Honeycomb Wet Scrubber For Acidic Gas Control: Modeling And Long-term Test Results

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Research

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

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1

18   **Abstract**

19   A laboratory scale, 1.0 CMM wet scrubber packed with water-absorbing honeycomb material

20   (HWS) with a very large geometric surface area of 480 m^2 m^{-3} and a low pressure drop developed in

21   our previous study was shown to achieve a very high removal efficiency for gaseous acidic gases but

22   there were no long-term test data. In this study, the HWS scaled up to operate at 100 CMM flow rate

23   was tested for removing mixed acidic gases at a semiconductor fab for a very long period of 3.5 years.

24   Results showed that the removal efficiency for the mixed gases emitted from the fab always maintained

25   as high as > 95% for HF, CH_3COOH, HCl, HNO_3, HNO_2, and H_2SO_4 with the inlet concentrations

26   ranging from supper-ppmv to sub-ppmv, respectively, during 3.5-yr period. With water jet cleaning of

27   the honeycomb modules once per year, the pressure drop of the HWS remained to be very low at 0.5-

28   0.8 cmH_2O, indicating minimal scaling in the HWS existed. Additionally, the predicted height and

29   removal efficiencies of the HWS were very close to the experimental data. The excellent long-term

30   performance of the HWS warrants its potential applications in many areas in which liquid absorption

31   is the preferred treatment method and the theoretical equations can facilitate the design of the HWS.

32

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

34

1. Introduction

Inorganic acids such as HF, HCl, HNO_3, H_2SO_4, and CH_3COOH are widely used in semiconductor or photoelectric 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 TWEPA (Taiwan Environment Protection Agency). The standard stipulates that the total mass emission rate from all stacks must be $<$ 0.6 kg h^{-1} for HF, HCl, and HNO_3, and $<$ 0.1 kg h^{-1} for H_2SO_4, respectively, for a semiconductor fab, and the removal efficiency 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 $PM_{2.5}$ [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 the acidic gases [9-11] due to their small footprint, simple structure, and low capital cost [12]. However, the measured removal efficiency (RE) of most of the traditional WSs can't meet the required value of greater than 95% [1]. For instance, the removal efficiency of a vertical packed WS with 1,500 CMM flow rate were 70.6-93.4% for HF, HCl, HNO_2, HNO_3, and CH_3COOH at a high-tech factory while the removal efficiency of a lab-scale packed WS with 0.6-1.8 CMM flow rate was found to be as low as 76.8%-85.7% for H_2SO_4 mist [13]. When the removal efficiency of the WSs can't meet the standard value, they must follow the design criteria

52 and operating conditions in which the specific surface area of packing materials must be $\geq 90 \ m^2 \ m^{-3}$,

53 the residence time through the scrubbing section must be ≥ 0.5 s, a wetting factor must be $\geq 0.1 \ m^3 \ h^{-1}$

54 ^{1}, and the pH of scrubbing liquid must be ≥ 7 as shown in Table 1 [1].

55   Among all operating parameters, the inlet concentration was found to be the most important

56 parameter affecting the removal efficiency of the packed WS. The removal efficiency of a conventional

57 packed WS increased with the increasing inlet concentration at low inlet concentration (C_{in}) and

58 approached a constant value until C_{in} reached a certain value for HF, HCl, HNO_3, HNO_2, and

59 CH_3COOH [14]. For example, the removal efficiency of a multistage dual-flow sieve plate WS was

60 93.8% as C_{in} was as high as 100-300 ppmv for HCl [15] while a packed tower was found to have a

61 removal efficiency of less than 90% when C_{in} was lower than 1.0 ppmv for HF and HCl [9, 14]. The

62 WSs normally have high pressure drop resulting in high power consumption and operation cost [13,

63 16]. For example, a WS installed mesh MV filters operated at 90 CMM flow rate achieved > 95%

64 removal efficiency for NH_3 (L/G of 2.0 L m^{-3}) at a high pressure drop of > 1,411 Pa [17]. However,

65 there were no long-term test data provided. Therefore, it is a challenge to design a WS with a high

66 removal efficiency and low pressure drop which lasts for a long time in an actual plant.

67   The WSs are normally packed with random packing materials to enhance the removal efficiency.

68 The packing materials, which can be in the form of a saddle, ring or tellerette and made of ceramics,

69   glass, metal, porcelain, steel or plastics [18, 19], should have a high specific surface area to maximize

70   the gas-liquid contact surface for mass transfer [20] and liquid holdup. The removal efficiencies of

71   acidic gases were found to increase when the packing material size decreased [21]. For instance, the

72   removal efficiency for HCl was increased by 2.0-3.5% when the specific surface area of packing balls

73   was increased from 150 to 224 m^2 m^{-3} [22]. The packing materials also need to resist fouling and

74   corrosion, avoid particle clogging, and enhance water retention to minimize the pressure drop and

75   maintenance cost for long-term use. In addition, they need to provide good air distribution for uniform

76   absorption [13] and low pressure drop in the WSs. The pressure drop of a WS with conventional plastic

77   packing materials is about 413-830 Pa (4.2-8.5 cmH_2O) per meter of the packing column [23-25].

78   The removal efficiency can also be improved by increasing the L/G ratio due to the increasing

79   gas-liquid contacting surface area. The removal efficiency could increase from 82.8% to 97.5% as the

80   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

81   pressure drop [11, 23]. Adding surfactants in the scrubbing liquid can help enhance the removal

82   efficiency of the WSs due to the charge generation on the water surface by surfactants but the

83   surfactants are costly [10]. The overall efficiency of a packed WS for NH_3, HF, and HCl (C_{in} = 0.2-3

84   ppmv) was reported to increase from 56% to 83% at 1.1 s residence time and 10 CMM flow rate when

85   surfactants were added [9, 10]. The pH of the scrubbing solution is another factor that affects the

86   removal efficiency of the WSs for some weak acidic gases such as CH_3COOH but not for strong acidic

87   gases such as HCl [11]. There is no significant effect on the removal efficiency for CH_3COOH when

88   the pH is higher than 7.5 due to the gas dissolution limit while the removal efficiency for HCl and

89   HNO_3 is nearly constant for the pH ranging from 7-9 [11, 27]. To achieve good efficiency for mixed

90   acidic gases and save the operating cost, the pH of scrubbing liquid should be maintained > 7.5.

91   Most of the previous researches have focused on enhancing removal efficiency of WSs by

92   determining the optimal operating conditions [16, 26, 28], attempting to improve the performance of

93   the existing WSs operating in high-tech industries [9, 10, 22], and designing lab-scale WSs for short-

94   term study only [13, 19, 29]. A WS packed with a multi-parallel-plate (MPP) module (PPWS)

95   developed by our group can achieve very high removal efficiencies for HCl (> 99%), HNO_3 (> 98%),

96   and CH_3COOH (> 99%) and very low pressure drop (23.5 Pa) at a regulated residence time of 0.5 s, a

97   low inlet concentration (< 3 ppmv), and a high L/G ratio (18.5 L m^{-3}) [11]. It is because that the PPWS

98   uses polypropylene plates coated with nano-TiO_2 packing material with 326.67 m^2 m^{-3} specific surface

99   area to enhance the hydrophilicity for scrubbing liquid to form a uniform liquid film. However, this

100 packing material requires a time-consuming and meticulous procedure for preparation and assembly

101 with a precise gap of 3 mm. The pilot-scale 1.0 CMM WS with the honeycomb module (HWS) was

102 further developed with the removal efficiency as high as the PPWS and the packing module is easy to

103 fabricate [14]. The HWS showed nearly 100% for HF (C_{in} = 0.1-0.4 ppmv) and 99% for HCl (C_{in} =

104 3.4-10.8 ppmv) and CH_3COOH (C_{in} = 0.7-6.5 ppmv) at an optoelectronic factory. Similar to the MPP

105 module, the honeycomb packing material made of water-absorbing polypropylene (PP) fabric (0.875

106 kg m^{-1}) shows the higher specific surface area (483.2 m^2 m^{-3}) and lower pressure drop than most of the

107 packing materials as shown in Table 2.

108   Thus, the honeycomb packing material with the uniform arrangement and 3-mm gap becomes a

109 promising packing material for an efficient WS. But the removal efficiency of these small scale HWS

110 and PPWS were obtained when these WSs were run for several days. There are no scale-up, actual

111 HWS which runs in a fab for the long-term to control the soluble gas emission. In this study, the HWS

112 was scaled up as an actual device in a semiconductor manufacturing fab at 100 CMM flow rate to

113 remove acidic gases and odor. The removal efficiencies of the HWS for the mixed gases containing

114 HF, HCl, HNO_3, HNO_2, H_2SO_4, and CH_3COOH were tested for 13 times over 3.5 years for the removal

115 efficiency while the pressure drop was also measured to make sure that there was no fouling occurred

116 in the system. The theoretical analysis was conducted to predict the height of the packing module and

117 the removal efficiency of the HWS and the traditional WS to further justify the outstanding

118 performance of the HWS.

119 **2. Materials and methods**

2.1. Field test

121   The experimental setup of the HWS installed at a semiconductor manufacturing fab which

122   produces DRAM in Hsinchu, Taiwan to reduce its emission and bad odor of acidic gases is shown in

123   Fig. 1a. The HWS, which is registered as a patent in Taiwan (No. I569867 and M466724), is an add-

124   on central scrubber device mainly to remove high concentration acidic gas pollutants collected from

125   about 40 local scrubbers before a traditional central packed tower which has low removal efficiency

126   with high emitted HF and CH_3COOH concentrations (e.g., typically > 10 ppmv without the present

127   HWS) and bad odor. The HWS consists of 64 honeycomb modules placed in parallel in four rows (16

128   modules per row) as shown in Fig. 1b, spray nozzles installed at the top of honeycomb modules, and

129   a liquid reservoir with conductivity and pH meters. Each honeycomb elements were scrolled in a

130   cylindrical casing (D x Z = 300 x 300 mm). The operating parameters of the HWS are shown in Table

131   1, which meet the design criteria for a packed tower to achieve the efficiency-based emission standards

132   for acidic gases. In this study, the HWS with the specific surface area of the packing material of 480

133    $m^2 m^{-3}$ was designed to operate at 100 CMM flow rate with 0.4 m s^{-1} face velocity and 0.5 s retention

134   time. The liquid flow rate was about 2,600 L min^{-1} which was controlled by a flow meter and the L/G

135   ratio was maintained at 26 L m^{-3}. NaOH solution was used as the scrubbing solution with the pH

136   maintained at 7.5-8. The theoretical pressure drop of 59.8 Pa (0.58 cmH$_2$O) was calculated by the

137 method in Chien et al. [11], which is higher than the pressure drop (23.5 Pa) of the lab-scale, 1.0 CMM

138 HWS in the previous study due to the increasing L/G ratio.

139 The HWS was tested for the removal efficiency of inorganic gaseous acidic pollutants including

140 HF, HCl, HNO_2, HNO_3, H_2SO_4, and CH_3COOH from August 2016 to March 2020 (3.5 years) with 13

141 times (11/08/2016, 12/09, 11/10, 15/11, 13/12, 13/03/2017, 15/06, 15/08, 22/12, 02/05/2018,

142 31/01/2019, 6/12, and 19/03/2020). The testing frequency was set at once per month in the first 5

143 months (5 tests) and then extended to once per three months in the next 12 months (4 tests), once per

144 six months (2 tests), and finally once per year (1 test). The latest testing time (19/03/2020) was about

145 3 months after the 12th test when the pH meter was not functioning well when the NaOH solution was

146 not pumped into the tank to maintain the pH $\geq$ 7.5. The result of the 12th test is shown in Fig. S1 in

147 Section S1 in the supplementary information (SI). Two porous denuder samplers (PDS) with a 2 L min[-]

148 [1] flow rate [4, 30-32] were used to sample mixture gases at the inlet and outlet of the HWS

149 simultaneously in 20-60 min depending on inlet concentrations. The PDS consists of a Teflon filter for

150 sampling particles followed by two porous-metal discs coated with solutions containing 10 ml, 5%

151 (w/v) sodium carbonate and 1% (w/v) glycerol in 1:1 (v/v) methanol/water solution for sampling

152 inorganic acidic and basic gases. An ion chromatography (model 883, Metrohm AG, Switzerland) was

153 used to quantify the concentrations of gases and particles. The particulate phase concentration was

154 found to be much smaller than the corresponding gaseous species and is not a target of this study. The

155 RE of the acidic gases was then calculated as:

156

157 $$RE(\%) = \left[1 - \frac{C_{out}}{C_{in}}\right] \times 100\% \qquad (1)$$

158

159 where C_{out} (ppbv) was the outlet concentrations of HF, HCl, HNO_2, HNO_3, H_2SO_4, or CH_3COOH in

160 the gas phase. Four tons of makeup water per day was required to compensate for water evaporation

161 and maintain the conductivity of 0.3-0.5 mS cm^{-1} in the water reservoir. The pressure drop of the HWS

162 was also measured at five testing times (11/08/2016, 13/12, 15/08/2017, 31/01/2019, and 19/03/2020)

163 during the 3.5-yr period.

164 *2.2. Theoretical prediction*

165   The theoretical removal efficiency of the HWS is calculated by using Eqs. (2) and (3) and Eq. (4)

166 based on the convection-diffusion theory of Gormley and Kennedy (RE_{GK}, %) [33, 34] and two film-

167 theory (RE_{CA}, %) [18], respectively, as follows:

168

169 $$RE_{GK}(\%) = \left[1 - [0.82\exp(-11.5\xi) + 0.097\exp(-70.1\xi)]\right] \times 100\% \quad \text{for } \xi < 0.009 \qquad (2)$$

170

171 $$\text{RE}_{GK}(\%) = \left[1 - \left[1 - 5.50\xi^{2/3} + 3.77\xi\right]\right] \times 100\% \quad \text{for } \xi \geq 0.009 \tag{3}$$

172

173 where ξ is the dimensionless parameter which is calculated as $\xi = D_gLZ/QW$, where D_g is the diffusion

174 coefficient of the gaseous pollutant in the gas phase ($m^2\ s^{-1}$) and calculated by the Fuller-Schettler-

175 Giddings equation [35], L is the total length of the PP fabric plate (m); Z is the height of the honeycomb

176 module (m); W is the gap of the module (m); Q is the airflow rate ($m^3\ s^{-1}$). The RE_{CA} is calculated as

177 [18]:

178

179 $$\text{RE}_{CA}(\%) = \left[1 - \exp\left(-\frac{ZK_ga_t}{G_{my}}\right)\right] \times 100\% \tag{4}$$

180

181 where a_t is the total specific surface area of the module ($m^2\ m^{-3}$); K_g is the overall mass transfer

182 coefficient (mole $s^{-1}\ m^{-2}$) which is calculated as $1/K_g = 1/k_g + m/k_w$ in which k_g and k_w are the mass

183 transfer coefficient of the gaseous pollutants in the gas phase and the liquid phase, respectively (mole

184 $s^{-1}\ m^{-2}$) and m is the dimensionless Henry's law constant (H) or the dimensionless effective Henry's

185 law constant (H^*); G_{my} is the molar flux of the air through the HWS and calculated as $G_{my} = V_m/S$ in

186 which V_m is the molar flow rate of the air (mole s^{-1}) and S is the empty cross-sectional area of the

187 honeycomb module (m^2).

188   The HWS is the wetted wall column whose the mass transfer coefficients in the gas phase (k_g)

189   and liquid phase (k_w) are calculated as [36]:

190

191    $$k_g = \frac{0.023\pi N_{Re}^{0.83} N_{Sc}^{0.44} D_g P}{W} \tag{6}$$

192    $$k_w = 0.422\sqrt{\frac{D_w \Gamma}{\rho_w B_F^2}} \tag{7}$$

193

194   where P is the total pressure (atm); N_{Re} is the Reynolds number which is calculated as $N_{Re} = W'G_g/\mu_g$,

195   where W' is the equivalent inner diameter of the honeycomb module (m) which is calculated as W' =

196    $\sqrt{4S/\pi}$, G_g is the mass flow rate of the air (kg s^{-1} m^{-2}), and μ_g is the air viscosity (kg m^{-1} s^{-1}); N_{Sc} is the

197   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

198   diffusion coefficient of the gaseous pollutant in the liquid phase (m^2 s^{-1}) which is calculated by the

199   Stokes-Einstein equation [37]; Γ is the liquid mass flow rate based on the wetted perimeter (kg s^{-1} m$^-$

200   ^{1}); ρ_w is the liquid density (kg m^{-3}); $B_F = (3\mu_w\Gamma/\rho_w g)^{1/3}$ in which μ_w is the liquid viscosity (kg m^{-1} s^{-1})

201   and g is the gravity acceleration (m s^{-2}).

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

203

204    $$Z_{CA} = \left[\frac{G_{my}}{K_g a_t}\right] \times \left[\int_{C_{out}}^{C_{in}} \frac{dC}{\left(1 - m\frac{V_m}{L_m}\right)C + \left(\frac{V_m}{L_m}C_{in} - C_{w,out}\right)}\right] \tag{8}$$

205

206   where L_m is the liquid molar flow rate (mole s^{-1}); $C_{w,out}$ is the outlet concentration of the gaseous

207   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

208    $C_{w,in}$ is the inlet concentration of the gaseous pollutant in the liquid phase (ppbv). Assuming that the

209   honeycomb modules are replaced by the traditional packing materials, the removal efficiency of the

210   wet scrubber packed with different raschig rings (equivalent diameter (D_e) = 6, 13 and 25 mm) and

211   berl saddle (D_e = 25 mm) are studied. The small raschig rings with D_e equal to 6 mm and 13 mm have

212   the specific surface areas 50% higher than (710 m^2 m^{-3}) and similar (370 m^2 m^{-3}) [36] to that of the

213   honeycomb module, respectively. Whereas, the commonly used raschig ring (D_e = 25 mm and a_t = 190

214   m^2 m^{-3}) [36] and berl saddle (D_e = 25 mm and a_t = 250 m^2 m^{-3}) [36] have the specific surface areas

215   much lower than that of the honeycomb module. The gas phase and liquid phase mass transfer

216   coefficients of the WS packed with the raschig rings and berl saddle are typically calculated by Eqs.

217   (S1) and (S2), respectively shown in Section S2 in the SI. The theoretical height of the raschig rings

218   and the berl saddle is then calculated by Eq. (8) and compared with that of the honeycomb module.

219   The pressure drop of the traditional WS is determined based on the correlation of $\dfrac{U_t{}^2 a_t \rho_g \mu_w{}^{0.2}}{g\epsilon^2 \rho_w}$ and

220    $\dfrac{G_w}{G_g}\sqrt{\dfrac{\rho_g}{\rho_w}}$ and compared to that of the HWS [36].

221   **3. Results and discussion**

3.1. Field test results

Fig. 2 shows the removal efficiency of the HWS versus time with a wide range of the inlet concentrations of 31,874–180,685 ppbv for HF, 12,552–72,582 ppbv for CH_3COOH; 1,384–15,203 ppbv for HCl, 1,234–12,390 ppbv for HNO_3, 201–6,017 ppbv for HNO_2, and 240–1,583 ppbv for H_2SO_4. Among these gases, HF and CH_3COOH are dominant and constitute 64.3 ± 9.95 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 figure shows that the removal efficiencies of each species are all very high, which are 95.6-98.3% ($RE_{ave} = 97.2 \pm 0.8\%$) for HF (Fig. 2a), 95.0-98.3% ($RE_{ave} = 96.9 \pm 1.1\%$) for CH_3COOH (Fig. 2a), 96.1-98.5% ($RE_{ave} = 97.7 \pm 0.7\%$) for HCl (Fig. 2b), 95.1-98.8% ($RE_{ave} = 97.5 \pm 1.2\%$) for HNO_3 (Fig. 2b), 95.0-97.9% ($RE_{ave} = 96.3 \pm 0.9\%$) for HNO_2 (Fig. 2c), and 95.2-97.6% ($RE_{ave} = 96.5 \pm 0.7\%$) 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 removal efficiency of the acidic gases follows the order as HCl>HNO_3>HF>HNO_2>CH_3COOH>H_2SO_4. Among these gases, HCl has the highest removal efficiency during 3.5 years although HCl has the lowest H ($H_{HCl} = 1.1 < H_{HNO2} = 49 < H_{CH3COOH} = 8.8 \times 10^3 < H_{HF} = 1.3 \times 10^4 < H_{HNO3} = 2.1 \times 10^5 < H_{H2SO4} = 2.9 \times 10^9 \, M \, atm^{-1}$) [38, 39]. On the other hand,

239   the dissociation constants of HCl ($K_{HCl} = 1.7 \times 10^6$ M atm^{-1}) is several orders of magnitude higher than

240   other acidic gases ($K_{H2SO4} = 10^3$, $K_{HNO3} = 15.4$; $K_{HF} = 6.3 \times 10^{-4}$; $K_{HNO2} = 5.1 \times 10^{-4}$; $K_{CH3COOH} = 1.7$

241    $\times 10^{-5}$ M atm^{-1}) [11, 38]. It indicates that H just reflects the physical solubility of the gases while

242   additional dissociation and chemical reaction constants also play role in the absorption process by WSs

243   [16, 38], which is explained in detail in Section 3.2.

244   It is seen that the high removal efficiency of the HWS does not change from the first operating

245   day (11/08/2016) until the latest testing day (19/03/2020) for CH$_3$COOH (1st: 96.3%; 13th: 97.4%) and

246   HCl (1st: 97.8%; 13th: 97.8%), respectively, while it is slightly decreases for 1.3% for HF (1st: 98.0%;

247   13th: 96.7%), 2.8% for HNO$_3$ (1st: 97.9%; 13th: 95.1%), 2.0% for HNO$_2$ (1st: 97.9%; 13th: 95.9%), and

248   1.2% for H$_2$SO$_4$ (1st: 96.9%; 13th: 95.7%), respectively. It may be due to the slight increasing pressure

249   drop which is to be discussed later. It is to be noted that the HWS was cleaned manually only once

250   every year by spraying water jet from the top of the modules during the test period (three cleaning

251   times in total, in July 2017, April 2018 and July 2019, respectively), which helped maintain $\geq$ 95%

252   high removal efficiency during this long-term test. In comparison, the packing materials of the

253   traditional packed tower need to be washed once every quarter to reduce particle clogging. After each

254   yearly maintenance denoted as "M" in Fig. 2, the removal efficiency is increased slightly for HF (1st

255   M: +1.4%; 2nd M: +1.5%) and is not affected for other gases which all maintain to be very high.

256   Unlike other traditional WSs, the removal efficiency of the HWS is not affected by the inlet

257   concentration, which is always higher than 95% for all acidic gases when C_{in} varies from sub-ppmv

258   (709 ± 324 ppbv in average) to supper-ppmv ($105,505 \pm 41,039$ ppbv in average) as shown in Fig. 3.

259   However, it is seen that the removal efficiencies of the HWS increase slightly by 3.5%, 2.2%, and

260   1.5% when the inlet concentration increases by 8.2, 5.6, and 3.9 times for HNO_3, HCl, and H_2SO_4,

261   respectively as shown in Fig. 3a. The slightly positive correlation of the removal efficiency and the

262   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

263   (p-value $= 0.02 < 0.05$), respectively. It is due to the variation of the inlet concentrations of HNO_3 and

264   HCl from low ($< 3,000$ ppbv) to high concentrations ($> 3,000$ ppbv). In Fig. 3a, it is found that the

265   removal efficiency of H_2SO_4 is lower than those of HNO_3 and HCl since the inlet concentration of

266   H_2SO_4 is lower than 1,600 ppbv and 9.2 times lower than HNO_3 and 11.2 times lower than HCl in

267   average. In comparison, the inlet concentrations of HF and CH_3COOH are always higher than 12,500

268   ppbv. Therefore, the removal efficiency also remains high and constant without an increasing trend, as

269   shown in Fig. 3b. The removal efficiency for HNO_2 also shows no dependence on the inlet

270   concentration which also varies from low ($< 3,000$ ppbv) to high concentrations (> 3000 ppbv) but

271   does not range widely ($C_{in-ave} = 2,899 \pm 1,596$ ppbv) as compared to that of HCl ($C_{in-ave} = 8,493 \pm$

272    $3,566$ ppbv) and HNO_3 ($C_{in-ave} = 6,969 \pm 3,353$ ppbv) with a wide concentration range (Fig. 3b).

273   During the 3.5-yr period, the pressure drop is found to be very small and increases only slightly

274   from 49.0 to 78.4 Pa (0.5 to 0.8 cmH_2O), which is also close to the theoretical value of 59.8 Pa (0.58

275   cmH_2O). The slight increase in the pressure drop during 3.5 years implies that only slight fouling of

276   the honeycomb material might have occurred which could not be cleaned thoroughly by water jet. In

277   Fig. 4a, it is found that the removal efficiency is decreased by 2.8% for HNO_3, 2.0% for HNO_2, 1.6%

278   for HF, and 1.2% for H_2SO_4 as the pressure drop is increased by 29.4 Pa (0.3 cmH_2O). The removal

279   efficiency shows a good negative correlation with the pressure drop for HNO_3 (R^2 = 0.88; p-value =

280   0.02 < 0.05) with 95% confident interval and H_2SO_4 (R^2 = 0.73; p-value = 0.06 < 0.1), HF (R^2 = 0.70;

281   p-value = 0.08 < 0.1), and HNO_2 (R^2 = 0.67; p-value = 0.09 < 0.1) with 90% confident interval showing

282   a slight influence of the pressure drop on the removal efficiency for these gases. On the other hand,

283   the removal efficiency of HCl and CH_3COOH does not show the dependence on the pressure drop as

284   shown in Fig. 4b since HCl has high dissociation constant and CH_3COOH has a very high inlet

285   concentration.

286   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

287   previous study [14] while the present scale-up, actual HWS device operated at a higher flow rate of

288   100 CMM, a higher L/G ratio of 26 L m^{-3}, and the same retention time of 0.5 s. Although the removal

289   efficiencies of HF, HCl, and CH_3COOH for the present HWS are not as high as those in the previous

290   study, which are about 99% for HF and HCl and nearly 100% for CH_3COOH, the long-term test results

291   show that the HWS always achieved removal efficiency higher than 95% stipulated in the emission

292   standard [1]. The inlet concentrations in the previous study (C_{in} = 100-450 ppbv for HF, 3,000-11,000

293   ppbv for HCl, and 600-6500 ppbv for CH_3COOH) were lower than the current HWS. Since there are

294   64 modules placed in parallel inside the HWS with some unused cross-section area (or dead space),

295   the required L/G ratio in the present study needs to be 1.4 times higher, which causes the pressure drop

296   to increase from 23.5 Pa to 49.0 Pa but it is still much lower than that of most traditional WSs.

297   The emission rates of each species in 12 sampling times were calculated from the outlet

298   concentrations and the air flow rate, and found to be less than 0.03, 0.04, 2.4×10^{-3}, 4.0×10^{-3}, $3.2 \times$

299   10^{-3}, and 1.8×10^{-3} kg h^{-1} for HF, CH_3COOH, HCl, HNO_3, HNO_2, and H_2SO_4, respectively. It indicates

300   that the exhausted gaseous acidic pollutants from the manufacturing process of the fab are well

301   controlled with only 2.5%, 3.0%, 0.3%, 0.4%, 0.2% and 0.6%, respectively, of the total regulated mass

302   emission rates (0.1 kg h^{-1} for H_2SO_4 and 0.6 kg h^{-1} for other gases) for a semiconductor manufacturing

303   fab. In addition, the bad odor due to high HF and CH_3COOH discharge concentrations disappears after

304   the installation of the present HWS.

305   *3.2. Results of theoretical prediction*

306   Fig. 5 shows the theoretical removal efficiencies of the HWS with the D x Z = 0.3 x 0.3 m for

307 different acidic gases. It is seen that the RE_{GK} is about 100% for all the acidic gases indicating that the

308 pollutants diffuse completely from the gas phase into the liquid phase fast enough without limiting the

309 ensuring dissociation and reaction processes of the pollutants in the water film. Whereas, the RE_{CA}

310 based on H (RE_{CA-H}) is 99.7% for HF, 98.0% for CH_3COOH, 12.6% for HCl, 99% for HNO_3, 91.9%

311 for HNO_2, and 97.8% for H_2SO_4, respectively indicating that the mass transfer resistance across the

312 gas-liquid interface is high for HCl, moderate for HNO_2, and very low for other acidic gases. It is

313 because that the solubility of the HCl and HNO_2 in the liquid is several orders of magnitude lower than

314 other acidic gases. The RE_{CA} based on H^* (RE_{CA-H^*}) of HCl and HNO_2 is increased to 99.3% and

315 99.2%, respectively when H^* is used. This implies that the dissociation and reaction are the rate

316 limiting steps in the HWS and the chemical reaction enhances the absorption rate and increases the

317 dissolved capacity of the liquid film in the HWS. The RE_{CA-H^*} results are close to the experimental

318 data with less than +3% biases.

319   To achieve the removal efficiency as high as the experimental data, the theoretical heights based

320 on H^* (Z_{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

321 for HNO_2, and 0.16 m for H_2SO_4, respectively, as shown in Fig. S2 in the SI. It indicates that the

322 theoretical values are very similar to the designed value of 0.3 m for mixed acidic gas control. The

323 theoretical heights based on H (Z_{CA-H}) are also shown in Fig. S2 in the SI, which demonstrate that it is

324 required longer module to achieve the removal efficiency $\geq 95\%$ for HCl ($Z_{CA-H} = 8.5$ m) and HNO$_2$

325 ($Z_{CA-H} = 0.4$ m) when the scrubbing NaOH solution is not added. In summary, the equations based on

326 the two-film theory can be used to predict the theoretical removal efficiency of the HWS and

327 theoretical height of the honeycomb module with good accuracy.

328   Fig. 6 shows the theoretical removal efficiency (RE$_{CA-H*}$) and the theoretical height (Z_{CA-H*}) of

329 the WS packed with the traditional packing materials including raschig rings ($D_e = 6$, 13, and 25 mm)

330 and berl saddle ($D_e = 25$ mm). As compared with the honeycomb module with the similar specific

331 surface area, theoretical removal efficiencies of the WS packed with 13-mm raschig ring are high for

332 HF, HCl and H$_2$SO$_4$ (RE$_{CA-H*} \geq 99.9\%$) but low for CH$_3$COOH (RE$_{CA-H*} = 62.3\%$) and HNO$_2$ (RE$_{CA-}$

333 $_{H*} = 17.2\%$). It is because CH$_3$COOH has the lowest dissociation constant among these acidic gases

334 and both Henry's law and dissociation constants of HNO$_2$ are several orders of magnitude lower than

335 other acidic gases. When the 6-mm raschig ring with a higher specific surface area than the honeycomb

336 module is used, the removal efficiencies can be achieved 95% for CH$_3$COOH and 52.6% for HNO$_2$,

337 respectively. However, the removal efficiencies of the traditional WS packed with 6-mm rachig rings

338 are still lower than those of the HWS for CH$_3$COOH ($Z_{CA-H*} = 98.1\%$) and HNO$_2$ ($Z_{CA-H*} = 99.2\%$). It

339 implies that HWS enhances the mass transfer rate of CH$_3$COOH and HNO$_2$ which have low solubility

340 and dissociation ability due to its multiple small gaps with uniform water-absorbing surface.

341 Additionally, the theoretical pressure drop of the WS packed with the 6-mm and 13-mm raschig rings

342 are about 490 Pa (5.0 cmH$_2$O) and 245 Pa (2.5 cmH$_2$O), respectively, which are much higher than that

343 of the HWS (59.8 Pa or 0.58 cmH$_2$O). The theoretical heights of the WS packed with the 6-mm and

344 13-mm raschig rings need to be as large as 1.34 m and 5.3 m, respectively to achieve the removal

345 efficiency higher than 95% for all the acidic gases for mixed gas control as shown in Fig. 6b. That is,

346 the HWS is an economical control device for mixed gas control as compared with the traditional WS.

347   To reduce the pressure drop, the WS is usually packed with bigger packing materials with lower

348 specific surface areas such as 25-mm raschig ring or 25-mm berl saddle. However, the removal

349 efficiency of the traditional WS packed with 25-mm perl saddle is not only as low as 29.1% for

350 CH$_3$COOH and 6.42% for HNO$_2$ but just 92% for H$_2$SO$_4$, which does not meet the removal efficiency

351 requirement. It is because that the inlet concentrations of H$_2$SO$_4$ are very low (C$_{in}$ = 0.24-1.58 ppmv)

352 as compared to those of the acidic gases. Although the removal efficiency of the WS packed with 25-

353 mm raschig ring is also as high as 95.3% for H$_2$SO$_4$, it is still lower than that of the HWS and the WS

354 packed with 6-mm and 13-mm raschig rings, which is due to its lower specific surface area. This

355 indicates that the HWS outperforms the traditional WS with the similar or even larger specific surface

356 areas for the removal efficiency of the mixed gas and pressure drop.

357 **4. Conclusion**

358   This work evaluated the long-term performance of the honeycomb wet scrubber for removing

359   gaseous acidic gases and odor at a semiconductor manufacturing fab in 3.5 years from August 2016 to

360   March 2020. The results of 12 removal efficiency tests indicated that the HWS is capable of removing

361   the gaseous acidic gases and odor with the inlet concentrations varying from low concentrations of

362   240–1,583 ppbv for H_2SO_4 and 201–6,017 ppbv for HNO_2 to moderate concentrations of 1,234–12,390

363   ppbv for HNO_3 and 1,384–15,203 ppbv for HCl, and high concentrations of 12,552–72,582 ppbv for

364   CH_3COOH and 31,874–180,685 ppbv for HF. All removal efficiency data, which are 95.6%–98.3%

365   for HF, 95.0%–98.3% for CH_3COOH, 96.1%–98.5% for HCl, 95.1%–98.8% for HNO_3, 95.0%–97.9%

366   for HNO_2, and 95.2%–97.6 for H_2SO_4, respectively, met the regulation for the control devices in

367   Taiwan. The pressure drop of the HWS maintained as low as 49.0–78.4 Pa (0.5-0.8 cmH_2O) and

368   increased only slightly during the 3.5-yr period, which is much lower than that of the traditional WSs.

369   The theoretical removal efficiency and the theoretical height of the HWS module were close to the

370   experimental data indicating that the theoretical equations can facilitate the design of the HWS. The

371   HWS showed higher theoretical removal efficiency and lower theoretical height for the mixed gas

372   control, especially for the acidic gases with low solubility and dissociation ability as compared with

373   the WS packed with the traditional packing materials. That is, the honeycomb wet scrubber can be

374   used as an efficient device for mixed acidic gas and odor control with high removal efficiency and low

375 pressure drop for the long-term.

376 **<u>Declarations</u>**

377 **Acknowledgments**

382 **Authors' contributions**

383 YLL and BTW designed the study and carried out the field tests. GYL and ZL contributed to data

384 analysis. GHH drafted and edited the manuscript. LTC contributed to data analysis, modeling, and

385 manuscript preparation. DYHP and CJT critically reviewed and edited the final manuscript. All the

386 authors read and approved the final manuscript.

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390 **Availability of data and materials**

391 All data generated or analyzed during this study are included in this article and its supplementary

392 information file. The raw data are available from the corresponding author upon reasonable request.

Competing interests

394 The authors declare they have no competing interests.

Nomenclature

a_t	Specific surface area (m^2 m^{-3})	N_{RE}	Reynolds number
C_{in}	Inlet concentration (ppbv)	N_{Sc}	Schmidt number
C_{out}	Outlet concentration (ppbv)	Q	Airflow rate (m^3 s^{-1})
$C_{w,in}$	Inlet concentration of the gaseous pollutant in the liquid phase (ppbv)	R	Gas constant (m^3 Pa K^{-1} $mole^{-1}$)
$C_{w,out}$	Out concentration of the gaseous pollutant in the liquid phase (ppbv)	RE	Experimental removal efficiency (%)
D	The inner diameter of the packing material (m)	RE_{CA}	Theoretical removal efficiency based on two film theory (%)
D_e	The equivalent diameter of the raschig ring (m)	RE_{GK}	Theoretical removal efficiency based on diffusion theory (%)
D_g	Gas-phase diffusion coefficient (m^2 s^{-1})	S	Empty cross-section surface area (m^2)
D_w	Liquid-phase diffusion coefficient (m^2 s^{-1})	T	Temperature (K)
g	Gravity acceleration (m s^{-2})	U_t	Superficial air velocity (m s^{-1})
G_g	The mass flow rate of the air (kg s^{-1} m^{-2})	V_m	The molar flow rate of the air ($mole$ s^{-1})

G_w	Mass flow rate of the liquid (kg s^{-1} m^{-2})	W	Gap distance of the honeycomb module (m)
G_{my}	The molar flux of the air (mole s^{-1} m^{-2})	W'	The equivalent diameter of the honeycomb module (m)
H	Henry's low constant (atm M^{-1})	Z	Height of the honeycomb module (m)
H*	Effective Henry's low constant (atm M^{-1})	Є	Fractional voids in dry packing
K	Dissociation constant (M atm^{-1})	ρ_g	Air density (kg m^{-3})
K_g	Overall mass transfer coefficient (mole s^{-1} m^{-2})	ρ_w	Liquid density (kg m^{-3})
k_g	Gas phase mass transfer coefficient (mole s^{-1} m^{-2})	μ_g	Air viscosity (kg s m^{-1})
kw	Liquid phase mass transfer coefficient (mole s^{-1} m^{-2})	μ_w	Liquid viscosity (kg s m^{-1})
L	The total length of the PP fabric plate (m)	ξ	Dimensionless parameter
L_m	Liquid molar flow rate (mole s^{-1})	Γ	Liquid mass flow rate based on the wetted perimeter (kg s^{-1} m^{-1})

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486 **Table 1** Operating parameters of the HWS at a semiconductor fab and the design criteria of TWEPA.

Operating parameter	Value	Criteria	Unit
[a]Wetting factor	0.1	≥ 0.1	$m^2\ h^{-1}$
Retention time	0.5	≥ 0.5	sec
Pressure drop	49~78.4	—	Pa
Airflow rate	100	—	$m^3\ min^{-1}$
Liquid flow rate	2,600	—	$L\ min^{-1}$
pH	7.5~9.0	≥ 7	—
Packing material	483	≥ 90	$m^2\ m^{-3}$

487 [a] Wetting factor = scrubbing water flow rate divided by (the specific surface area of the packing materials times the

488 horizontal cross-sectional area of the tower).

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Table 2 Some typical packing materials.

Packing materials	[a] D×Z or L×W_T×Z (cm)	Bulk density (kg m^{-3})	Specific surface area (m^2 m^{-3})	L/G ratio (L m^{-3})	Pressure drop (Pa)	Reference
Honeycomb	**30*30**	**160**	**483**	**26**	**[b] 59.8**	**Present**
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]

501   [a] Packing dimensions: diameter × height (D×Z) or length × width × height (L×W_T×Z); [b] Theoretical value

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509 **Figure captions**

510 **Fig. 1 a** Experimental setup of the HWS at a semiconductor fab, and **b** pictorial view of honeycomb

511 packing installed in the HWS.

512 **Fig. 2** Removal efficiency versus test date for **a** HF and CH_3COOH, **b** HCl and HNO_3, and **c** HNO_2

513 and H_2SO_4 (Note: (**M**): maintenance day).

514 **Fig. 3** Removal efficiency versus inlet concentration for **a** HCl, HNO_3, and H_2SO_4, and **b** HF,

515 CH_3COOH, and HNO_2.

516 **Fig. 4** Removal efficiency versus pressure drop for **a** HF, HNO_2, HNO_3, and H_2SO_4, and **b** HCl and

517 CH_3COOH.

518 **Fig. 5** Comparison of the experimental and theoretical removal efficiencies for different acidic gases

519 (Note: the data are slightly shifted to avoid overlapping).

520 **Fig. 6** Comparison of **a** the theoretical removal efficiency, and **b** the theoretical height of the traditional

521 WS and the HWS (Note: the data are slightly shifted to avoid overlapping).
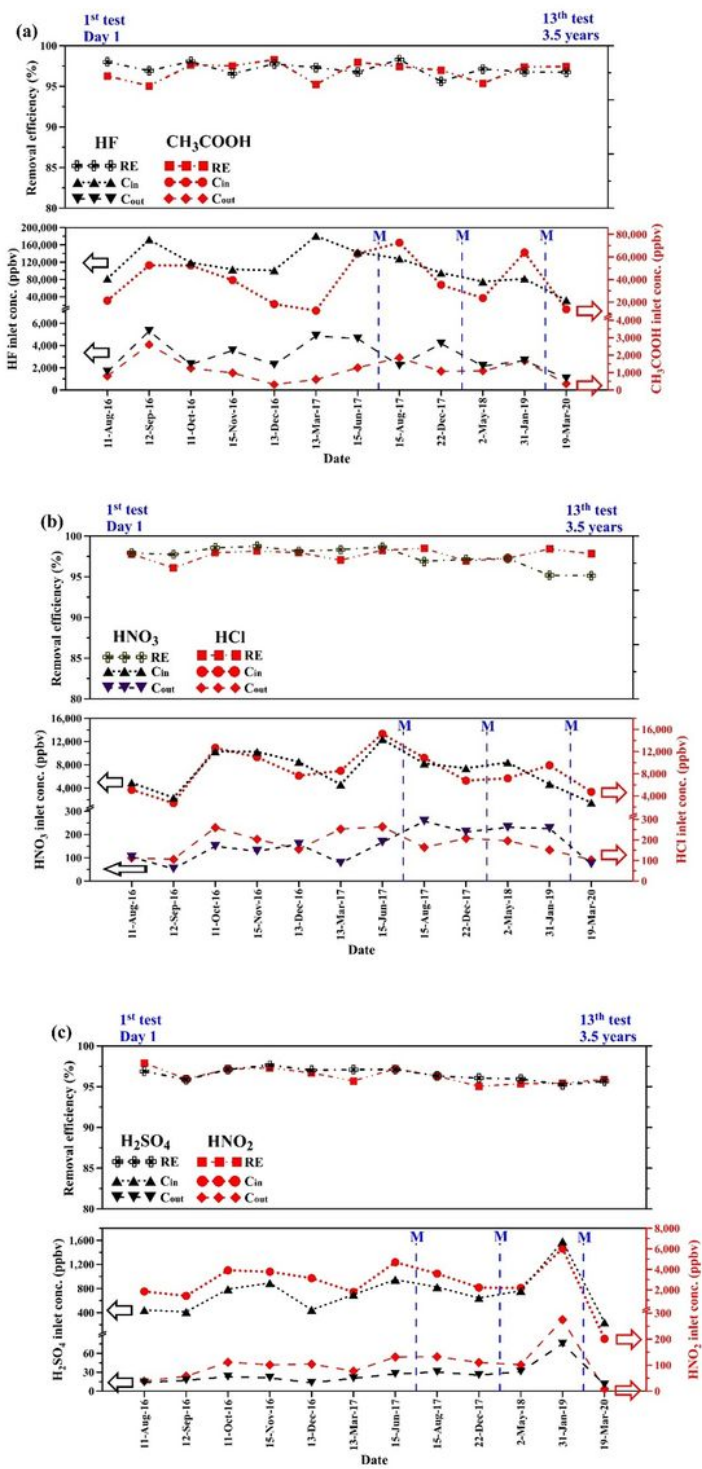
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Figures

(a)

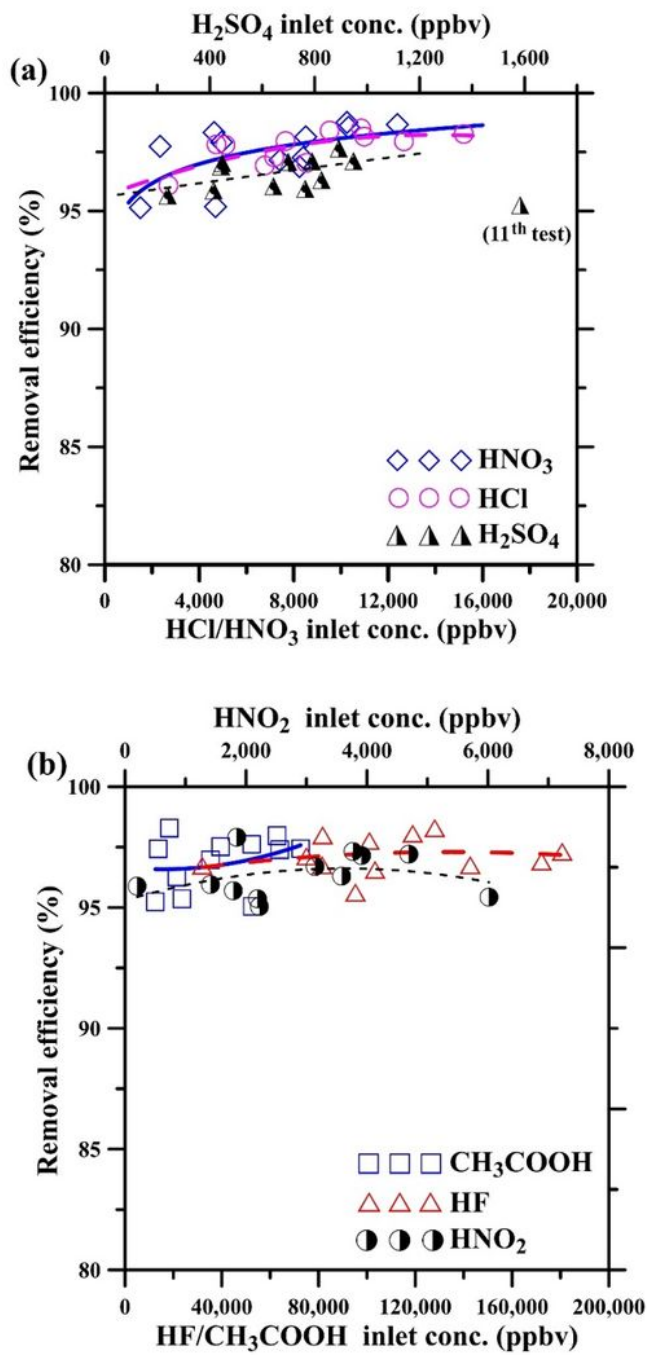Honeycomb wet scrubber — Scrubbing liquid — exhaust gas
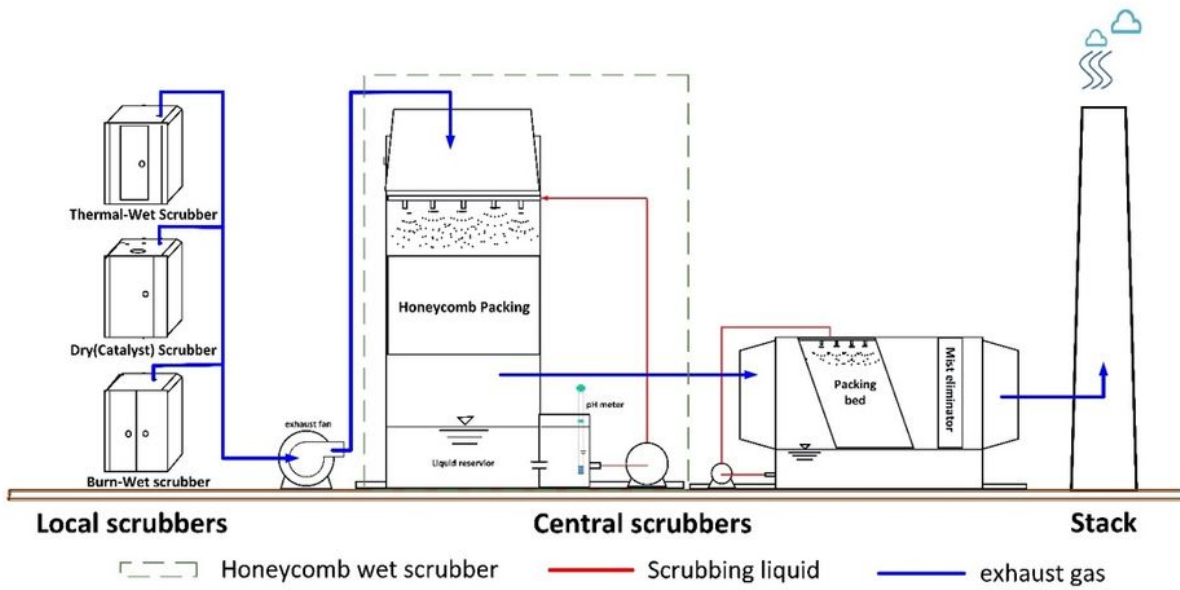
(b)

Figure 1

a Experimental setup of the HWS at a semiconductor fab, and b pictorial view of honeycomb packing installed in the HWS.

Figure 2

Removal efficiency versus test date for a HF and CH3COOH, b HCl and HNO3, and c HNO2 and H2SO4 (Note: (M): maintenance day).

Figure 3

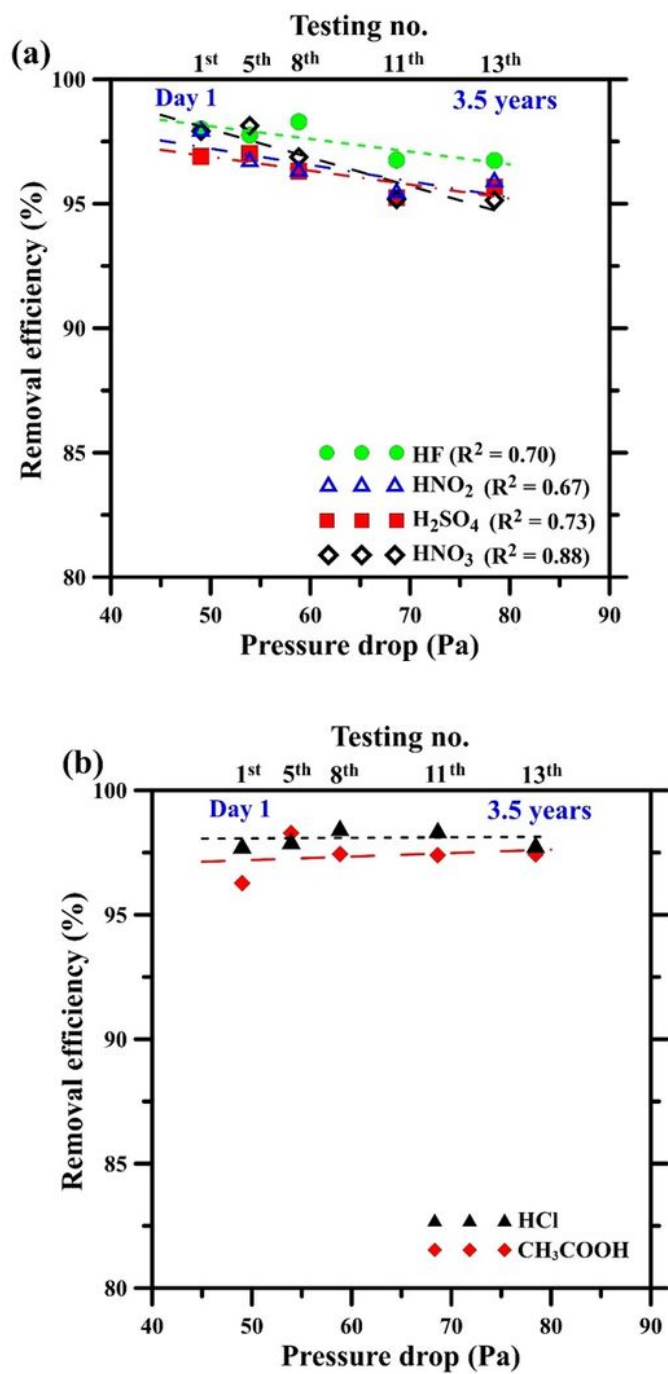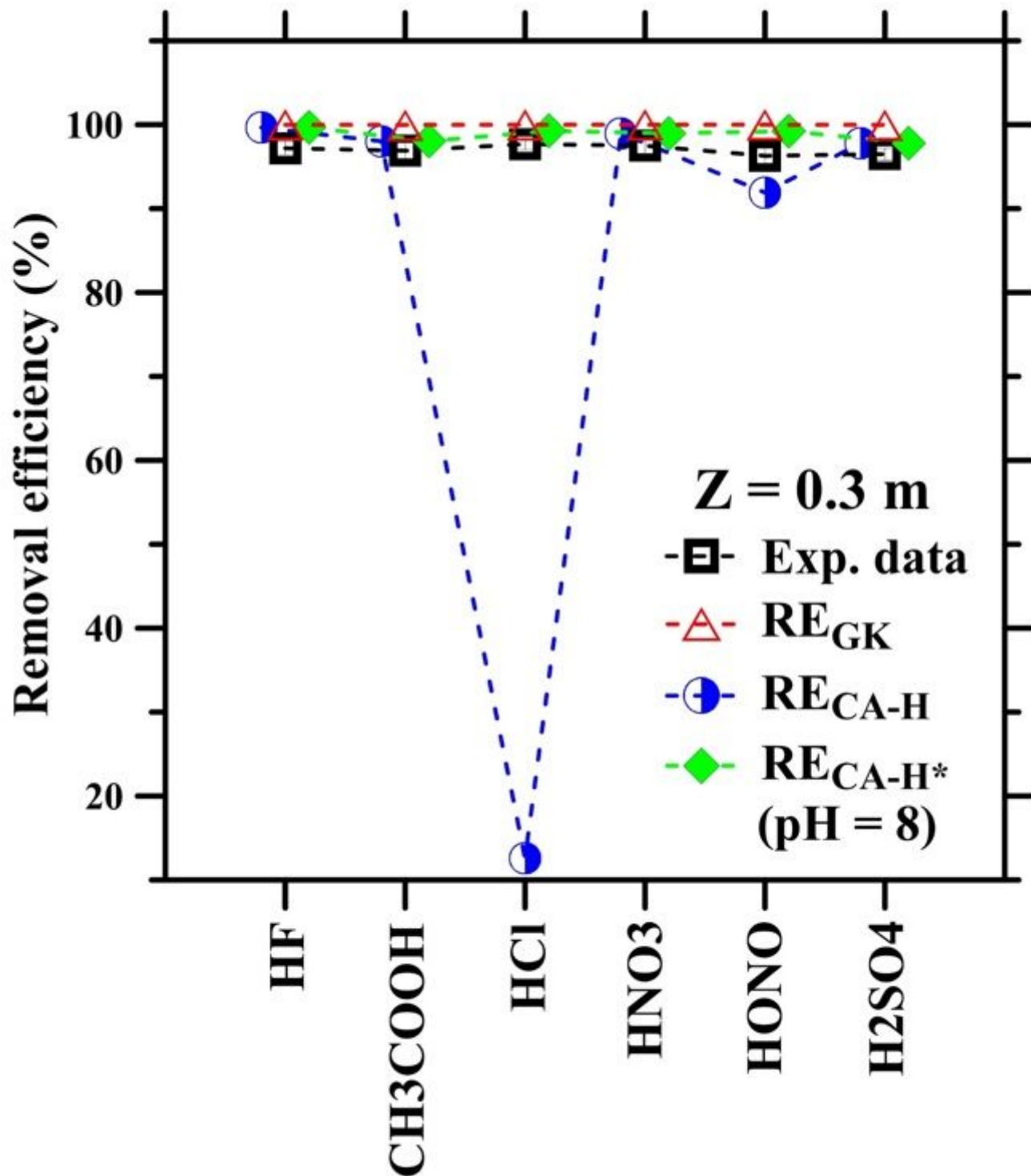Removal efficiency versus inlet concentration for a HCl, HNO3, and H2SO4, and b HF, CH3COOH, and HNO2.

Figure 4

Removal efficiency versus pressure drop for a HF, HNO2, HNO3, and H2SO4, and b HCl and CH3COOH.

Figure 5

Comparison of the experimental and theoretical removal efficiencies for different acidic gases (Note: the data are slightly shifted to avoid overlapping).
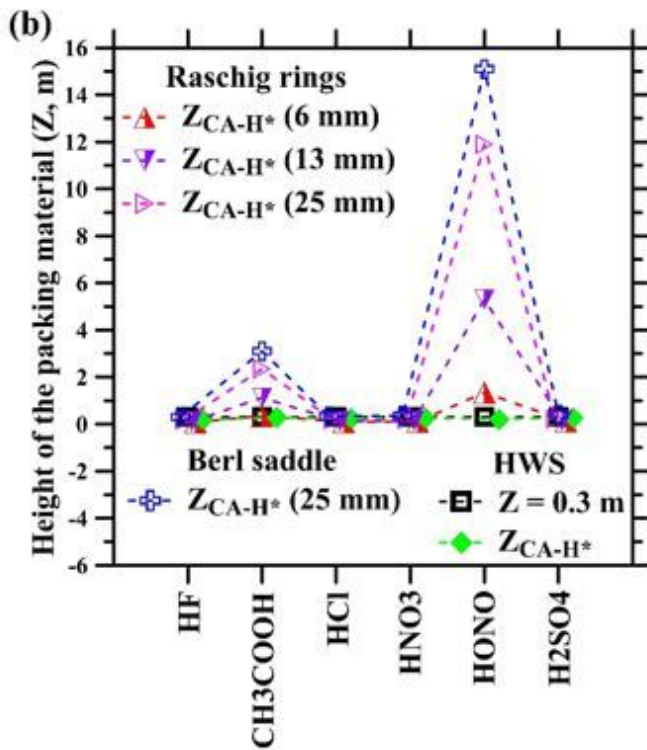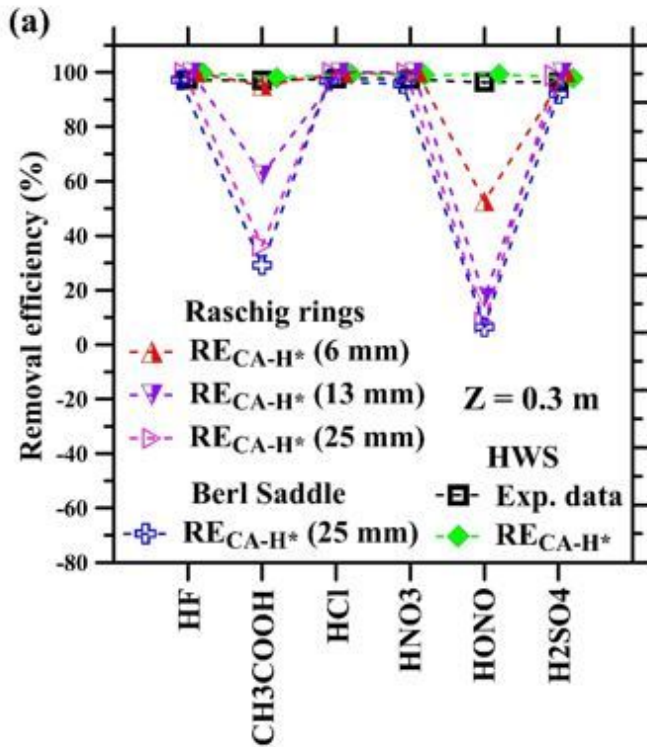
Figure 6

Comparison of a the theoretical removal efficiency, and b the theoretical height of the traditional WS and the HWS (Note: the data are slightly shifted to avoid overlapping).