{in}} = 0.7\text{--}6.5 \text{ ppmv}$) at an optoelectronic factory. Similar to the MPP module, the

Table 1 Operating parameters of the HWS at a semiconductor fab and the design criteria of Taiwan Environmental Protection Agency

Operating parameter	Value	Criteria	Unit
^a Wetting factor	0.1	≥ 0.1	$\text{m}^2 \text{ h}^{-1}$
Retention time	0.5	≥ 0.5	sec
Pressure drop	49 ~ 78.4	–	Pa
Airflow rate	100	–	$\text{m}^3 \text{ min}^{-1}$
Liquid flow rate	2600	–	L min^{-1}
pH	7.5 ~ 9.0	≥ 7	–
Packing material	483	≥ 90	$\text{m}^2 \text{ m}^{-3}$

^aWetting factor = scrubbing water flow rate divided by (the specific surface area of the packing materials times the horizontal cross-sectional area of the tower)

honeycomb packing material made of water-absorbing polypropylene (PP) fabric (0.875 kg m^{-1}) has a higher specific surface area ($483 \text{ m}^2 \text{ m}^{-3}$) and lower pressure drop than most of the packing materials as shown in Table 2.

Thus, the honeycomb packing material with a uniform arrangement and 3-mm gap becomes a promising packing material for an efficient WS. But the RE of these small scale HWS and PPWS were obtained when these WSs were run for several days. There is no scale-up, actual HWS which runs in a fab for the long-term to control the soluble gas emission. In this study, the HWS was scaled up as an actual device in a semiconductor manufacturing fab at a 100 CMM flow rate to remove acidic gases and odor. The REs of the HWS for the mixed gases containing HF, HCl, HNO_3 , HNO_2 , H_2SO_4 , and CH_3COOH were tested 13 times over 3.5 years while the pressure drop was also measured to make sure that there was no fouling occurred in the system. The theoretical analysis was conducted to predict the height of the packing module and the RE of the HWS and the traditional WS to further justify the outstanding performance of the HWS.

Materials and methods

Field test

The experimental setup of the HWS installed at a semiconductor manufacturing fab that produces DRAM in Hsinchu, Taiwan to reduce its emission and bad odor of acidic gases is shown in Fig. 1a. The HWS, which is registered as a patent in Taiwan (No. I569867 and M466724), is an add-on central scrubber device mainly to remove high concentration of acidic gas pollutants collected from about 40 local scrubbers before a traditional central packed tower which has low RE with high emitted HF and CH_3COOH concentrations (e.g., typically > 10 ppmv without the present HWS) and bad odor. The HWS, whose schematic diagram is shown in Fig. 1b, consists of 64 honeycomb modules placed in parallel

in four rows (16 modules per row) as shown in Fig. S1 in the Supplementary Materials (SM), spray nozzles installed at the top of honeycomb modules, and a liquid reservoir with conductivity and pH meters. Each honeycomb element was scrolled in a cylindrical casing ($D \times Z = 300 \times 300 \text{ mm}$). The operating parameters of the HWS are shown in Table 1, which meet the design criteria for a packed tower to achieve the efficiency-based emission standards for acidic gases. In this study, the HWS with the specific surface area of the packing material of $480 \text{ m}^2 \text{ m}^{-3}$ was designed to operate at a 100 CMM flow rate with a 0.4 m s^{-1} face velocity and 0.5 s retention time. The liquid flow rate was about 2600 L min^{-1} which was controlled by a flow meter and the L/G ratio was maintained at 26 L m^{-3} . NaOH solution was used as the scrubbing solution with the pH maintained at 7.5–8. The theoretical pressure drop of 59.8 Pa ($0.58 \text{ cm H}_2\text{O}$) was calculated by the method in Chien et al. [11], which is higher than the pressure drop (23.5 Pa) of the lab-scale, 1.0 CMM HWS in the previous study due to the increased L/G ratio.

The HWS was tested for the RE of inorganic gaseous acidic pollutants including HF, HCl, HNO_2 , HNO_3 , H_2SO_4 , and CH_3COOH from August 2016 to March 2020 (3.5 yr) with 13 times (11/08/2016, 12/09, 11/10, 15/11, 13/12, 13/03/2017, 15/06, 15/08, 22/12, 02/05/2018, 31/01/2019, 6/12, and 19/03/2020). The testing frequency was set at once per month in the first 5 months (5 tests) and then extended to once per three months in the next 12 months (4 tests), once per six months (2 tests), and finally once per year (1 test). The latest testing time (19/03/2020) was about 3 months after the 12th test when the pH meter was not functioning well when the NaOH solution was not pumped into the tank to maintain a pH of higher than 7.5. The result of the 12th test is shown in Fig. S2 in Section S1. Two porous denuder samplers (PDS) with a 2 L min^{-1} flow rate [4, 30–32] were used to sample mixture gases at the

Table 2 Some typical packing materials

Packing materials	^a $D \times Z$ or $L \times W_T \times Z$ (cm)	Bulk density (kg m^{-3})	Specific surface area ($\text{m}^2 \text{ m}^{-3}$)	L/G ratio (L m^{-3})	Pressure drop (Pa)	Reference
Honeycomb	30*30	160	483	26	^b 59.8	This study
Honeycomb	30*30	160	483	18.5	23.5	[14]
MPPM	30*30*30	920	327	18.5	23.5	[11]
Polypropylene ball (2.5 cm)	47*40*40	139	379	1.5–3	< 196	[21]
Ceramic intalox saddle (2.5 cm)	30*61	610	250	1.4–2.1	265–1932	[29]
Raschig ring (1.3 cm)	30*61	–	350	1.4–2.1	294–1058	[29]
Polyethylene knit packing	14*10	–	150	3.0	~ 2000	[27]
SS wire mesh packing	14*10	–	500	3.0	~ 2000	[27]

^aPacking dimensions: diameter \times height ($D \times Z$) or length \times width \times height ($L \times W_T \times Z$); ^bTheoretical value

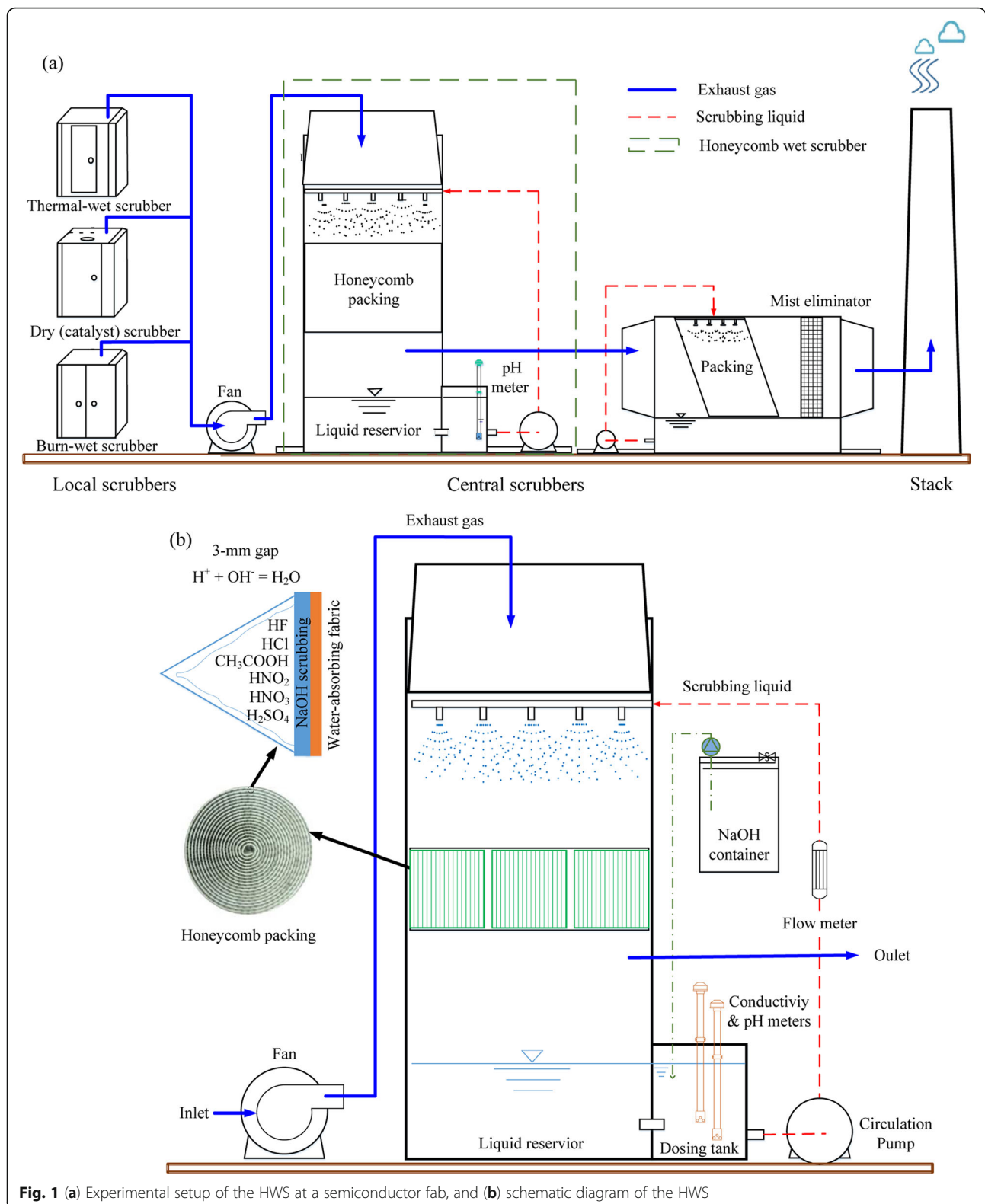


Fig. 1 (a) Experimental setup of the HWS at a semiconductor fab, and (b) schematic diagram of the HWS

inlet and outlet of the HWS simultaneously in 20–60 min depending on inlet concentrations. The PDS consists of a Teflon filter for sampling particles followed by

two porous-metal discs coated with solutions containing 10 mL, 5% (w/v) sodium carbonate, and 1% (w/v) glycerol in 1:1 (v/v) methanol/water solution for sampling

inorganic acidic and basic gases. An ion chromatography (model 883, Metrohm AG, Switzerland) was used to quantify the concentrations of gases and particles. The particulate phase concentration was found to be much smaller than the corresponding gaseous species and is not a target of this study. The RE of the acidic gases was then calculated as:

$$RE(\%) = \left[1 - \frac{C_{out}}{C_{in}} \right] \times 100\% \quad (1)$$

where C_{out} (ppbv) was the outlet concentrations of HF, HCl, HNO_2 , HNO_3 , H_2SO_4 , or CH_3COOH in the gas phase. The makeup water of 4 t d^{-1} was required to compensate for water evaporation and maintain the conductivity of $0.3\text{--}0.5 \text{ mS cm}^{-1}$ in the water reservoir. The pressure drop of the HWS was also measured at five testing times (11/08/2016, 13/12, 15/08/2017, 31/01/2019, and 19/03/2020) during the 3.5-yr period.

Theoretical prediction

The theoretical RE of the HWS is calculated by using Eqs. (2), (3) and (4) based on the convection-diffusion theory of Gormley and Kennedy (RE_{GK} , %) [33, 34] and two film theory (RE_{CA} , %) [18], as follows:

$$RE_{GK}(\%) = [1 - [0.82 \exp(-11.5\xi) + 0.097 \exp(-70.1\xi)]] \times 100\% \text{ for } \xi < 0.009 \quad (2)$$

$$RE_{GK}(\%) = \left[1 - \left[1 - 5.50\xi^{2/3} + 3.77\xi \right] \right] \times 100\% \text{ for } \xi \geq 0.009 \quad (3)$$

where ξ is the dimensionless parameter which is calculated as $\xi = D_g LZ / (QW)$, where D_g is the diffusion coefficient of the gaseous pollutant in the gas phase ($\text{m}^2 \text{ s}^{-1}$) and calculated by the Fuller-Schettler-Giddings equation [35], L is the total length of the PP fabric plate (m); Z is the height of the honeycomb module (m); W is the gap of the module (m); and Q is the airflow rate ($\text{m}^3 \text{ s}^{-1}$). The RE_{CA} is calculated as [18]:

$$RE_{CA}(\%) = \left[1 - \exp\left(-\frac{ZK'_g a_t}{G_{my}}\right) \right] \times 100\% \quad (4)$$

where a_t is the total specific surface area of the module ($\text{m}^2 \text{ m}^{-3}$); K_g is the overall mass transfer coefficient ($\text{mol s}^{-1} \text{ m}^{-2}$), which is expressed as $K'_g = K_g P / RT$, where P is the total pressure (Pa), T is the gas temperature (K), R is the gas constant ($\text{Pa m}^3 \text{ K}^{-1} \text{ mol}^{-1}$), and K_g (m s^{-1}) is calculated as $1/K_g = 1/k_g + m/k_w$ in which k_g and k_w are

the mass transfer coefficient of the gaseous pollutants in the gas phase and the liquid phase, respectively (m s^{-1}) and m is the dimensionless Henry's law volatility constant (H) or the dimensionless effective Henry's law volatility constant (H^*); G_{my} is the molar flux of the air through the HWS and calculated as $G_{my} = V_m / S$ in which V_m is the molar flow rate of the air (mol s^{-1}) and S is the empty cross-sectional area of the honeycomb module (m^2).

The HWS is the wetted wall column whose mass transfer coefficients in the gas phase (k_g) and liquid phase (k_w) are calculated as [36]:

$$k_g = \frac{0.023\pi N_{Re}^{0.83} N_{Sc}^{0.44} D_g}{W} \quad (5)$$

$$k_w = 0.422 \sqrt{\frac{D_w \Gamma}{\rho_w B_F^2}} \quad (6)$$

where N_{Re} is the Reynolds number which is calculated as $N_{Re} = WG_g / \mu_g$, where G_g is the mass flow rate of the air ($\text{kg s}^{-1} \text{ m}^{-2}$), and μ_g is the air viscosity ($\text{kg m}^{-1} \text{ s}^{-1}$); N_{Sc} is the Schmidt number and calculated as $N_{Sc} = \mu_g / \rho_g D_g$ in which ρ_g is the air density (kg m^{-3}); D_w is the diffusion coefficient of the gaseous pollutant in the liquid phase ($\text{m}^2 \text{ s}^{-1}$) which is calculated by the Stokes-Einstein equation [37]; Γ is the liquid mass flow rate based on the wetted perimeter ($\text{kg s}^{-1} \text{ m}^{-1}$); ρ_w is the liquid density (kg m^{-3}); and $B_F = (3\mu_w \Gamma / \rho_w g)^{1/3}$ in which μ_w is the liquid viscosity ($\text{kg m}^{-1} \text{ s}^{-1}$) and g is the gravity acceleration (m s^{-2}).

The theoretical height of the honeycomb module (Z_{CA} , m) can be calculated as [18]:

$$Z_{CA} = \left[\frac{G_{my}}{K_g a_t} \right] \times \left[\int_{C_{out}}^{C_{in}} \frac{dC}{\left(1 - m \frac{V_m}{L_m} \right) C + \left(\frac{V_m}{L_m} C_{in} - C_{w,out} \right)} \right] \quad (7)$$

where L_m is the liquid molar flow rate (mol s^{-1}); $C_{w,out}$ is the outlet concentration of the gaseous pollutant in the liquid phase (ppbv) and it is calculated as $C_{w,out} = C_{w,in} + (V_m / L_m) \times (C_{in} - C_{out})$ in which $C_{w,in}$ is the inlet concentration of the gaseous pollutant in the liquid phase (ppbv). Assuming that the honeycomb modules are replaced by the traditional packing materials, the RE of the wet scrubber packed with different Raschig rings (equivalent diameter (D_e) = 6, 13, and 25 mm) and Berl

Saddle ($D_e = 25$ mm) were studied. The small Raschig rings with D_e equal to 6 mm and 13 mm have the specific surface areas 50% higher than ($710 \text{ m}^2 \text{ m}^{-3}$) and similar ($370 \text{ m}^2 \text{ m}^{-3}$) [36] to that of the honeycomb module. Whereas, the commonly used Raschig ring ($D_e = 25$ mm and $a_t = 190 \text{ m}^2 \text{ m}^{-3}$) [36] and Berl Saddle ($D_e = 25$ mm and $a_t = 250 \text{ m}^2 \text{ m}^{-3}$) [36] have specific surface areas much lower than that of the honeycomb module. The gas-phase and liquid-phase mass transfer coefficients of the WS packed with the Raschig rings and Berl Saddle are typically calculated by Eqs. (S1) and (S2), respectively shown in Section S2 of the SM. The theoretical height of the Raschig rings and Berl Saddle is then calculated by Eq. (7) and compared with that of the honeycomb module. The pressure drop of the traditional WS is determined based on the correlation of $U_t^2 a_t \rho_g \mu_w^{0.2} / (g \epsilon^2 \rho_w)$ and $G_w \sqrt{\rho_g} / (G_g \sqrt{\rho_w})$ and compared to that of the HWS [36].

Results and discussion

Field test results

Figure 2 shows the RE of the HWS versus time with a wide range of the inlet concentrations of 31,870–180,700 ppbv for HF, 12,550–72,580 ppbv for CH_3COOH ; 1384–15,200 ppbv for HCl, 1234–12,390 ppbv for HNO_3 , 201–6017 ppbv for HNO_2 , and 240–1583 ppbv for H_2SO_4 . Among these gases, HF and CH_3COOH are dominant and constitute $64.3 \pm 10.0\%$ and $25.4 \pm 9.3\%$ of the total mass concentration of the mixed gases, respectively while HCl, HNO_3 , HNO_2 , and H_2SO_4 account for only $4.0 \pm 1.7\%$, $3.5 \pm 1.7\%$, $2.1 \pm 1.4\%$, and $0.6 \pm 0.3\%$, respectively. The results show that the REs of all species are very high, which are 96–98% ($\text{RE}_{\text{ave}} = 97 \pm 1\%$) for HF (Fig. 2a), 95–98% ($\text{RE}_{\text{ave}} = 97 \pm 1\%$) for CH_3COOH (Fig. 2a), 96–99% ($\text{RE}_{\text{ave}} = 98 \pm 1\%$) for HCl (Fig. 2b), 95–99% ($\text{RE}_{\text{ave}} = 98 \pm 1\%$) for HNO_3 (Fig. 2b), 95–98% ($\text{RE}_{\text{ave}} = 96 \pm 1\%$) for HNO_2 (Fig. 2c), and 95–98% ($\text{RE}_{\text{ave}} = 97 \pm 1\%$) for H_2SO_4 (Fig. 2c), respectively. That is, the HWS shows a very good performance for mixed gas control with a wide inlet concentration range during the 3.5-yr long period.

The RE of the acidic gases follows the order as $\text{HCl} > \text{HNO}_3 > \text{HF} > \text{HNO}_2 > \text{CH}_3\text{COOH} > \text{H}_2\text{SO}_4$. Among these gases, HCl has the highest RE during 3.5 yr although HCl has the lowest Henry's law solubility constant ($H_{\text{HCl}} = 1.1 < H_{\text{HNO}_2} = 49 < H_{\text{CH}_3\text{COOH}} = 8.8 \times 10^3 < H_{\text{HF}} = 1.3 \times 10^4 < H_{\text{HNO}_3} = 2.1 \times 10^5 < H_{\text{H}_2\text{SO}_4} = 2.9 \times 10^9 \text{ M atm}^{-1}$) [38, 39]. On the other hand, the dissociation constants of HCl ($K_{\text{HCl}} = 1.7 \times 10^6 \text{ M}$) is several orders of magnitude higher than other acidic gases ($K_{\text{H}_2\text{SO}_4} = 10^3$, $K_{\text{HNO}_3} = 15.4$; $K_{\text{HF}} = 6.3 \times 10^{-4}$; $K_{\text{HNO}_2} = 5.1 \times 10^{-4}$; $K_{\text{CH}_3\text{COOH}} = 1.7 \times 10^{-5} \text{ M}$) [11, 38]. It

indicates that Henry's law constant just reflects the physical solubility of the gases while additional dissociation and chemical reaction constants also play an important role in the absorption process by WSs [16, 38], which is explained in detail later.

It is seen that the high RE of the HWS does not change from the first operating day (11/08/2016) until the latest testing day (19/03/2020) for CH_3COOH (1st: 97%; 13th: 97%) and HCl (1st: 98%; 13th: 98%), respectively, while it slightly decreases for 1% for HF (1st: 98%; 13th: 97%), 3% for HNO_3 (1st: 98%; 13th: 95%), 2.0% for HNO_2 (1st: 98%; 13th: 96%), and 1% for H_2SO_4 (1st: 97%; 13th: 96%), respectively. It may be due to the slightly increased pressure drop which is to be discussed later. It is noted that the HWS was cleaned manually only once every year by spraying water jet from the top of the modules during the test period (three cleaning times in total, in July 2017, April 2018, and July 2019, respectively), which helps maintain $\geq 95\%$ high RE during this long-term test. In comparison, the packing materials of the traditional packed tower need to be washed once every quarter to reduce particle clogging. After each yearly maintenance denoted as "M" in Fig. 2, the RE of HF increases slightly (1st M: +1.4%; 2nd M: +1.5%) while the REs of other acidic gases still maintain to be very high.

Unlike other traditional WSs, the RE of the HWS is not affected by the inlet concentration, which is always higher than 95% for all acidic gases when C_{in} varies from sub-ppmv (709 ± 324 ppbv in average) to super-ppmv ($105,500 \pm 41,040$ ppbv in average) as shown in Fig. 3. However, it is seen that the REs of the HWS increase slightly by 4, 2, and 2% when the inlet concentration increases by 8.2, 5.6, and 3.9 times for HNO_3 , HCl, and H_2SO_4 , respectively as shown in Fig. 3a. The slightly positive correlation of the RE and the inlet concentration of HNO_3 and HCl was found with R^2 of 0.37 (p -value = $0.03 < 0.05$) and R^2 of 0.53 (p -value = $0.02 < 0.05$), respectively. It is due to the variation of the inlet concentrations of HNO_3 and HCl from low (< 3000 ppbv) to high concentrations (> 3000 ppbv). In Fig. 3a, it is found that the RE of H_2SO_4 is lower than HNO_3 and HCl since the inlet concentration of H_2SO_4 is lower than 1600 ppbv and 9.2 times lower than HNO_3 and 11.2 times lower than HCl in average. In comparison, the inlet concentrations of HF and CH_3COOH are always higher than 12,500 ppbv. Therefore, the RE also remains high and constant without an increasing trend, as shown in Fig. 3b. The RE of HNO_2 also shows no dependence on the inlet concentration which varies from low (< 3000 ppbv) to high concentrations (> 3000 ppbv) but does not range widely ($C_{\text{in-ave}} = 2899 \pm 1596$ ppbv) as compared to HCl ($C_{\text{in-ave}} = 8493 \pm 3566$ ppbv) and HNO_3 ($C_{\text{in-ave}} = 6969 \pm 3353$ ppbv) with a wide concentration range (Fig. 3b).

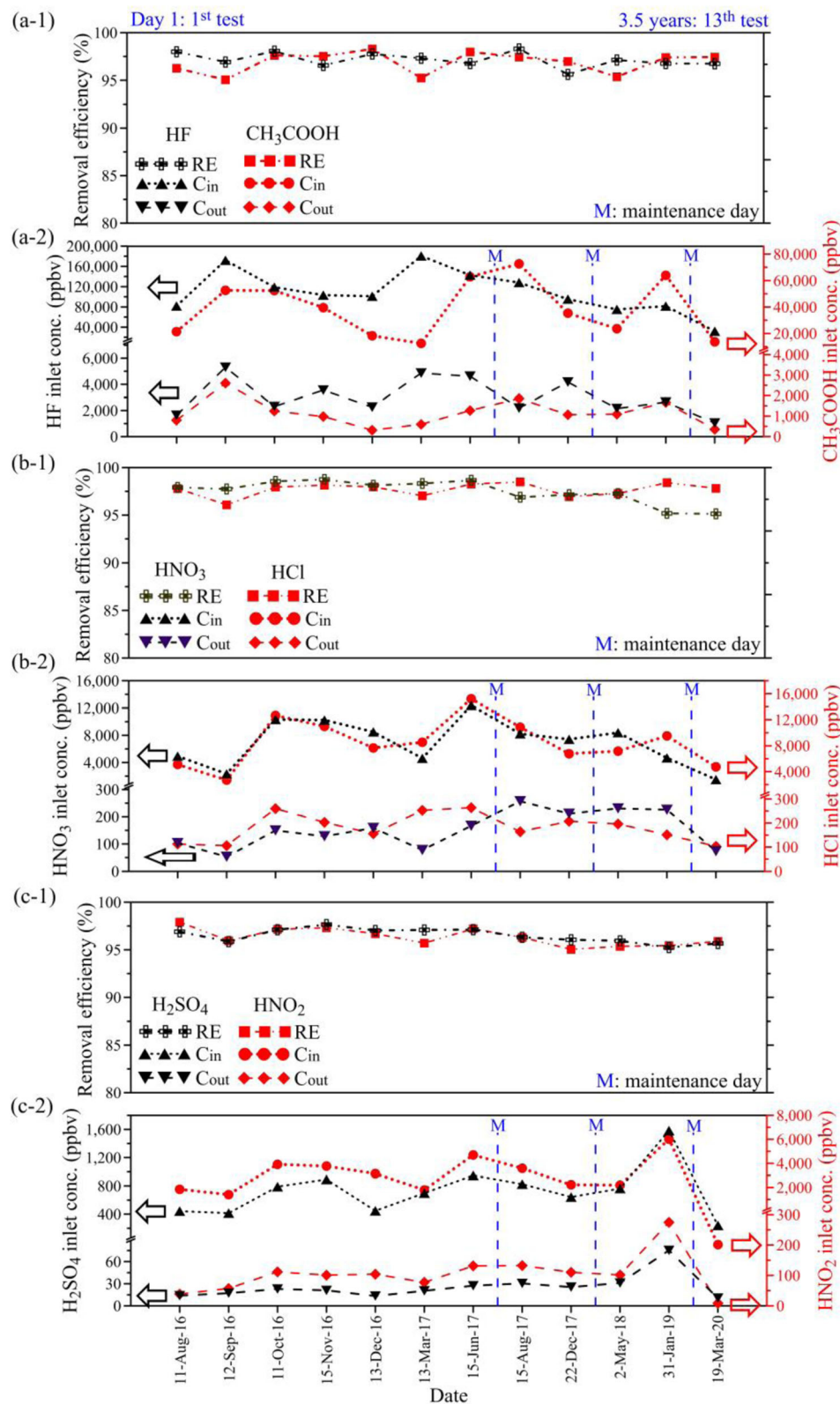
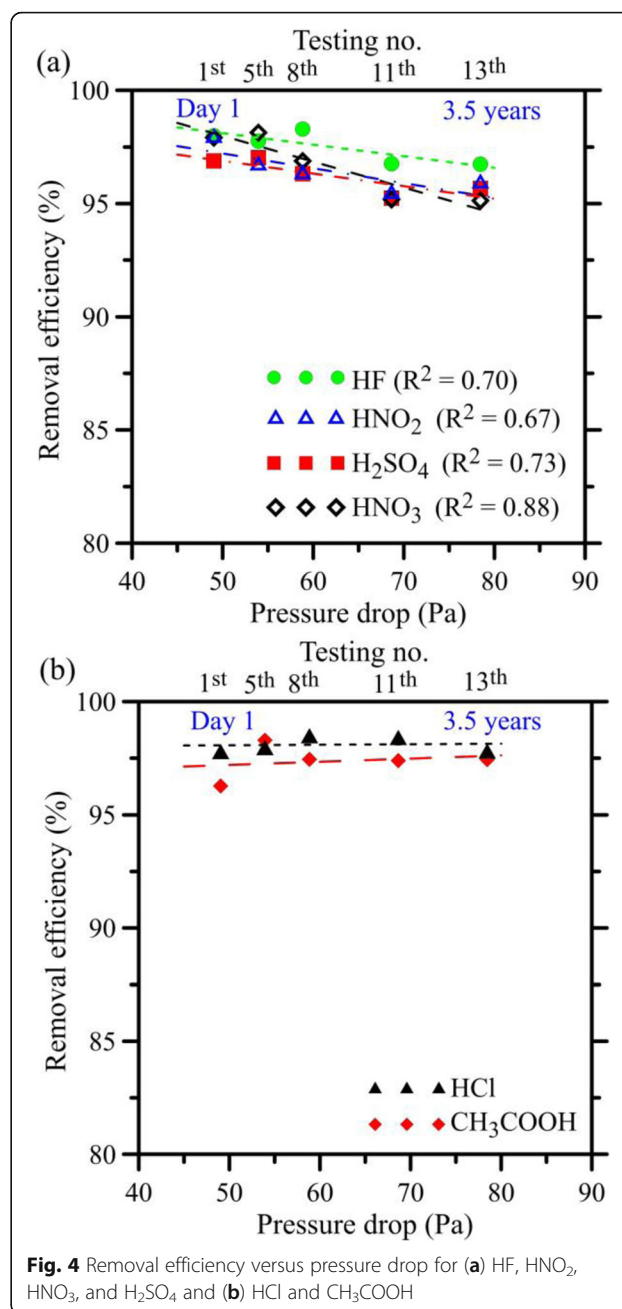
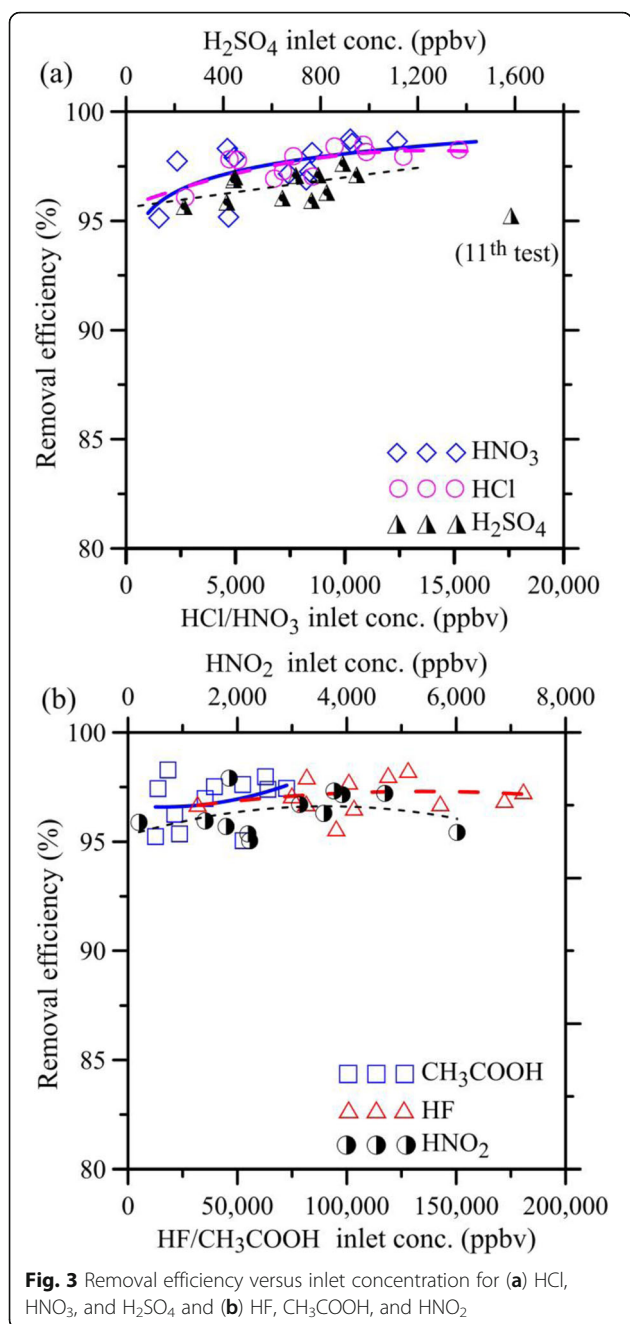


Fig. 2 Removal efficiency and inlet and outlet concentrations versus test date for (a) HF and CH₃COOH, (b) HCl and HNO₃, and (c) HNO₂ and H₂SO₄



During a 3.5-yr period, the pressure drop is found to be very small and increases only slightly from 49.0 to 78.4 Pa (0.5 to 0.8 cm H₂O), which is also close to the theoretical value of 59.8 Pa (0.58 cm H₂O) as shown in Fig. S3 in the SM. The slight increase in the pressure drop during 3.5 yr implies that only slight fouling of the honeycomb material might have occurred which could not be cleaned thoroughly by water jet. In Fig. 4a, it is found that the RE decreases by 3% for HNO₃, 2% for HNO₂ and HF, and 1% for H₂SO₄ as the pressure drop increases by 29.4 Pa (0.3 cm H₂O). The RE shows a good

negative correlation with the pressure drop for HNO₃ ($R^2 = 0.88$; p -value = $0.02 < 0.05$) with 95% confident interval and H₂SO₄ ($R^2 = 0.73$; p -value = $0.06 < 0.1$), HF ($R^2 = 0.70$; p -value = $0.08 < 0.1$), and HNO₂ ($R^2 = 0.67$; p -value = $0.09 < 0.1$) with 90% confident interval. It indicates a slight influence of the pressure drop on the RE for these gases. On the other hand, the RE of HCl and CH₃COOH does not show the dependence on the pressure drop as shown in Fig. 4b since HCl has high dissociation constant and CH₃COOH has a very high inlet concentration.

The lab-scale HWS operated at a flow rate of 1.0 CMM and an L/G ratio of 18 L m^{-3} in the previous study [14] while the present scale-up, actual HWS device operated at a higher flow rate of 100 CMM, a higher L/G ratio of 26 L m^{-3} , and the same retention time of 0.5 s. Although the REs of the present HWS for HF, HCl, and CH_3COOH are not as high as $\sim 99\%$ for HF and HCl and $\sim 100\%$ for CH_3COOH achieved in the previous study, the long-term test results show that the present HWS always reaches the RE higher than 95% stipulated in the emission standard [1]. The inlet concentrations in the previous study ($C_{\text{in}} = 100\text{--}450$ ppbv for HF, $3000\text{--}11,000$ ppbv for HCl, and $600\text{--}6500$ ppbv for CH_3COOH) were lower than the present HWS. Since there are 64 modules placed in parallel inside the HWS with some unused cross-section area (or dead space), the required L/G ratio in the present study needs to be 1.4 times higher, which causes the pressure drop to increase from 23.5 to 49.0 Pa but it is still much lower than that of most traditional WSs.

The emission rates of each species in 12 sampling times were calculated from the outlet concentrations and airflow rate and is found to be less than 0.03, 0.04, 2.4×10^{-3} , 4.0×10^{-3} , 3.2×10^{-3} , and $1.8 \times 10^{-3} \text{ kg h}^{-1}$ for HF, CH_3COOH , HCl, HNO_3 , HNO_2 , and H_2SO_4 , respectively. It indicates that the exhaust gaseous acidic pollutants from the manufacturing process of the fab are well controlled with only 2.5, 3.0, 0.3, 0.4, 0.2, and 0.6%, respectively, of the total regulated mass emission rates (0.1 kg h^{-1} for H_2SO_4 and 0.6 kg h^{-1} for other gases) for a semiconductor manufacturing fab. In addition, the bad odor due to high HF and CH_3COOH discharge concentrations disappears after the installation of the present HWS.

Results of theoretical prediction

Fig. 5 shows the theoretical REs of the HWS with $D \times Z = 0.3 \times 0.3 \text{ m}$ for different acidic gases. It is seen that the RE_{GK} is about 100% for all the acidic gases indicating that the pollutants diffuse completely from the gas phase into the liquid phase fast enough without limiting the ensuring dissociation and reaction processes of the pollutants in the water film. Whereas, the RE_{CA} based on Henry's law constant ($\text{RE}_{\text{CA-H}}$) is close to 100% for HF, 98% for CH_3COOH , 13% for HCl, 99% for HNO_3 , 92% for HNO_2 , and 98% for H_2SO_4 , respectively indicating that the mass transfer resistance across the gas-liquid interface is high for HCl, moderate for HNO_2 , and very low for other acidic gases. It is because that the solubility of the HCl and HNO_2 in the liquid is several orders of magnitude lower than other acidic gases. The RE_{CA} based on the effective Henry's law constant ($\text{RE}_{\text{CA-H}^*}$) of HCl and HNO_2 is as high as 99%, when the effective Henry's law constant is used for prediction. This implies that the dissociation and reaction are the rate-limiting steps in the HWS and the chemical reaction enhances the absorption rate and increases the dissolved capacity of the liquid film in the HWS. The $\text{RE}_{\text{CA-H}^*}$

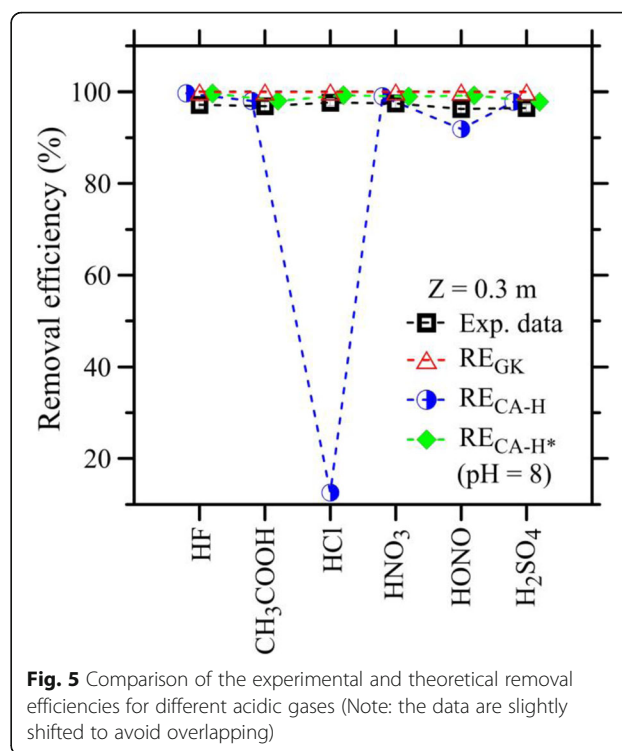
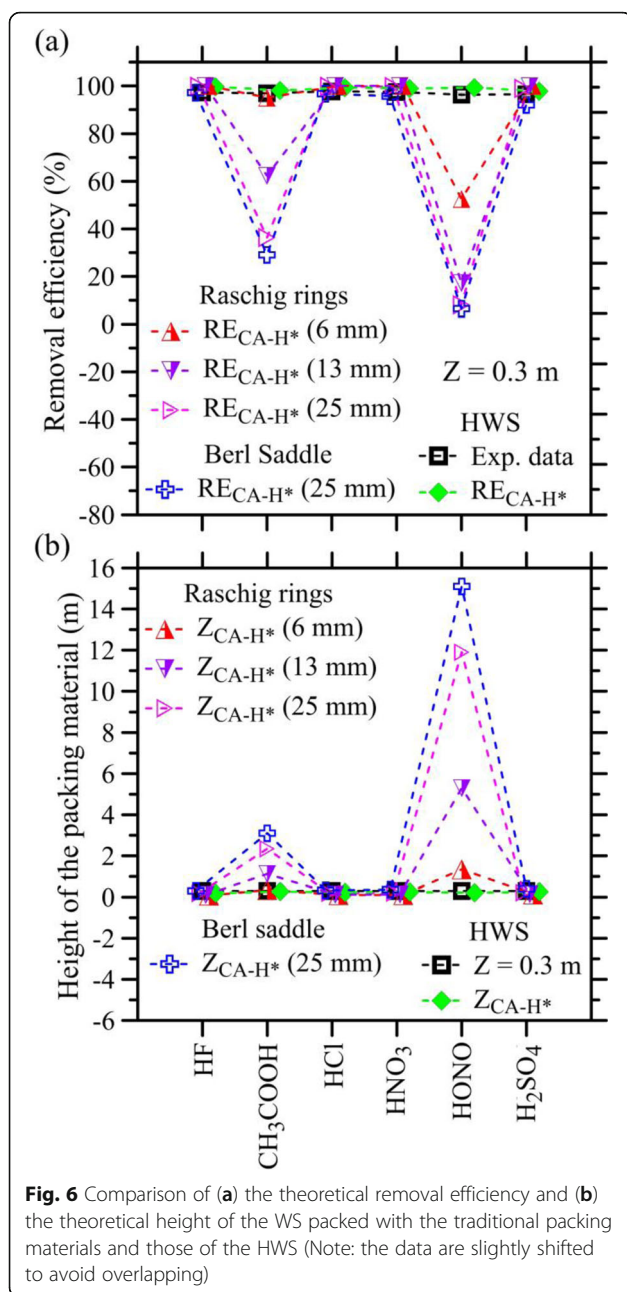


Fig. 5 Comparison of the experimental and theoretical removal efficiencies for different acidic gases (Note: the data are slightly shifted to avoid overlapping)

results are close to the experimental data with less than +3% biases.

To achieve the RE as high as the experimental data, the theoretical heights based on the effective Henry's law constant ($Z_{\text{CA-H}^*}$) are 0.19 m for HF, 0.27 m for CH_3COOH , 0.23 m for HCl, 0.25 m for HNO_3 , 0.21 m for HNO_2 , and 0.16 m for H_2SO_4 , respectively, as shown in Fig. S4 in the SM. It indicates that the theoretical values are very similar to the designed value of 0.3 m for mixed acidic gas control. The theoretical heights based on Henry's law constant ($Z_{\text{CA-H}}$) are also shown in Fig. S4 in the SM, which demonstrates that it is required a longer module to achieve the RE of higher than 95% for HCl ($Z_{\text{CA-H}} = 8.5 \text{ m}$) and HNO_2 ($Z_{\text{CA-H}} = 0.4 \text{ m}$) when the scrubbing NaOH solution is not added. In summary, the equations based on the two-film theory can be used to predict the theoretical RE of the HWS and the theoretical height of the honeycomb module with good accuracy.

Figure 6 shows the $\text{RE}_{\text{CA-H}^*}$ and $Z_{\text{CA-H}^*}$ of the WS packed with the traditional packing materials including Raschig rings ($D_e = 6, 13, \text{ and } 25 \text{ mm}$) and Berl Saddle ($D_e = 25 \text{ mm}$). As compared to the honeycomb module with a similar specific surface area, the theoretical REs of the WS packed with 13-mm Raschig rings are high for HF, HCl, and H_2SO_4 ($\text{RE}_{\text{CA-H}^*} \geq 99\%$) but low for CH_3COOH ($\text{RE}_{\text{CA-H}^*} = 62\%$) and HNO_2 ($\text{RE}_{\text{CA-H}^*} = 17\%$). It is because that CH_3COOH has the lowest dissociation constant among these acidic gases and both Henry's law and dissociation constants of HNO_2 are several orders of magnitude lower than other acidic gases. When the 6-



mm Raschig ring with a higher specific surface area than the honeycomb module is used, the REs can be achieved of 95% for CH₃COOH and 53% for HNO₂, respectively. However, the REs of the traditional WS packed with 6-mm Raschig rings are still lower than those of the HWS for CH₃COOH ($Z_{CA-H^*} = 98\%$) and HNO₂ ($Z_{CA-H^*} = 99\%$). It implies that HWS enhances the mass transfer rate of CH₃COOH and HNO₂ which have low solubility and dissociation ability due to multiple small gaps with uniform water-absorbing surface. Additionally, the theoretical pressure drop of the WS packed with 6- or 13-mm Raschig rings is about 490 Pa (5.0 cm H₂O) or 245 Pa (2.5 cm H₂O),

respectively, which is much higher than that of the HWS (0.58 cm H₂O). The theoretical height of the WS packed with 6- or 13-mm Raschig rings needs to be as large as 1.34 or 5.3 m, respectively to achieve the RE higher than 95% for all the acidic gases for mixed gas control as shown in Fig. 6b. That is, the HWS is an economical control device for mixed gas control as compared with the traditional WS.

To reduce the pressure drop, the traditional WS is usually packed with bigger packing materials with lower specific surface areas such as 25-mm Raschig rings or 25-mm Berl Saddles. However, the RE of the traditional WS packed with 25-mm Berl Saddle is not only as low as 29% for CH₃COOH and 6% for HNO₂ but just 92% for H₂SO₄, which does not meet the RE requirement. It is because that the inlet concentrations of H₂SO₄ are very low ($C_{in} = 0.24\text{--}1.58$ ppmv) as compared to those of the acidic gases. Although the RE of the WS packed with 25-mm Raschig rings is also as high as 95% for H₂SO₄, it is still lower than that of the HWS and the WS packed with 6- or 13-mm Raschig rings, which is due to its lower specific surface area. This indicates that the HWS outperforms the traditional WS with similar or even larger specific surface areas for the RE of the mixed gases and pressure drop.

Conclusions

This work evaluated the long-term performance of the honeycomb wet scrubber for removing gaseous acidic gases and odor at a semiconductor manufacturing fab in 3.5 yr from August 2016 to March 2020. The results of 12 times of RE tests indicated that the HWS is capable of removing the gaseous acidic gases and odor with the inlet concentrations varying from low concentrations of 240–1583 ppbv for H₂SO₄ and 201–6017 ppbv for HNO₂ to moderate concentrations of 1234–12,390 ppbv for HNO₃ and 1384–15,200 ppbv for HCl, and high concentrations of 12,550–72,580 ppbv for CH₃COOH and 31,874–180,700 ppbv for HF. All RE data, which are 96–98% for HF, 95–98% for CH₃COOH, 96–99% for HCl, 95–99% for HNO₃, 95–98% for HNO₂, and 95–98% for H₂SO₄, respectively, met the regulation for the control devices in Taiwan. The pressure drop of the HWS maintained as low as 49.0–78.4 Pa (0.5–0.8 cm H₂O) and increased slightly during the 3.5-yr period only, which is much lower than that of the traditional WSs. The theoretical RE and the theoretical height of the HWS module were close to the experimental data indicating that the theoretical equations can facilitate the design of the HWS. The HWS showed higher theoretical RE and lower theoretical height for the mixed gas control, especially for the acidic gases with low solubility and dissociation ability as compared with the WS packed with the traditional packing materials. That is, the honeycomb wet scrubber can be used as an efficient device for mixed acidic gases and odor control with high RE and low pressure drop for the long-term.

Nomenclature

a_t	Specific surface area ($\text{m}^2 \text{m}^{-3}$)	N_{RE}	Reynolds number
C_{in}	Inlet concentration (ppbv)	N_{Sc}	Schmidt number
C_{out}	Outlet concentration (ppbv)	Q	Airflow rate ($\text{m}^3 \text{s}^{-1}$)
$C_{w,in}$	Inlet concentration of the gaseous pollutant in the liquid phase (ppbv)	R	Gas constant ($\text{m}^3 \text{Pa K}^{-1} \text{mol}^{-1}$)
$C_{w,out}$	Out concentration of the gaseous pollutant in the liquid phase (ppbv)	P	Total pressure (Pa)
D	The inner diameter of the packing material (m)	RE	Experimental removal efficiency (%)
D_e	The equivalent diameter of the raschig ring (m)	RE_{CA}	Theoretical removal efficiency based on two film theory (%)
D_g	Gas-phase diffusion coefficient ($\text{m}^2 \text{s}^{-1}$)	RE_{CK}	Theoretical removal efficiency based on diffusion theory (%)
D_w	Liquid-phase diffusion coefficient ($\text{m}^2 \text{s}^{-1}$)	S	Empty cross-section surface area (m^2)
g	Gravity acceleration (m s^{-2})	T	Temperature (K)
G_g	The mass flow rate of the air ($\text{kg s}^{-1} \text{m}^{-2}$)	U_t	Superficial air velocity (m s^{-1})
G_w	The mass flow rate of the liquid ($\text{kg s}^{-1} \text{m}^{-2}$)	V_m	The molar flow rate of the air (mol s^{-1})
G_{my}	The molar flux of the air ($\text{mol s}^{-1} \text{m}^{-2}$)	W	Gap distance of the honeycomb module (m)
H	Henry's law volatility constant (atm M^{-1})	Z	Height of the honeycomb module (m)
H^*	Effective Henry's law volatility constant (atm M^{-1})	ϵ	Fractional voids in dry packing
K	Dissociation constant (M)	ρ_g	Air density (kg m^{-3})
K'_g	Overall mass transfer coefficient ($\text{mol s}^{-1} \text{m}^{-2}$)	ρ_w	Liquid density (kg m^{-3})
K_g	Overall mass transfer coefficient (m s^{-1})	μ_g	Air viscosity (kg s m^{-1})
k_g	Gas-phase mass transfer coefficient (m s^{-1})	μ_w	Liquid viscosity (kg s m^{-1})
kw	Liquid-phase mass transfer coefficient (m s^{-1})	ξ	Dimensionless parameter
L	The total length of the PP fabric plate (m)	Γ	Liquid mass flow rate based on the wetted perimeter ($\text{kg s}^{-1} \text{m}^{-1}$)
L_m	Liquid molar flow rate (mol s^{-1})		

Supplementary Information

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Additional file 1.

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Authors' contributions

YLL and BTW designed the study and carried out the field tests. GYL and ZL contributed to data analysis. GHH drafted and edited the manuscript. LTC contributed to data analysis, modeling, and manuscript preparation. DYHP and CJT critically reviewed and edited the final manuscript. All the authors read and approved the final manuscript.

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Availability of data and materials

All data generated or analyzed during this study are included in this article and its supplementary materials file. The raw data are available from the corresponding author upon reasonable request.

Declarations

Competing interests

The authors declare they have no competing interests.

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