

Assessment of adsorptive filter for removal of formaldehyde from indoor air

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ABSTRACT

We examine vary face velocity and initial formaldehyde concentration to investigate formaldehyde removal performance of the coconut shell activated carbon (AC) adsorptive filter media. AC surface is smooth and distributed with small pores. The surface of AC is also rather uneven, coarse and porous. Irregular layer structure were formed the amorphous. C, O, Mg, P etc. were detected, which showed the existence of MgO in AC. AC surface area is $1333.3304 \text{ m}^2 \text{ g}^{-1}$, ketone -C=O bonds were successfully grafted on carbon. At any given face velocity, the experimental results present that adsorption capacity is increased and breakthrough time is reduced as initial concentration is increased. The breakthrough behavior of AC adsorptive filter could henceforth be investigated with confidence using the breakthrough curves predicted by the Yoon-Nelson model. Among three kinetic models, the experimental and calculated results show that the correlation coefficient and mean absolute performance error (MAPE) of Pseudo-second-order kinetic model presented the best approximation of the adsorption kinetic process dynamics than Pseudo-First Order Kinetic model and Intraparticle Diffusion model. Both of the

25 Intraparticle diffusion model and membrane diffusion affected the overall rate of AC adsorptive
26 filter media adsorption process by more than one step. The equilibrium data of AC adsorptive filter
27 media was found to best fit to the Langmuir model. The D-R equation predicted the equilibrium
28 capacity of formaldehyde onto AC at relative pressures 0.151.

29
30 **Keywords:** *Adsorption; breakthrough; kinetic model; Formaldehyde*

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38 **1. Introduction**

39 Human beings typically spend more than 90 % of their waking hours in indoor environments.
40 As such, pollutants found in indoor air may in many cases represent a higher hazard to human
41 health than those present in the outdoor environment (**Brunsgaard et al. 2012**). Recent changes to
42 Building Regulations have demanded much higher air-tightness requirements for residential and
43 commercial buildings for reasons of improved energy conservation As a result there exists the
44 potential for increased concentrations of volatile organic compounds (VOCs) within indoor
45 environments. The negative impact on health and well-being and comfort (**Chuck and Kim, 2013**)
46 of occupants in air-tight housing is well documented. Among VOCs, formaldehyde (HCHO) has
47 long been established as a hazardous and harmful substance (**Mui et al., 2008**). The primary
48 source of formaldehyde found as a pollutant in indoor air is pressed wood products manufactured

49 using urea formaldehyde resins (**U.S. Consumer Product Safety Commission, 2015**).

50 There are several studies on air cleaning technologies for indoor formaldehyde removal. UV
51 photolysis is limited due to its trend to produce ozone and radicals with harmful effects.
52 Photocatalytic oxidation has been utilized for ppm levels of contaminant degradation and ppbv
53 concentrations need to be measured, and their main deficiencies are the short lifetime of the catalyst
54 and the creation of harmful by-products. Cold or non-thermal plasma are not yet on the market.
55 (**Luengas et al., 2015**) These major technical issues also have limitations like high energy
56 consumption and low removal efficiency (**Gilbert et al., 2008, Mo et al., 2008**). Biological
57 degradation (**Lu et al., 2012**) and botanical air filtration are still under development, currently
58 operating at low efficiency and with insufficient understanding of long-term safety (**Luengas et al.,**
59 **2015; Wang et al., 2014**). At this time, neither of these techniques is broadly employed for indoor
60 pollutant removal (**Lu et al., 2012**). Activated carbon adsorption is a simple, effective, and low cost
61 method to reduce the concentrations of formaldehyde removal in indoor air (**Lu et al., 2015**).

62 The primary purpose of this study characterized the physical and chemical properties of the AC
63 by Scanning electron microscope (SEM), X-ray diffraction (XRD), Energy Dispersive X-Ray
64 Spectroscopy (EDS), Brunauer–Emmett–Teller (BET), and Fourier transform infrared spectroscopy
65 (FTIR) analysis. To investigate the equilibrium, breakthrough behavior, and the applicable
66 assessment of three kinetic models: Pseudo-first-order, Pseudo- second-order and intraparticle
67 diffusion. The latter assessment was conducted under various experimental conditions ,using the
68 correlation coefficient (R^2) and the mean absolute percentage error (MAPE) to characterize the
69 kinetic data. As a result, this study provided the essential parameters to consider when designing
70 and operating an adsorptive purifier for removal of formaldehyde from indoor air, and
71 implementing strategies to improve air quality in the indoor environment.

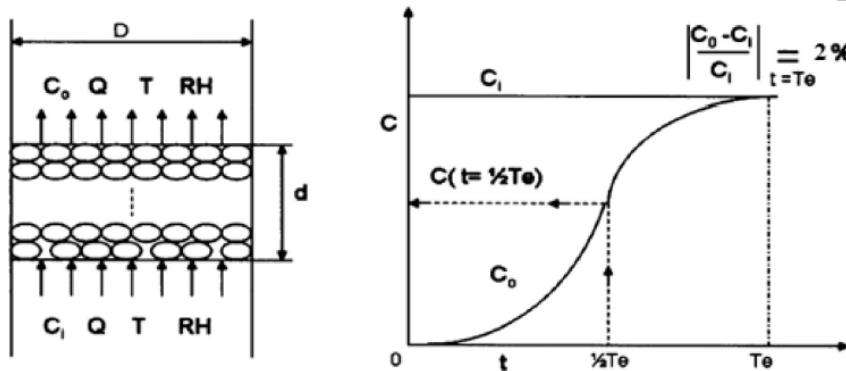
72 **2. Materials and methods**

73 **2.1. Principle**

74 **2.1.1. Adsorptive capacity**

75 This study used the testing method of VanOsdell et al. (1996) shown in Fig. 1. The mixed air
76 passes through the adsorbent. Removal efficiency (η) was calculated with the initial concentration
77 (C_0). Adsorption capacity (q_t) is defined as follow (Shiue et al. 2010):

78
$$q_t = C_0 V \int \eta(t) dt \times \frac{1}{0.082} \times \frac{1}{273 + K} \times M \times 10^{-6} \quad (1)$$



79
80 **Fig. 1. Schematic diagram of testing theory**

81 **2.1.2. Breakthrough**

82 Generally, it is not practicable to measure activated carbon filter breakthrough time against
83 formaldehyde concentration in real indoor environments. When the outlet concentration reached 2%
84 of the initial concentration, the breakthrough time was determined (Dehdashti et al., 2011). While
85 changing only formaldehyde concentration, simplified breakthrough time estimation is shown as
86 (Cheng, 2008; Cheng and Tsai, 2007; Shiue et al, 2010):

87
$$\frac{t_{b,1}}{t_{b,2}} = \left(\frac{C_{0,1}}{C_{0,2}} \right)^\alpha \quad (2)$$

88 where t_b is the breakthrough time, C_0 is the initial (inlet) concentration of adsorbate (ppmv), α is the
89 constant. Equation (2) predicted the performance of the activated carbon filter.

90 Yoon and Nelson (1984) initially developed a simple model that proposed the rate of decrease
91 in the probability of adsorption for each adsorbate molecule is proportional to the probability of

92 adsorption of an adsorbate (P_{ads}) and the probability of breakthrough of the adsorbate ($1-P_{ads}$):

$$93 \quad \frac{-dP_{ads}}{dt} = k'P_{ads}(1-P_{ads}) \quad (3)$$

94 Integrating Eq (3), and defining τ as the time required for 50% breakthrough (i.e., $P_{ads} = 0.5$,
95 hereinafter referred to as the stoichiometric breakthrough time),

$$96 \quad t_b = \tau + \frac{1}{k'} \ln \frac{C_t}{C_0 - C_t} \quad (4)$$

97 where k' is a rate constant, C_t is the breakthrough (effluent) concentration of adsorbate. The
98 calculation of theoretical breakthrough curves for a single-component system requires the
99 determination of the parameters k' and t for the adsorbate of interest.

100 In accordance with Eq. (4), the value of k' can be got from the slope of the plot $\ln [C_t/(C_0 - C_t)]$
101 versus breakthrough time t , and the value of t (50% breakthrough time) can be decided as the time
102 at $Q = 0.5$ (i.e., $C_t = C_0/2$). Applying k' and t of Eq. (4), one may cause the complete breakthrough
103 curve for a given set of experimental conditions. Furthermore, Yoon and Nelson (1984) have
104 proposed the following equation:

$$105 \quad K = k't \quad (5)$$

106 where K is a proportionality constant for a given adsorbate and determined type of adsorbent. Both
107 k' and t are alternatively rely on the concentration of the adsorbate and the face velocity.

108 2.1.3. Kinetic model

109 The Pseudo-first-order equation is expressed by Lagergren equation (1898) as follow:

$$110 \quad \log(q_e - q_t) = \log q_e - \frac{k_1}{2.303} t \quad (6)$$

111 where q_e is the amount of adsorbed formaldehyde per unit weight of adsorbent, q_t is the amount of
112 adsorbate adsorbed at time t .

113 The pseudo-second-order model is showed as: (Ho and McKay, 1999)

$$114 \quad \frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \quad (7)$$

115 The intraparticle diffusion model is described as follows: (Guibal, 1998)

$$116 \quad q_t = k_i t^{1/2} + C \quad (8)$$

117 2.1.4. Error analysis

118 **Mandal et al., (2015)** used correlation coefficient (R^2) and mean absolute performance error
119 (MAPE) values to verify the experimental data for the most suitable kinetic model. These expected
120 indices were computed by utilizing the following equations:

$$121 \quad R^2 = \frac{[\sum_{i=1}^n (q_{pre} - \bar{q}_{pre})(q_{exp} - \bar{q}_{exp})]^2}{\sum_{i=1}^n (q_{pre} - \bar{q}_{pre})^2 \sum_{i=1}^n (q_{exp} - \bar{q}_{exp})^2} \quad (9)$$

$$122 \quad MAPE = \frac{1}{n} \sum_{i=1}^n \left| \frac{q_{exp} - q_{pre}}{q_{exp}} \right| \times 100 \quad (10)$$

123 where q_{pre} and q_{exp} are the expected and measured values of formaldehyde concentration,
124 respectively; \bar{q} illustrates the average values of related q ; n is the total number of observations.

125 2.1.5. Adsorption isotherm model

126 Between adsorbed and free formaldehyde in air, the equilibrium is given by the linearized
127 Langmuir isotherm model as per the following equation (Bernabe et al., 2015; Cheng and Tsai,
128 2007):

$$129 \quad \frac{C_e}{q_e} = \frac{1}{q_{max} K_L} + \left(\frac{1}{q_{max}}\right) C_e \quad (11)$$

130 where q_e is the adsorbed amount of formaldehyde per unit weight of adsorbent at equilibrium; C_e is
131 concentration of free formaldehyde in air; q_{max} is maximum adsorption capacity; and K_L is the
132 Langmuir's adsorption equilibrium constant related to the affinity of the binding sites, this
133 phenomena has been described before. (Bernabe et al., 2015; Cheng and Tsai, 2007)

134 Linearized the Freundlich isotherm model is shown as (Bernabe et al., 2015; Cheng and Tsai,

135 2007):

$$136 \quad \log q_e = \log k_f + 1/n \log C_0 \quad (12)$$

137 where k_F and n are Freundlich constants, which related to the adsorption intensity and adsorption
138 capacity, respectively. (Bernabe et al., 2015; Cheng and Tsai, 2007)

139 The Dubinin-Radushkevich (D-R) equation, which was initially formulated from the Polanyi
140 adsorption potential theory and represents molecular adsorption as a pore-filling phenomenon rather
141 than a layer-covering one, has been used to correct the Freundlich isotherms (Yao et al., 2009). In
142 accordance with the D-R equation the adsorption capacity (W), expressed as the adsorbed liquid
143 volume per unit mass of adsorbent, is related to the adsorption potential (A) as given by (Dubinin,
144 1966)

$$145 \quad \ln(W / W_0) = -kA^2 \quad (13)$$

146 and

$$147 \quad A = RT \ln(P_0 / P) \quad (14)$$

148 where W is the volumetric adsorption capacity, W_0 is the maximum adsorption space, k is the
149 parameter for each system of adsorbate and adsorbent, A is adsorption potential, R is the universal
150 gas constant, T is the absolute temperature, P_0 is the saturated vapor pressure at temperature T , and
151 P is the partial pressure of the adsorbate. Combined Eqs 13 and 14, the gas-phase partial pressure
152 related to the adsorption capacity as follows

$$153 \quad \ln W = \ln W_0 - k[RT(P / P_0)]^2 \quad (15)$$

154 2.2. Experimental methods

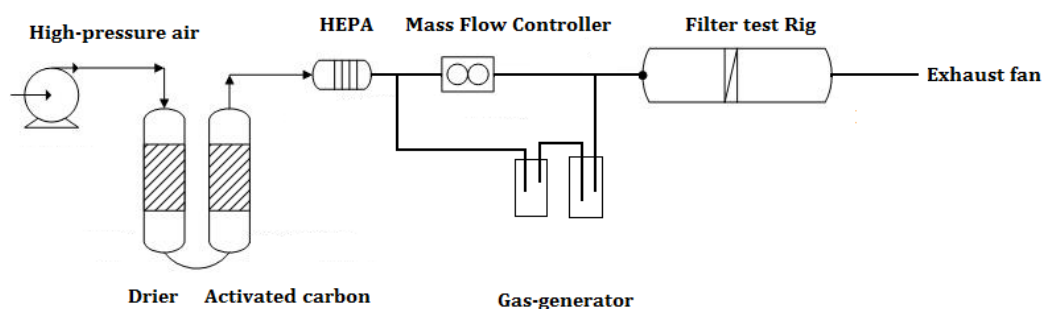
155 The 2.2 mm thickness AC-loaded nonwoven fabric filter media (provided by AIRREX Co.
156 Ltd.) was placed into a 10 cm × 10 cm testing rig located in a a temperature of 28 ±1°C by ambient
157 air-conditioning system. The testing airflow was controlled at relative humidity (RH) of 40 ± 2% by

158 adjusted primary split airflow. The characteristics of fabric filter media and schematic test system
159 diagram are shown in **Table 1** and **Fig. 2**, respectively.

160 **Table 1.** Characteristics of fabric filter media. (Shiue et al, 2010)

Raw material	Coconut shell
BET surface area (m^2g^{-1})	1145
Ash content (on dry basis) (%)	3%
Moisture content (%)	20~25%
pHzpc	6
Particle size, mesh	12 × 20 mesh
Thickness, e (mm)	2.2 mm
Weight per surface, W (g m^{-2})	702.8
Specific surface area, SBET (m^2g^{-1})	931.2
Pore volume, V_p (cm^3g^{-1})	0.6762
Microporous volume, $V_{\mu p}$ (cm^3g^{-1})	0.3264
Microporous volume percentage, % $V_{\mu p}$ (%)	0.48%
Median micropore diameter, $d_{\mu p}$	52.08 Å

161 The face velocity of filter media was rated at 0.25, 0.5, and 0.75 m/sec (VT110-VT115
162 Hotwire thermos-anemometer). A source of compressed air was first passed through a drier,
163 adsorbent, and high efficiency particulate air (HEPA) filter to generate a 'zero air'. This zero air
164 was then blended with formaldehyde via a gas-generator to produce the challenge gas entering the
165 test rig (**Fig. 1**). 99.9% grade formaldehyde solvent was filled into the gas-generator and submerged
166 in a water bath (temperature adjustable from 15°C to 25°C). The formaldehyde enters upstream of
167 the testing rig, controlled by passing air through a mass flow controller (Fujikin T1000).
168 Formaldehyde concentration was fixed at 0.25, 0.56 and 0.79 ppm with $\pm 2\%$ deviation. A real-time
169 formaldehyde analyzer (Formaldemeter htv-m) monitored the concentration of the challenge gas.



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Fig. 2. Schematic diagram of the experimental system setup

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2.3. Characterization

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Scanning electron microscope (SEM, ZEISS SIGMA Essential) was used to study the surface morphology of the adsorbent. X-ray diffraction (XRD) is for chemical characteristics and surface morphology of adsorbent study. Energy-dispersive X-ray spectrometry (EDS, ZEISS SIGMA Essential) was employed for the elemental analysis of the sample. To obtain an infrared spectrum of absorption or emission of adsorbent, Brunauer–Emmett–Teller (BET, Micrometrics ASAP2020) method was used to study the surface area. Fourier-transform infrared spectra (FTIR, Spectrum one) was applied.

180

3. Results and discussion

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3.1. Characterization

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3.1.1. Scanning Electron Microscope

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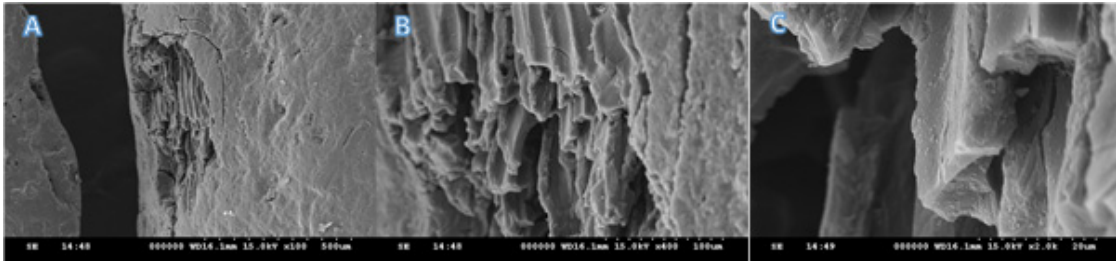
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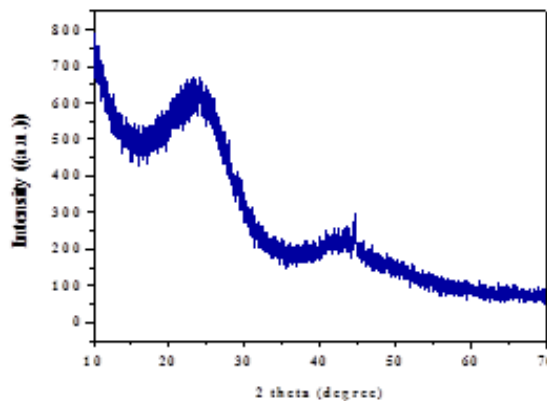
Fig. 3 shows the SEM image of AC under different magnification (100, 400, and 2,000) magnification. **Fig. 3(a)** indicated the surface is smooth and distributed with small pores. Moreover, it can observe a little crack and tiny particle near the pore. From **Fig. 3(b)** and **Fig. 3(c)** the surface of AC is rather uneven, coarse and porous. The shape is columnar and lamellar on the surface because the coconuts have the cellulose sintering at high temperature.



188
189 **Fig. 3.** SEM photographs of AC at (a) 100, (b) 400, (c) 2,000 magnifications.

190 **3.1.2. X-ray diffraction (XRD)**

191 **Fig. 4** indicated XRD pattern of AC. The graph was observed that AC is amorphous. The two
192 broad Bragg or diffraction peaks were showed around 22° and 43° indicated that the crystallites
193 were produced by two or more of these plates layer being stacked one above the other. Irregular
194 layer structure were formed the amorphous that the XRD diffraction peaks were broad.



195
196 **Fig. 4.** XRD image of AC.

197 **3.1.3. Energy Dispersive X-Ray Spectroscopy (EDS)**

198 **Fig. 5** illustrated the EDS detection area of AC, element such as: C, O, Mg, P etc. can be found
199 in the AC. AC is composed of C, O, and P accounting for 77.48%, 18.71%, 3.28% respectively. And
200 we can calculate the O/C ratio= 24.14% which means around 1.0 unit carbons are given by 24.14
201 unit oxygen. Some metal element like Mg with weight percentage 0.28%. Mg and P are detected in
202 the elemental analysis of AC. The EDS analysis showed that the Mg/C ratio=0.68%, which further

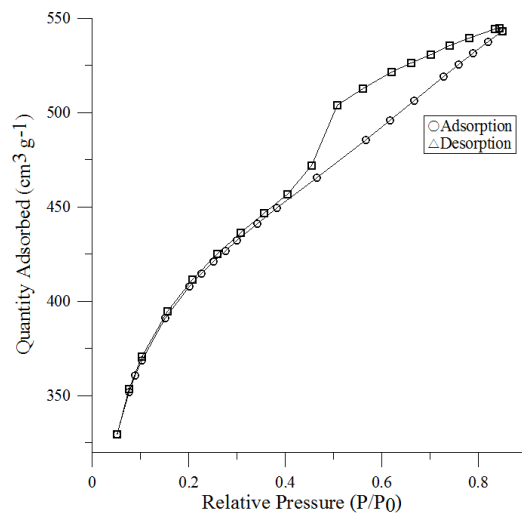
203 proves the existence of MgO in the coconuts activated carbons.



Fig. 5. EDS of AC.

207 3.1.4. Surface area and pore size

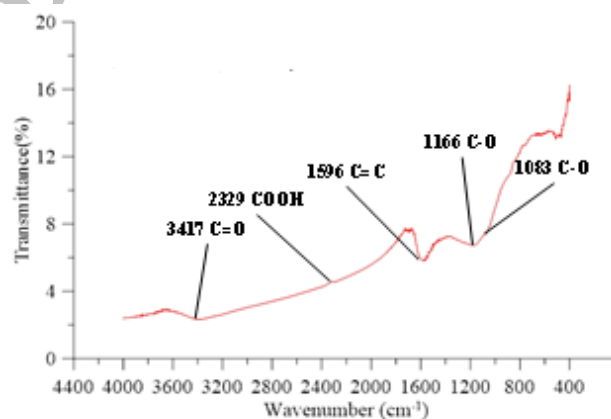
208 The BET analysis showed that the surface area was $1333.3304 \text{ m}^2 \text{ g}^{-1}$ after BET analysis. The
209 identified data showed that micropore volume of AC is $0.13 \text{ cm}^3 \text{ g}^{-1}$, meso volume is $0.45 \text{ cm}^3 \text{ g}^{-1}$,
210 total volume is $0.840442 \text{ cm}^3 \text{ g}^{-1}$; BJH adsorption average pore diameter is 2.21361 nm in the range
211 of $2.0\text{-}3.0 \text{ nm}$. The most important thing is the area between adsorption linear and desorption linear
212 in Fig. 6.



213
214 **Fig. 6.** Adsorption isotherms of N₂ at 77 K on AC.

215 **3.1.5. Fourier Transform Infrared Spectra**

216 **Fig. 7** shows FTIR spectroscopy in analysis of the functional groups. AC depended upon the
 217 experiments with molecular organic compounds that we can list the orders of the absorptive bands.
 218 It showed an intensive band 3417 cm⁻¹ has been explained by stretching of ketone -C=O bonds. A
 219 weak strength bonding at 2329 cm⁻¹ indicated the formation of O-H bonds from carboxylic acid
 220 (**Boehm 2002**). Near 1166 and 1086 cm⁻¹ has been explained by reverse stretching vibrations of
 221 carboxylic acids C-O bonds.

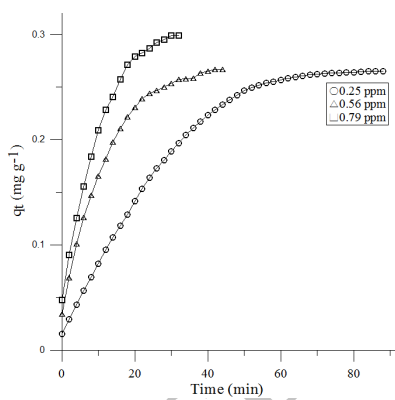


222
223 **Fig. 7.** FTIR image of AC.

224 **3.2. Adsorption capacity**

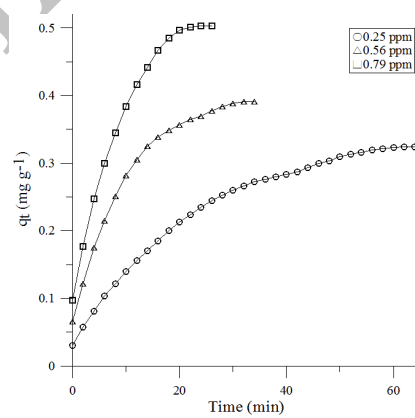
225 **Fig. 8** presents variations in formaldehyde adsorption capacity with different inlet
 226 concentrations at different face velocities. Equilibrium time (t_t) is defined as the time when the
 227 outlet concentration is 98% of the inlet concentration. The proportional relationship between natural
 228 logarithm of equilibrium time and formaldehyde adsorption capacity (Q) is presented as the formula
 229 (Scahill et al., 2004) in Table 2. These results are in agreement with Shiue et al. (2010) (flow rate
 230 of 0.076~0.152 m s⁻¹ at an inlet toluene concentration of 10~70 ppm). As shown in Fig. 8, if the
 231 inlet concentration of the adsorbate is increased, resulting in increased velocity of diffusion into the
 232 pores of AC, adsorption may reach equilibrium faster; the equilibrium time decreased from 86 to 30
 233 min, 64 to 24 min and 46 to 12 min, while adsorption capacity increased from 0.265 to 0.2991 mg g
 234 media⁻¹, 0.3244 to 0.4726 mg g⁻¹, and 0.4726 to 0.6019 mg g⁻¹ at 0.25, 0.5 and 0.75 m s⁻¹ face
 235 velocities, respectively.

236 (a)

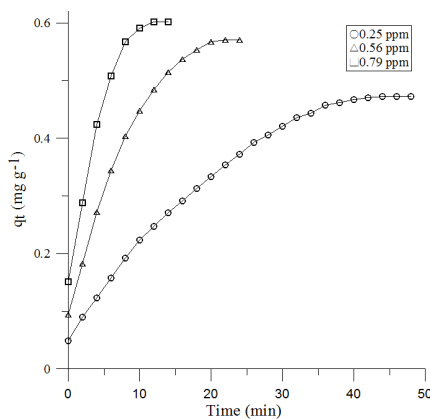


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(b)



238 (c)



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240 **Fig. 8.** Effect of concentration on formaldehyde adsorption capacity (a) 0.25m s⁻¹, (b) 0.5 m s⁻¹, (c)
 241 0.75m s⁻¹

242 **Table 2.** Correlations of formaldehyde adsorption capacity with time and concentrations.

Face velocity (m s ⁻¹)	C _i (ppm)	Adsorption capacity (<i>Q</i>) vs time (<i>t</i>)	Adsorption capacity (mg g ⁻¹)	<i>t</i> (min)
0.25	0.25	$Q = 0.081 \ln(t) - 0.0807, R^2 = 0.9596$	0.2650	76
	0.56	$Q = 0.0707 \ln(t) - 0.0098, R^2 = 0.9849$	0.2669	41
	0.79	$Q = 0.0836 \ln(t) - 0.0188, R^2 = 0.9861$	0.2989	33
0.5	0.25	$Q = 0.0902 \ln(t) - 0.005, R^2 = 0.9785$	0.3244	68
	0.56	$Q = 0.1022 \ln(t) - 0.0441, R^2 = 0.9904$	0.3906	38
	0.79	$Q = 0.1405 \ln(t) - 0.0638, R^2 = 0.9873$	0.5029	28
0.75	0.25	$Q = 0.1441 \ln(t) - 0.0815, R^2 = 0.9636$	0.4724	44
	0.56	$Q = 0.1695 \ln(t) - 0.0541, R^2 = 0.9905$	0.5701	19
	0.79	$Q = 0.1693 \ln(t) + 0.1882, R^2 = 0.9682$	0.6015	12

243 3.2. Breakthrough

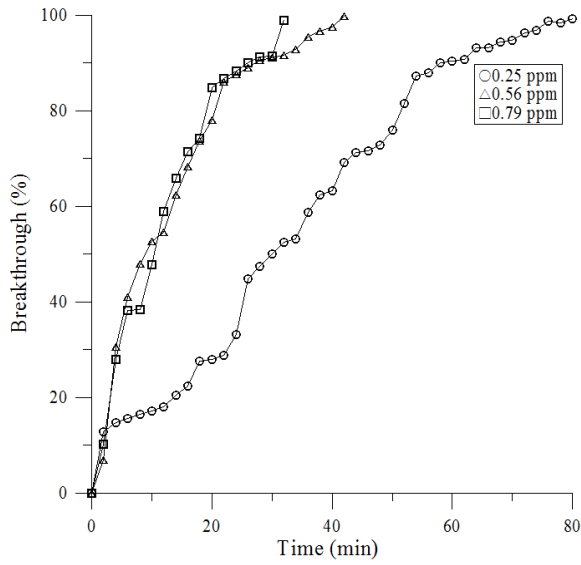
244 Complete breakthrough is specified as the point at which the measured formaldehyde
 245 concentration at the outlet equaled its inlet concentration. Breakthrough was executed by varying
 246 initial formaldehyde concentrations from 0.25 to 0.79 ppmv at 0.25 to 0.75 m s⁻¹ face velocity
 247 shown in **Fig. 9**. The breakthrough time was decreased from 78, 68, and 44 to 34, 28, and 14 min
 248 for 0.25 to 0.79 ppmv formaldehyde initial concentrations at 0.25 to 0.75 m s⁻¹ face velocity,
 249 respectively. This phenomena has been described before (**Jo and Chun, 2014; Cheng and Tsai,**
 250 **2007**). Additionally, the breakthrough behavior also was decreased from 78, 68, and 34 to 44, 28,
 251 and 14 min for 0.25 to 0.75 m s⁻¹ face velocity at formaldehyde initial concentration of 0.25, 0.56
 252 and 0.79 ppmv, respectively.

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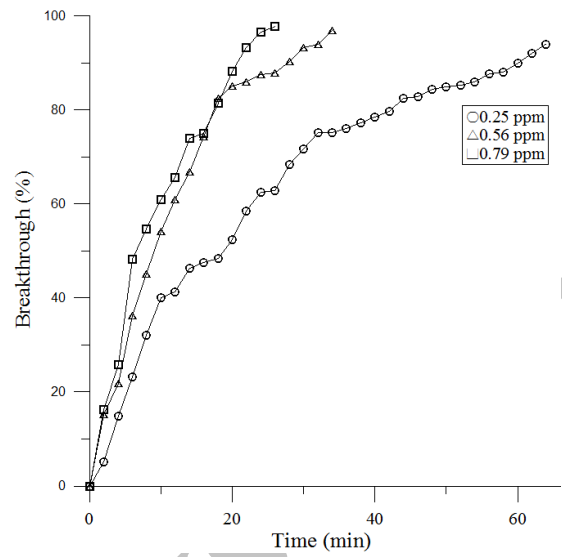
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256 (a)



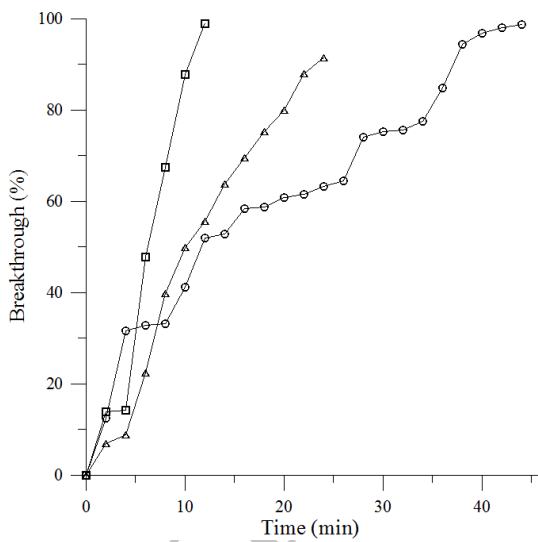
(b)



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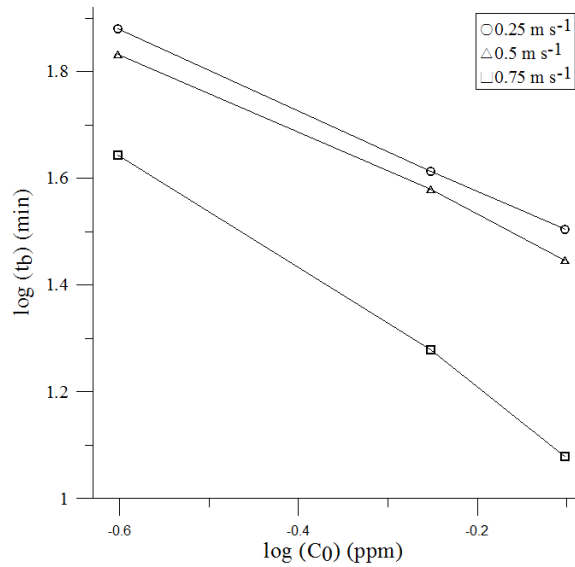
(c)



259

260 **Fig. 9.** The breakthrough curves of formaldehyde adsorption at various concentration and various
261 face velocity (a) 0.25 m s^{-1} , (b) 0.5 m s^{-1} , (c) 0.75 m s^{-1} .

262 The relationships between varies logarithmic formaldehyde initial concentrations and various
263 logarithmic breakthrough times are expressed in **Fig. 10** and **Table 3**. The slopes of the lines were
264 smaller for the formaldehyde with lower face velocity at the same concentration.



265

266

Fig. 10. The relationships between formaldehyde concentrations and breakthrough times.

267

Table 3. The relationships between formaldehyde concentrations and breakthrough times.

Face velocity (m s ⁻¹)	Breakthrough time vs Initial concentration
0.25	$t_b C_0^{0.1761} = 26.662, R^2 = 0.9998$
0.5	$t_b C_0^{0.1729} = 23.812, R^2 = 0.9975$
0.75	$t_b C_0^{0.077} = 9.526, R^2 = 0.9963$

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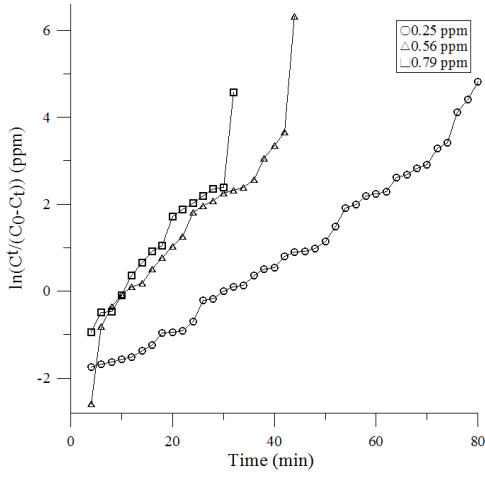
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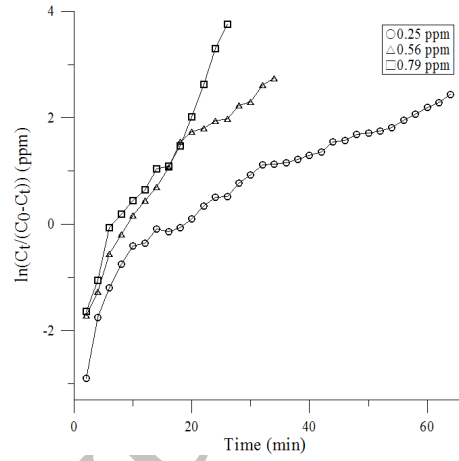
The plots of the equation 4 at various face velocities (**Fig. 11**) yielded linear curves with excellent correlations, the end-point of the regression analyses used 50% breakthrough (i.e., $\ln(c_b/c_i - c_b) = 0$). **Table 4** listed the values of the slopes (k') and the intercept of ($-k$). Both k' and τ are dependent on the adsorbate inlet concentration. The rate constant k' increased with increasing formaldehyde inlet concentration, as the stoichiometric breakthrough time τ decreased. Also, both k' and τ were significantly affected by the face velocity; i.e., the value of k' increased with increasing face velocity, while the value of τ decreased. Due to k' and τ act in reverse trend, one would predict that k would be a constant with a well-behaved breakthrough times presented in **Fig. 11**. Certainly, as interpreted in **Table 4**, the value of proportionality constant K (2.60 ± 0.47)

277 could be utilized as a basis of prevision for other hypothetical formaldehyde concentrations and face
 278 velocities.

279 (a)

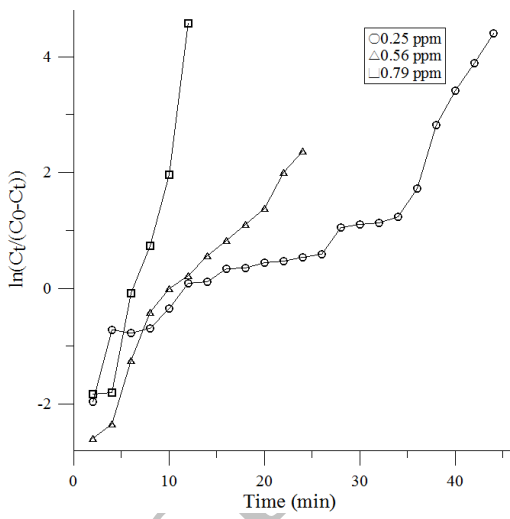


(b)



280

281 (c)



282

283 **Fig. 11.** Effect of concentration on $\ln[(C_i - C_b)/C_b]$ (a) 0.25 m s^{-1} , (b) 0.5 m s^{-1} , (c) 0.75 m s^{-1}

284 **Table 4.** The theoretical parameters $k' t$, and K

Face velocity (m s^{-1})	Concentration (ppm)	$k' t$ (min^{-1})	t(min)	K	R^2
0.25	0.25	0.081	40	3.232	0.981
	0.56	0.124	22	2.737	0.978
	0.79	0.143	16	2.294	0.972
0.5	0.25	0.056	32	1.792	0.973
	0.56	0.183	17	3.114	0.972
	0.79	0.201	13	2.613	0.972

0.75	0.25	0.086	22	1.892	0.973
	0.56	0.197	12	2.359	0.979
	0.79	0.565	6	3.389	0.972

285 3.3. Adsorption kinetic models

286 To determine the sorption mechanism, three common kinetic models, Pseudo-first-order,
 287 Pseudo-second-order, and intra-particle diffusion models were applied to empirically analyze
 288 kinetic sorption data.

289 3.3.1. Pseudo-first-order model

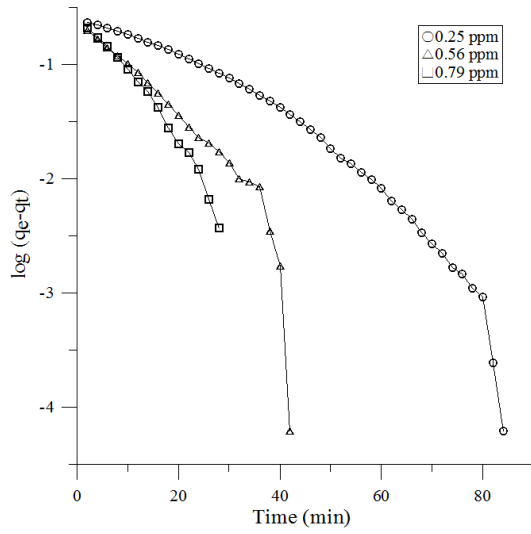
290 The plot of Pseudo-first order kinetics is shown in **Fig. 12**. **Table 4** summarized the calculated
 291 constants according to the Pseudo-first-order equation along with correlation coefficients (r_l^2). The
 292 adsorption amount $q_{e,1}$ of formaldehyde increased from 0.264, 0.324, and 0.472 to 0.298, 0.502, and
 293 0.601 mg g⁻¹ for 0.25 to 0.79 ppmv formaldehyde initial concentrations at 0.25 to 0.75 m/sec face
 294 velocity, respectively. It shows formaldehyde removal efficiency is dependent on initial
 295 concentration. In addition, the amount of formaldehyde adsorption decreased from 0.571, 0.787,
 296 and 0.986 to 0.291, 0.441, and 0.820 mg g media⁻¹ for 0.25 to 0.75 m s⁻¹ face velocity at 0.25, 0.56
 297 and 0.79 ppmv formaldehyde initial concentration, respectively. The removal efficiency of
 298 formaldehyde is also dependent on face velocity. Moreover, the calculated data of $q_{e,1}$ are closer to
 299 the experimental data. With the initial concentration increased from 0.25 to 0.79 ppm at face
 300 velocity 0.25 to 0.75 m s⁻¹, the rate constant (k_1) values were increased from 0.77 to 0.145, 0.082 to
 301 0.227 and 0.125 to 0.547 min⁻¹, respectively. The rate constant (k_1) of the pseudo-first-order rate
 302 model is linearly related to formaldehyde initial concentration (C_0) is shown in **Fig. 13**. This result
 303 is similar to the findings of many previous studies (**Saeid and Bahare, 2006**).

304

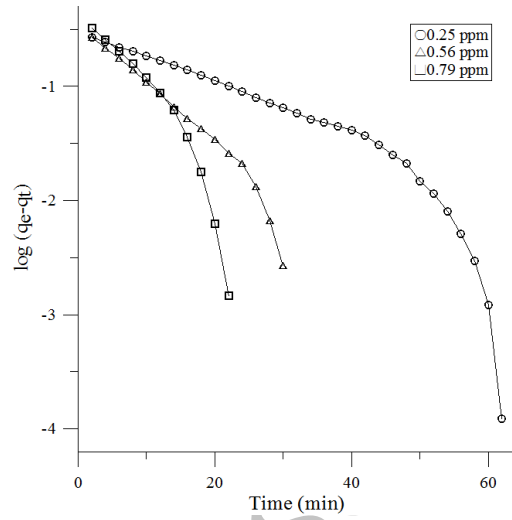
305

306

307 (a)

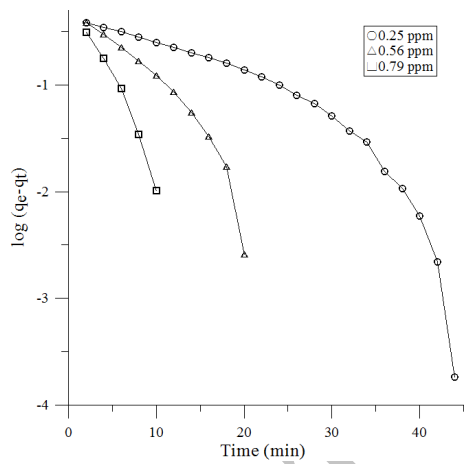


(b)



308

309 (c)



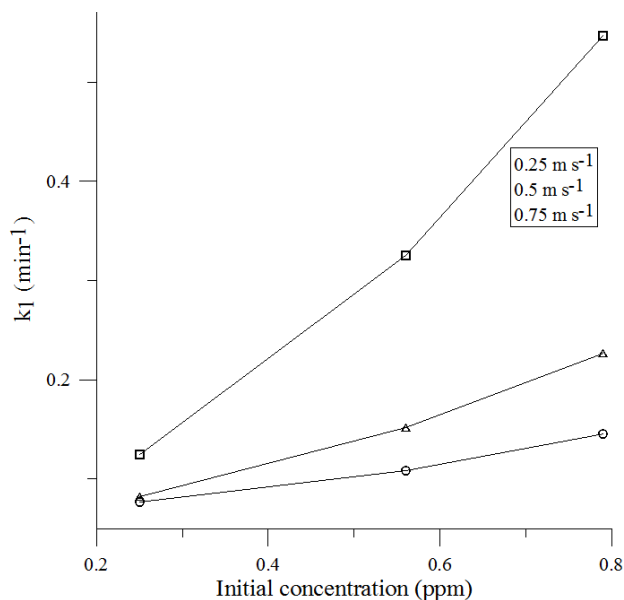
310

311 **Fig. 12.** Plot of Pseudo-first order kinetics at face velocity (a) 0.25 m s^{-1} , (b) 0.5 m s^{-1} , (c) 0.75 m
 312 s^{-1} .

313 **Table 4.** Fitting parameters of three kinetic models for formaldehyde adsorption.

Face velocity	Inlet concentration	Pseudo-first-order				Pseudo-second order				Intraparticle diffusion			
		$k_1 (\text{min}^{-1})$	$q_{e,1} (\text{mg g}^{-1})$	r_1^2	MAPE (%)	$k_2 (\text{g mg}^{-1} \text{min}^{-1})$	$q_{e,2} (\text{mg g}^{-1})$	r_2^2	MAPE (%)	$k_i (\text{mg g}^{-1} \text{min}^{-1/2})$	C	r_i^2	MAPE (%)
0.25 m s^{-1}	0.25 ppm	0.077	0.571	0.915	161.51	0.323	0.309	0.96	5.49	0.031	0.019	0.846	12.43
	0.56 ppm	0.108	0.349	0.969	14.96	0.683	0.329	0.984	2.35	0.037	0.053	0.813	7.89
	0.79 ppm	0.145	0.291	0.977	25.27	0.715	0.35	0.973	4.63	0.049	0.057	0.867	6.4
0.5 s^{-1}	0.25 ppm	0.082	0.787	0.803	88.69	0.4	0.402	0.972	3.92	0.041	0.048	0.895	6.69
	0.56 ppm	0.142	0.548	0.959	20.69	0.689	0.436	0.983	3.67	0.06	0.087	0.832	6.57
	0.79 ppm	0.227	0.441	0.896	119.21	0.716	0.522	0.972	5.46	0.089	0.11	0.871	5.67

	0.25 ppm	0.125	0.986	0.795	156.27	0.409	0.498	0.94	5.94	0.072	0.055	0.924	8.72
0.75 m s ⁻¹	0.56 ppm	0.225	0.825	0.896	64.36	0.705	0.785	0.965	6.12	0.109	0.103	0.876	7.06
	0.79 ppm	0.547	0.82	0.878	66.19	0.721	0.919	0.974	7.92	0.133	0.153	0.843	5.93



314

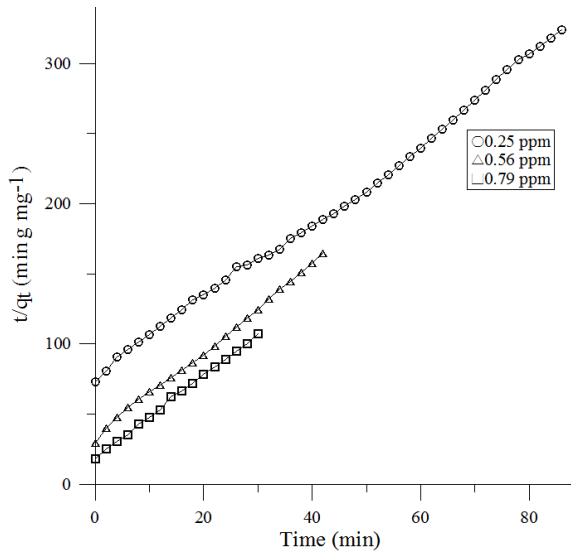
315 **Fig. 13.** The relationship between the Pseudo-first-order rate constant and formaldehyde initial
316 concentration.

317 3.3.2. Pseudo-second-order model

318 **Fig. 13** presents a plot of the Pseudo-second order kinetics for the formaldehyde adsorption
319 onto AC filter media. **Table 4** summarizes the calculated constants (k_2) of the Pseudo-second-order
320 equation along with correlation coefficients (r_2^2). The adsorption amount ($q_{e,2}$) of the formaldehyde
321 increased from 0.309, 0.329, and 0.715 to 0.498, 0.785, and 0.721 mg g⁻¹media⁻¹ for 0.25 to 0.79
322 ppmv formaldehyde initial concentrations at 0.25 to 0.75 m/sec face velocity, respectively. The
323 formaldehyde removal is based on initial concentration. In addition, the amount of formaldehyde
324 adsorption $q_{e,2}$ increased from 0.309, 0.402, and 0.498 to 0.35, 0.522, and 0.919 mg g⁻¹media⁻¹ for
325 0.25 to 0.75 m/sec face velocity at 0.25, 0.56 and 0.79 ppmv formaldehyde initial concentration,
326 respectively. It shows that the formaldehyde removal efficiency is also dependent on face velocity.
327 With initial concentration increased from 0.25 to 0.79 ppm at face velocity 0.25 to and 0.75 m s⁻¹,
328 the rate constant (k_2) values were increased from 0.323, 0.4, and 0.409 to 0.715, 0.716, and 0.721 g

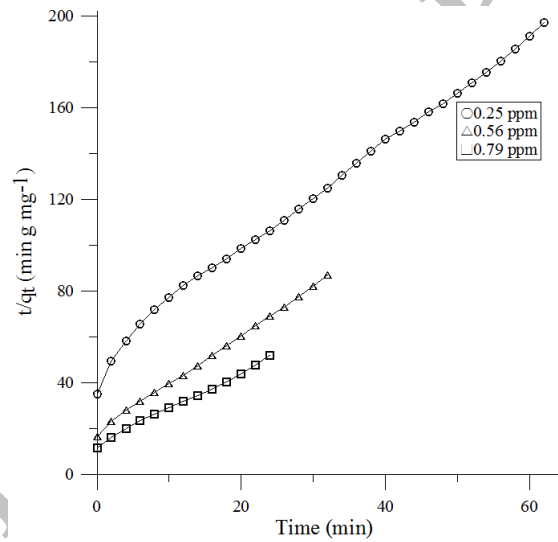
329 $\text{mg}^{-1} \text{min}^{-1}$, respectively. Moreover, the calculated $q_{e,2}$ values are closer to the experimental data, the
 330 rate constant (k_2) of the pseudo-second-order rate model related to formaldehyde initial
 331 concentration (C_0) is shown in **Fig. 14**. This result is similar to the findings of many previous
 332 studies (**Saeid and Bahare, 2006**).

333 (a)

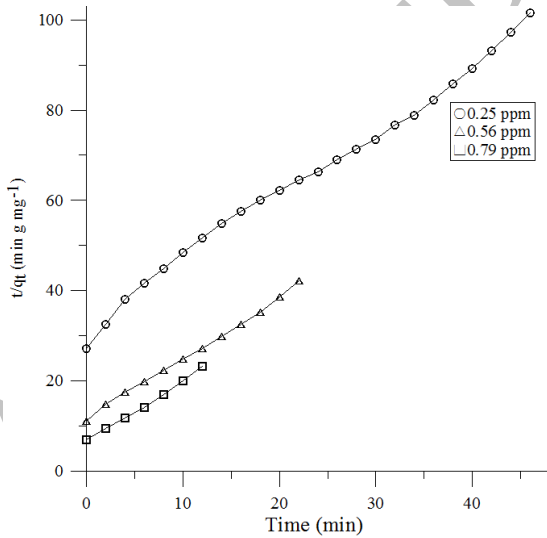


334

(b)

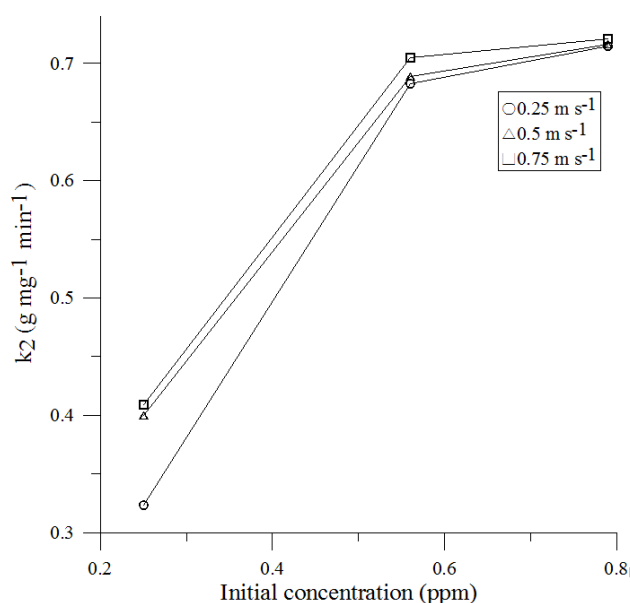


335 (c)



336

337 **Fig. 13.** Plot of Pseudo-second order kinetic for the adsorption of formaldehyde at face velocity (a)
 338 0.25 m s^{-1} , (b) 0.5 m s^{-1} , (c) 0.75 m s^{-1} .



339

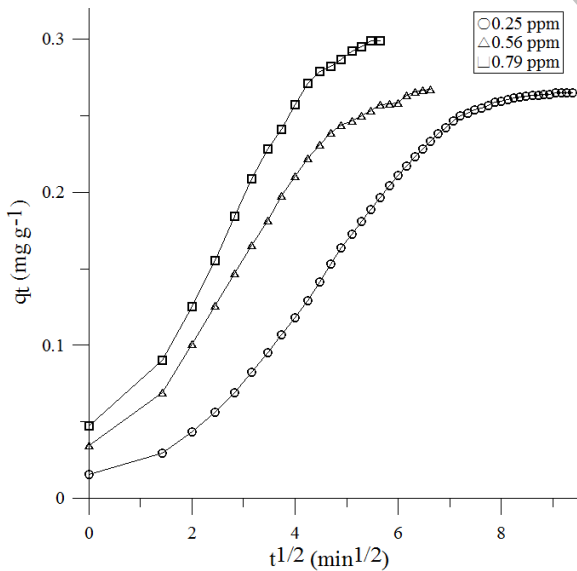
340 **Fig. 14.** The relationship between the Pseudo-second-order rate constant and formaldehyde initial
 341 concentration.

342 3.3.3. Intraparticle diffusion model

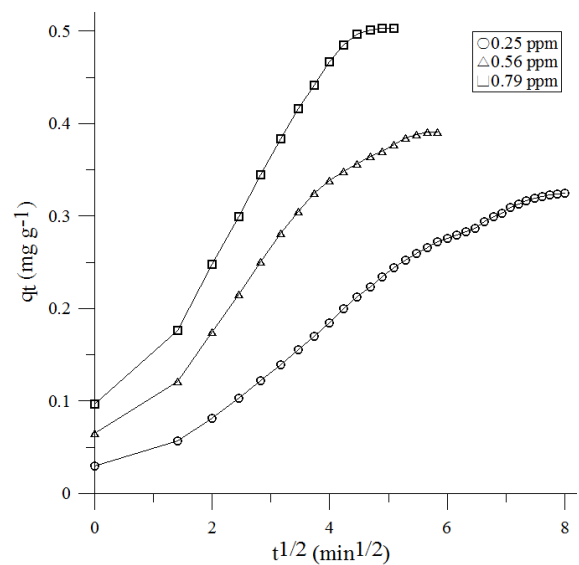
343 **Fig. 15** shows the plot of intraparticle diffusion model. The rate constants (k_i) are obtained
 344 from the slope of the line on second stage. C value of the intercept demonstrated some degree
 345 boundary layer thickness. It is larger to promote the greater effect of the boundary layer. k_i and C
 346 value with its correlation coefficients are shown in **Table 4**. The adsorption process presented three
 347 steps: stage 1: the adsorbate molecules diffused in the adsorption and adsorbed on the outer surface
 348 (Shiue and Hu 2012; Tang et al. 2016; Hu et al., 2017; Shiue et al. 2017). Stage 1 is finished
 349 before 2 min. average at 0.25 to 0.79 ppm inlet formaldehyde concentrations of 0.25, 0.5, and 0.75
 350 m s⁻¹ face velocity, respectively. Stage 2: this stage is a developed process and k_i is the highest,
 351 resulting that the intraparticle diffusion is the key controlling factor of adsorption rate. (Shiue and
 352 Hu 2012; Tang et al. 2016; Hu et al., 2017; Shiue et al. 2017) It kept and continues from 2 min to
 353 25~49, 16~36, and 9~25 mins of AC adsorptive filter media at 0.25 to 0.79 ppm inlet concentration
 354 of 0.25, 0.5, and 0.75 m s⁻¹ face velocities, respectively. Stage 3: this stage is the adsorption
 355 equilibrium process, where the intraparticle diffusion further slows down, and the adsorption rate

356 decreases significantly due to the adsorbates almost filled the micropores and mesopores. (Shiue
 357 and Hu 2012; Tang et al. 2016; Hu et al., 2017; Shiue et al. 2017) It starts after the above
 358 mentioned time schedule at 0.25 to 0.79 ppm inlet formaldehyde concentration of 0.25, 0.5, and
 359 0.75 m s⁻¹ face velocity, respectively. The formaldehyde is slowly removed via intraparticle
 360 diffusion into the particles and is finally kept in the micropores. Generally, intraparticle diffusion
 361 rate constant k_i is the slope of the stage 2 line. The values of k_i arise from 0.031 to 0.049 min^{-1/2}
 362 (0.25m s⁻¹), from 0.041 to 0.089 min^{-1/2} (0.5m s⁻¹), and from 0.072 to 0.133 min^{-1/2}(0.75m s⁻¹) of AC
 363 adsorptive filter media when increase from 0.25ppm to 0.79ppm, respectively. Nevertheless, the
 364 linear intercept C is not zero and larger, showing that the membrane diffusion in stage 3 also
 365 affected the adsorption rate. (Tang et al. 2016) Three stage of the intraparticle diffusion model
 366 fitted the adsorption data well. The formaldehyde adsorption onto AC adsorptive filter media is
 367 affected by combined intraparticle diffusion model and membrane diffusion (Tang et al. 2016).

368 (a)



(b)



369

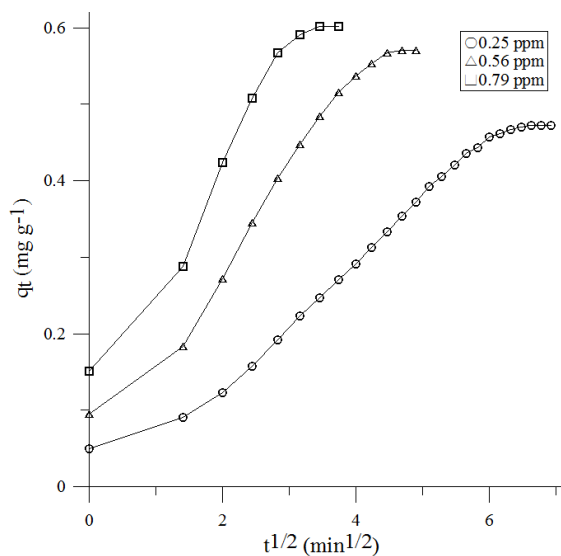
370

371

372

373

374 (c)



375

376 **Fig. 15.** Plot of intraparticle diffusion model at face velocity (a) 0.25 m s^{-1} , (b) 0.5 m s^{-1} , (c) 0.75 m
377 s^{-1} .

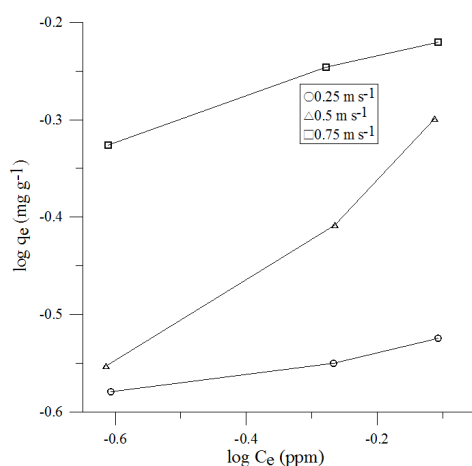
378 3.3.4. Error Analysis

379 The correlation coefficients of the Pseudo-second order kinetic model (Average value = 0.969)
380 is higher than the Pseudo-first order kinetic model (Average value = 0.899) and the intraparticle
381 diffusion model (Average value = 0.863). This result is similar to the findings by Tsai et al. (2017)
382 and Adelodun et al. (2016). For the adsorption process, physisorption might be the controlling
383 mechanism by the highest correlation coefficient (Average value = 0.969) and the lowest mean
384 absolute percentage error value (Average MAPE <8%) verification. Only the intraparticle diffusion
385 model fits the experimental data. Thus, it was determined to be the rate limiting step for the initial
386 period of the adsorption process.

387 3.4. Adsorption isotherms

388 The plots of the Freundlich and Langmuir isotherm models are presented as **Figs. 16** and **17**,
389 respectively. The isotherm constants and maximum adsorption capacity (q_{max}) values are listed in
390 **Table 5**. According to the linear regression method, the formaldehyde adsorption by AC adsorptive
391 filter media exhibits a better fit to Langmuir equation was statistically confirmed by giving

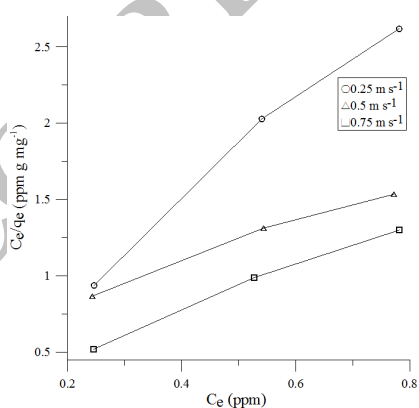
392 greater R^2 values closer to unity ($R^2 > 0.987$). This denotes that the Langmuir model may better
393 describe an adsorption isotherm for AC adsorptive filter media. This adsorptive behavior pointed
394 out that the adsorption occurs on a heterogeneous surface, which may be aspect to the various active
395 sites on AC adsorptive filter media. The overall adsorptive performance is prevailed as a physical
396 adsorption process. This phenomena has been described before (**Bernabe et al., 2015; Cheng and**
397 **Tsai, 2007**). The results show that the maximum formaldehyde adsorption capacity (q_{\max}) of AC
398 adsorptive filter media was 0.7862 mg g^{-1} at 0.75 m s^{-1} face velocity. From the observed results we
399 conclude that AC adsorptive filter media is potentially a good solution for formaldehyde removal.



400

401

Fig. 16. Formaldehyde adsorption with Freundlich adsorption isotherm.



402

403

Fig. 17. Formaldehyde adsorption with Langmuir adsorption isotherm.

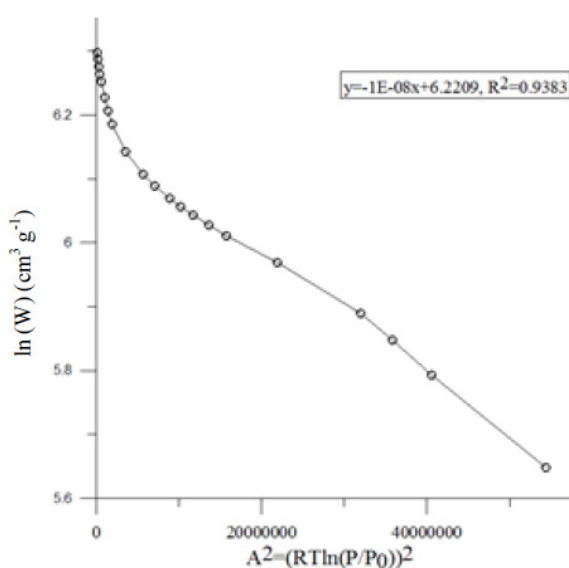
404

405

Table 5. Freundlich and Langmuir isotherm parameters

Face velocity (m s ⁻¹)	Freundlich isotherm			Langmuir isotherm		
	K _f (mg g ⁻¹)(L mg ⁻¹) ^{1/n}	1/n	R ²	q _{max} (mg g ⁻¹)	K _L (L mg ⁻¹)	R ²
0.25	0.5965	0.1061	0.9738	0.3167	15.2046	0.9872
0.5	0.6121	0.2133	0.9794	0.6864	8.058	0.9879
0.75	0.8242	0.4908	0.9886	0.7862	2.2	0.9923

407 Except than the Freundlich and Langmuir isotherms, the experimental data were also
 408 represented by the D–R equation. **Fig. 18** was plotted as $\ln W$ versus A^2 for AC filter from
 409 adsorption isotherms of N₂ data of **Fig. 3**. As shown in the figure, the characteristic curve for
 410 formaldehyde is simulated linearly at a lower A^2 range. Nevertheless, the isotherm data of
 411 formaldehyde cannot be linearly fitted over the entire range of abscissa. A clear transition was
 412 observed at $\sim 2.2 \times 10^7 \text{ J}^2 \text{ mol}^{-2}$ of A^2 , which is equal to a relative pressure (P/P_0) of 0.151 for the
 413 adsorbates to represent a good fit at lower levels of A^2 (high P/P_0) and of a big discrepancy at high
 414 A^2 (low P/P_0) in agreement with Shiue et al. (2011) research work. The results show that the D–R
 415 equation can simulate the experimental adsorption data except very low relative pressures. In the
 416 event of relative pressures lower than 0.151, the D–R equation may be difficult to utilize the
 417 simulation of the adsorption capacity for the tested adsorption systems.



418

419

Fig. 18. The D-R isotherm

420 4. Conclusions

421 This study concludes that formaldehyde removal by AC adsorptive filter media is effective in
422 the indoor environment. From SEM images it was found the micrographs show that AC surface is
423 smooth and distributed with small pores. The surface of AC is rather uneven, coarse and porous.
424 Irregular layer structure were formed the amorphous that the XRD diffraction peaks were broad. In
425 EDS analysis C, O, Mg, P etc. were detected, which showed that the Mg/C ratio=0.68% and further
426 proves the existence of MgO in AC. Therefore, the surface area is $1333.3304 \text{ m}^2 \text{ g}^{-1}$ according to
427 BET analysis by checking with FTIR, ketone -C=O bonds which is explained at sensible peak of
428 3417 cm^{-1} and weak strength bonding at 2329 cm^{-1} were successfully grafted on carbon.

429 The results presented that increased initial concentration of formaldehyde and face velocity
430 were associated with a higher adsorption capacity and faster breakthrough time. The breakthrough
431 behavior at low formaldehyde concentration can be linearly expressed by the relationship between
432 the natural logarithm of breakthrough time and the natural logarithm of initial concentration. It is
433 proved that the agreement between the Yoon and Nelson model and the experimental data is high
434 due to high correlation coefficients ($R^2 > 0.972$) for formaldehyde adsorption onto coconut activated
435 carbon filter media. For adsorption kinetics of formaldehyde removal by AC adsorptive filter media,
436 the Pseudo-second-order model with highest correlation coefficient ($0.94 < R^2 < 0.984$) and lowest
437 MAPE values (8 %) is more suitable than Pseudo-first-order-model, and intraparticle diffusion
438 model and membrane diffusion could predict the adsorption kinetics. The Langmuir equations is
439 well-fitted to AC adsorptive filter. The D-R equation successfully predicted the equilibrium
440 capacity of formaldehyde onto AC at high relative pressures. The overestimation of adsorption
441 capacity was detected when seeking to expect the formaldehyde adsorption capacity at relative
442 pressures 0.151.

443

444 **Acknowledgements**

445 The authors would like to acknowledge the supports from the Ministry of Science and
446 Technology with contract number 105-3011-F-027-001.

447 **Nomenclature**

448 C intercept of intraparticle diffusion model, mg kg^{-1}

449 C_e concentration of free formaldehyde in air, ppm

450 C_0 the inlet concentration, ppm

451 k_1 the pseudo-first-order rate coefficient, min^{-1}

452 k_2 the pseudo-second-order rate coefficient, $\text{g mg}^{-1} \text{min}^{-1}$

453 k_i the intraparticle diffusion rate constant, $\text{mg g}^{-1} \text{min}^{-1/2}$

454 K_L the Langmuir's adsorption equilibrium constant related to the affinity of the binding sites, L
455 mg^{-1}

456 q_e the amount of adsorbed formaldehyde per unit weight of adsorbent, mg g^{-1}

457 q_{max} maximum adsorption capacity, mg g^{-1}

458 q_t the amount of adsorbate adsorbed at time t , mg g^{-1}

459 t testing time, min

460 V the airflow rate, L min^{-1}

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