



## Adsorption of Low Concentration Formaldehyde in Air Using Ethylene-Diamine-Modified Diatomaceous Earth

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### ABSTRACT

Formaldehyde is one of the most common difficult-to-eliminate indoor air pollutants. Among the available methods of eliminating formaldehyde, adsorption is still the most commonly used due to its simplicity, efficiency, and economic viability. This study investigated the potential of diatomaceous earth (DE) adsorbent for formaldehyde (low concentration in air). DE was considered because of its high silica content and high porosity. It also examined the effect of adding ethylene-diamine (EDA) on the adsorption performance of DE. Amine groups have been proven to improve the adsorption of formaldehyde through their reaction that produces imine. The amount of added EDA was varied from 0.25 to 0.75 g per gram of DE. For pure DE adsorbent, the adsorption performance was 298 mg/g. Adding 0.75 g of EDA resulted in maximum DE adsorption performance (565 mg/g). EDA-modified DE was shown to be a potential adsorbent for removing formaldehyde in air.

**Keywords:** Calibration curve; Breakthrough point; Material synthesis; Amine; Indoor air pollution.

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### INTRODUCTION

Formaldehyde (HCHO) can come from a great number of sources like household products and materials commonly used for constructions and structural designs. It is also emitted by materials utilized for pipe, wall and thermal insulation, paint, furniture, and other wood products (Han *et al.*, 2012; Rezaee *et al.*, 2013). Thus, it is considered the most common aldehyde in the environment; and the probability of exposure to such potential hazard is very high (Panagopoulos *et al.*, 2011). Based on the standards and guidelines for indoor air quality, the 30-minute exposure threshold for formaldehyde can be as low as 0.1 mg/m<sup>3</sup> (WHO, 2010). This limit is lower compared to other air pollutants. Effects of going beyond the threshold limit vary from a simple sensory irritation, to a more serious nasal mucosa cytotoxic damage, to a critical upper respiratory tract cancer (Arts *et al.*, 2008; Gorbunov *et al.*, 2013).

This is very alarming, since various studies show that the

levels of indoor formaldehyde concentrations are much higher than the threshold value reported by the World Health Organization (WHO). In China, the mean concentration of formaldehyde in refurbished dwellings in urban areas is 0.238 mg/m<sup>3</sup>; while in office buildings, it is as high as 0.256 mg/m<sup>3</sup>. Similarly, the formaldehyde levels in the United Kingdom and Austria have reached concentrations of 0.171 and 0.115 mg/m<sup>3</sup>, respectively (WHO, 2010).

Thus, various techniques have been used to remove formaldehyde in air. These include phytoremediation, photocatalysis, biological degradation, and adsorption. Phytoremediation has a promising advantage of being less costly as compared to other specialized technological approaches; but the formaldehyde removal time is indefinite and can be long. Photocatalysis can easily degrade formaldehyde; but is more costly because of the utilization of high energy ultraviolet radiation. Biological degradation has longer service life because of the continuous metabolism of the microbes involved; but this may cause biohazard risks when microorganisms leak from the system or when pollutive by-products are formed.

For these reasons, and for its simplicity, efficiency, and economic viability, the most common method used to remove formaldehyde in air is adsorption. Adsorption is a process where organic molecules are held on the surface and in the

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pores of the adsorbent through weak forces of interactions. The efficiency of the said process is greatly determined by the characteristics of the adsorbent (Rezaee *et al.*, 2013).

Several studies used activated carbon (AC) as adsorbent (Ie *et al.*, 2014; Jo and Chun, 2014) because of its strong adsorption capacity, ease of separation, and regeneration capability. However, since the surface of activated carbon is only slightly polar, it does not efficiently remove polar organic molecules (Tsai *et al.*, 2004) like formaldehyde (Rezaee *et al.*, 2013). Other studies explored various adsorbents like bone char (Rezaee *et al.*, 2013), alumina (Agarwal *et al.*, 2011), and silica (Saeung and Boonamnuayvitaya, 2008; Kim *et al.*, 2011). Some of these studies tried to increase the adsorption capacity by improving interaction with formaldehyde through the introduction of amine groups to the surface of the adsorbent (Srisuda and Virote, 2007; Yu *et al.*, 2012; Lee *et al.*, 2013). In a study by Kim *et al.* (2011), various adsorbents like mesoporous material (MCM-41), crystalline microporous zeolite (HY) and amorphous silica (XPO-2412) were functionalized with three kinds of amine groups namely 3-aminopropyltriethoxysilane (APTES), N(b-aminoethyl) g-aminopropylmethyl dimethoxysilane (AEAPMDMS), and N1-(3-(trimethoxysilyl)-propyl) diethylenetriamine (TMSPDETA).

This study investigated the potential of commercially available diatomaceous earth (DE) as an adsorbent for the removal of low concentration formaldehyde in air. DE was considered because of its high silica content and high porosity, which are properties of good adsorbent (Tsai *et al.*, 2004). The experiment of Virote *et al.* (2005) proved that the adsorption capacity of mesoporous silica is around three times higher than that of AC. Diatomaceous, diatomite, or DE is a naturally occurring, soft sedimentary rock that consists of 86% to 94% silicon dioxide ( $\text{SiO}_2$ ) with significant amount of alumina ( $\text{Al}_2\text{O}_3$ ) and ferric oxide ( $\text{Fe}_2\text{O}_3$ ) (Tsai *et al.*, 2004).

This study further examined the effect of adding ethylenediamine (EDA) on the adsorption performance of DE. Numerous studies proved that the presence of amine group improved the adsorption of formaldehyde by mesoporous materials through their reaction that produces imine (Srisuda and Virote, 2007; Kim *et al.*, 2011). The adsorption

equilibrium was also determined using the Langmuir and Freundlich isotherms.

The study is limited to the synthesis and characterization of EDA-modified DE adsorbent, and the analysis of the effect of the various loading amount of EDA on formaldehyde adsorption.

## METHODS

### Adsorbent Modification

Diatomaceous earth (DE) was modified using three different loading amounts of EDA: 0.25 g, 0.50 g and 0.75 g per 1.0 g of DE. Anhydrous EDA of varying amounts were first mixed with 336 mL of ethanol (95%, SCRC); allowed to stand for 30 minutes; and then mixed with 6 g of DE in a round bottom flask. The samples ( $\text{DE}_{0.25}$ ,  $\text{DE}_{0.50}$ , and  $\text{DE}_{0.75}$ ) were refluxed for 2 hours at  $78^\circ\text{C}$  to thermally accelerate the reaction between EDA and DE. Finally, the samples were placed in a petri dish and oven dried for 8–12 hours at a temperature of  $60^\circ\text{C}$ , after removing the alcohol using a rotary evaporator (ELEYA 1-1000) for about 30–40 minutes at  $78^\circ\text{C}$  (Zhaoming, 1997). The surface properties of the modified adsorbents were analyzed using SEM (Hitachi S4500); and the functional groups on their surfaces were determined using FTIR spectrophotometer (Tensor 27).

These modified adsorbents, along with unmodified DE, were then examined as formaldehyde adsorbents using the set-up in Fig. 1.

### Adsorption Set-up

The system consists of an adsorbent tube, a cartridge sampler, a nitrogen ( $\text{N}_2$ ) tank, three flow meters, a mixing chamber, a pump, and a formaldehyde solution in a flask submerged in water bath.

The adsorbent tube was prepared by placing 0.10 g of adsorbent in a polytetrafluoroethylene (PTFE) tube (ID: 7 mm, L: 8 cm); securing both sides of the tube with fiber glass; and closing both ends with a syringe filter (0.25 mm,  $0.2\ \mu\text{m}$  PVDF membrane). The cartridge sampler, shown in Fig. 2, is a 2,4-dinitrophenylhydrazine (DNPH)-coated silica gel cartridge sampler (SKC 226-119) (Ho *et al.*, 2013). The mixing chamber was covered with aluminum foil to prevent

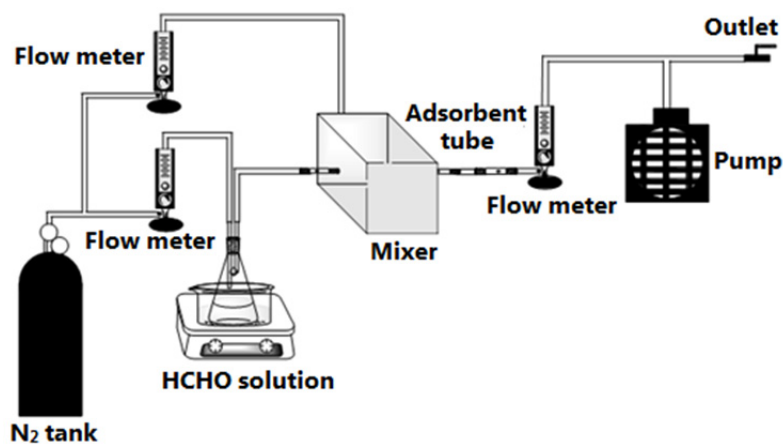


Fig. 1. Formaldehyde (HCHO) adsorption set-up.

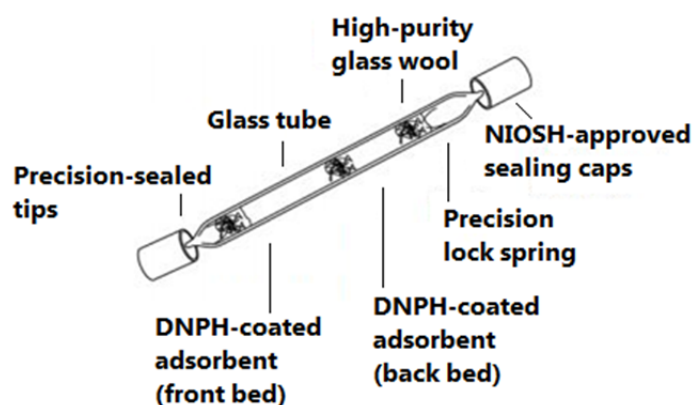


Fig. 2. 2,4-Dinitrophenylhydrazine (DNPH)-coated silica gel cartridge sampler (SKC 226-119).

evaporation of the sample. Each flow meter was calibrated using DC-2 dry test gas meter. Connectors used were PTFE tubes because this type of polymer is temperature and chemical resistant, has non-stick properties, and has low coefficient of friction.

#### Steady-State Analysis

To determine the steady-state condition for sampling, the system was operated at a constant temperature of 25°C, without the adsorbent tube and cartridge sampler. The outlet was closed for 20 minutes while allowing the two (2) gas streams to flow into the mixer: pure nitrogen gas (1000 mL/min); and carrier nitrogen gas (different flow rates) to purge the formaldehyde vapor. The outlet was then opened and the mixture of formaldehyde and nitrogen was pumped out of the mixer. The system was allowed to run until the concentration of formaldehyde coming out from the mixer remains constant (steady state). The concentration was checked every 10 minutes through a formaldehyde meter (Formaldemeter htV) starting from the point the vapor was released from the system. The concentration of formaldehyde was varied by adjusting the amount of carrier nitrogen gas. Then a plot of flow rate versus concentration was generated.

#### Preparation of the Calibration Curve

After allowing the system to stabilize, a blank was run by attaching one end of the cartridge sampler (SKC 226-119) to the mixer while the outlet end was connected to the sampling pump with flexible tubing. The mixture of formaldehyde and nitrogen was allowed to flow in the sampler for 10 minutes at a rate of 1 mL/min (NIOSH 2016). After sampling, the cartridge was capped and placed in the refrigerator at 4°C for 8–12 hours to allow further reaction between DNPH and formaldehyde. This sample served as the blank.

For the samples, the same procedure was followed. However; the adsorbent tube was already incorporated in the system: one end of the adsorbent tube was directly connected to the mixer while the other end was linked to the cartridge sampler.

Each sorbent section of the DNPH-coated silica gel cartridge was removed from the glass with the aid of a specialized cutter and was prepared for HPLC analysis in order to prepare a standard calibration curve prior to the main

experiment. To generate a standard curve, the formaldehyde meter (Formaldemeter htV) reading was plotted against the absorbance reading obtained from the HPLC analysis.

#### Determination of Adsorption Capacity

The adsorption capacities for formaldehyde vapor of DE and EDA-modified DE are tested using five concentrations of formaldehyde vapor. To illustrate the equilibrium characteristics of the adsorption process, the data obtained were fitted into two models, the Langmuir and Freundlich isotherms. Eqs. (1) and (2)

$$\frac{1}{q_e} = \frac{1}{K_L q_{max}} \times \frac{1}{C_e} + \frac{1}{q_{max}} \quad (1)$$

where  $q_e$  is the equilibrium amount of adsorbed gas on the adsorbent (mg/g),  $C_e$  is the concentration of the adsorbate at equilibrium condition (ppmv),  $q_{max}$  is the maximum amount of gas adsorbed on the adsorbent (mg/g) and  $K_L$  is the adsorption equilibrium constant (L/mg).

$$q_e = K_F C_e^n \quad (2)$$

where  $K_F$  and  $n$  are Freundlich constants, which indicate the adsorption capacity and adsorption intensity, respectively. The magnitude of the adsorption intensity,  $n$ , shows the suitability of the system. If the value of  $n$  is greater than one, then the system has favorable adsorption conditions (Srisuda and Virote, 2008).

#### Breakthrough Point Determination

The breakthrough measurements were made with a constant contact time of 60 minutes. The DNPH-coated silica gel cartridge was replaced every 10 minutes for 30 minutes then every five (5) min for 10 minutes. The breakthrough curve for every sample was generated by plotting time against the equilibrium concentration.

## RESULTS AND DISCUSSION

#### Adsorbent Characteristics

The FTIR spectra of the adsorbents are shown in Fig. 3.

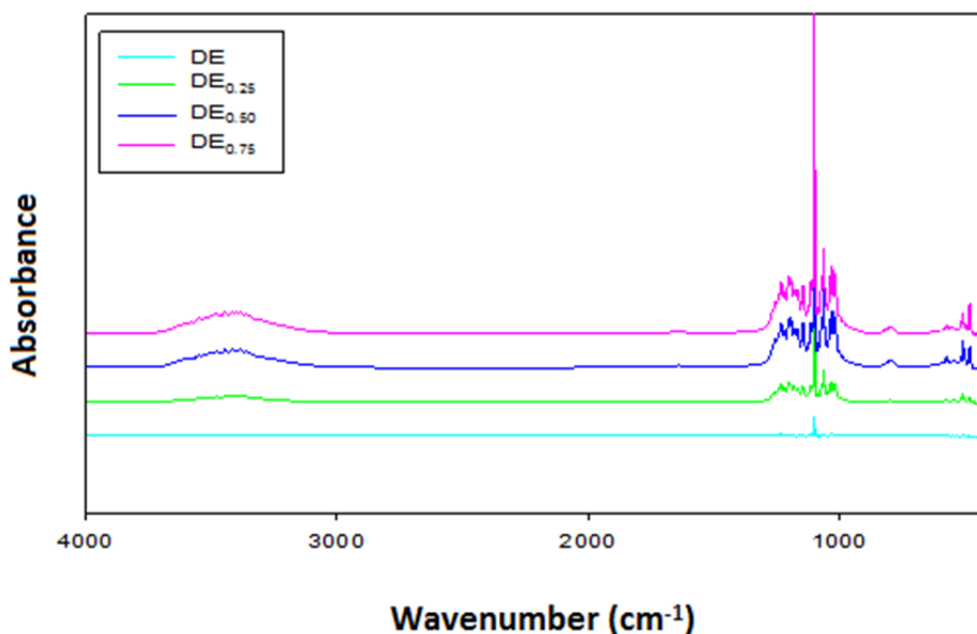


Fig. 3. FTIR spectra for DE, DE<sub>0.25</sub>, DE<sub>0.50</sub> and DE<sub>0.75</sub>.

All samples, DE, DE<sub>0.25</sub>, DE<sub>0.5</sub>, and DE<sub>0.75</sub>, had strong and sharp peaks at around 1090 to 1010 cm<sup>-1</sup> and 625 to 480 cm<sup>-1</sup> due to the asymmetric stretching vibrations of the siloxane band (Si-O-Si) band. Small and broad peaks at 990 to 945 cm<sup>-1</sup> indicated the presence of Si-O bond stretching of the silanol group. These prominent peaks due to the Si-O-Si and Si-O linkages proved that the adsorbent, DE, is composed mostly of silica materials. On the other hand, the peak around 1145 to 1130 cm<sup>-1</sup> which can only be attributed to the modified samples, DE<sub>0.25</sub>, DE<sub>0.5</sub>, and DE<sub>0.75</sub>, indicated the N-H deformation peak. This proved that DE was successfully modified with EDA (Srisuda and Virote, 2008).

The surface structures of the adsorbents were also analyzed using scanning electron microscopy (SEM) at 7.0 kV with 4000x–5000x magnification, as seen in Fig. 4. The micrographs show that the modification did not alter the structure of DE, but instead created numerous macro pores that are desirable for effective adsorption.

#### Steady-State Analysis

The system used was allowed to attain steady state condition using different flow rates of carrier N<sub>2</sub> gas passing thru the 37% w/w formaldehyde solution. Fig. 5 shows that even at different flow rates, the system had similar trends and started to stabilize at the 90-minute mark. This suggests that sampling for formaldehyde adsorption can only be done after 90 minutes of stabilization. A linear relationship between carrier N<sub>2</sub> gas flow rates and formaldehyde concentrations at 90-min mark is shown in Fig. 6. The equation is

$$y = 1.0251x - 0.2435 \quad (3)$$

where  $y$  is the formaldehyde concentration and  $x$  is the flow rate of carrier N<sub>2</sub> gas. Correlation coefficient is 0.9808. From this relationship, the desired formaldehyde concentration

can be obtained by computing the reading at which the flow meter should be adjusted.

#### Calibration Curve

A sample chromatogram which displays the peak of the compound of interest and the time by which the compound was eluted is presented in Fig. 7. The chromatogram presents the separation between 2,4-Dinitrophenylhydrazine (DNPH) and formaldehyde wherein the first peak represents the DNPH while the second shows the formaldehyde peak. Using a 70:30 acetonitrile-water mobile phase, both DNPH and formaldehyde were eluted before the five-minute mark. This is confirmed by the EPA IP-6A Method, which states that formaldehyde peak can be seen approximately at the 4.08-minute mark while that of the DNPH is around the 2.34-minute mark.

The calibration curves were obtained from the linear regression of the peak area of formaldehyde versus concentrations. For the concentration from zero (0) to one (1) ppm, the regression equation obtained was

$$y = 1 \times 10^6 x - 1949.3 \quad (4)$$

where  $y$  is the peak area and  $x$  is the formaldehyde concentration in ppm. The correlation coefficient (0.9952) showed an excellent linearity at a range of 0–1 ppm (Fig. 8).

Similarly, the linear regression equation for the concentration range of one (1) to four (4) ppm (Fig. 9) also proved a good linearity since the correlation coefficient reached 0.9953.

$$y = 732979x + 286852. \quad (5)$$

where  $y$  is the peak area and  $x$  is the formaldehyde concentration in ppm. The formaldehyde concentrations of the samples were determined from the regression equations.

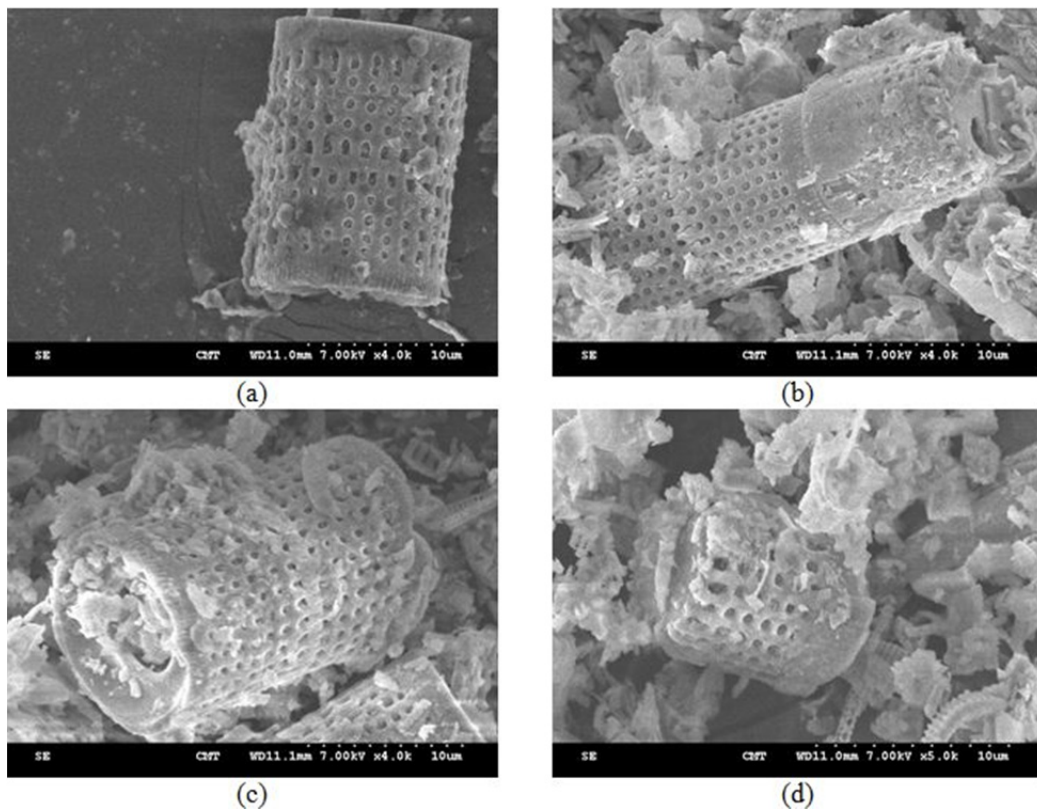


Fig. 4. SEM micrographs of (a) DE (b) DE<sub>0.25</sub> (c) DE<sub>0.50</sub> (d) DE<sub>0.75</sub>.

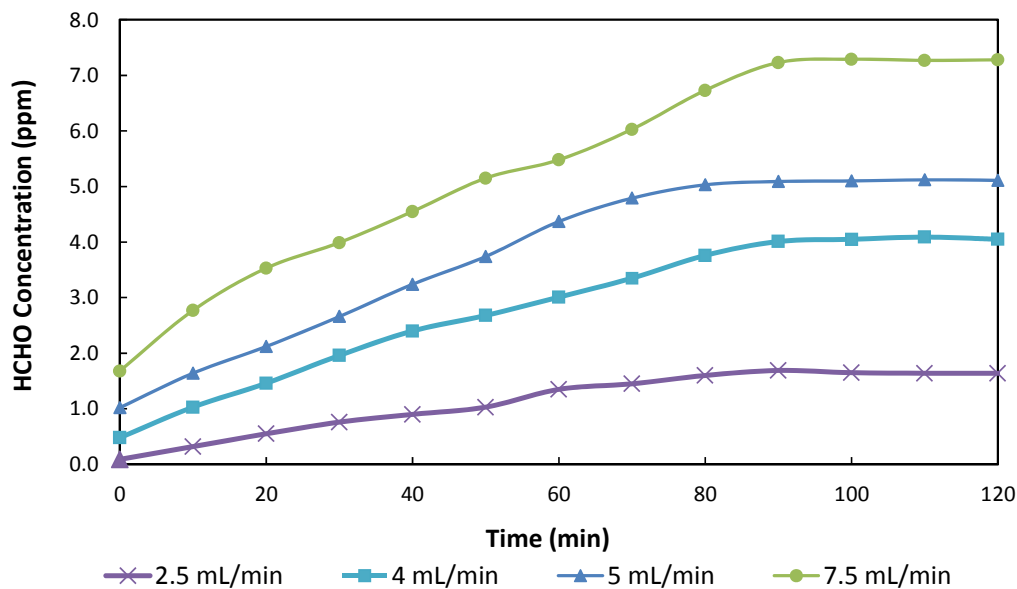


Fig. 5. Stability curve generated after 2 hours of continuous pumping.

#### Adsorption Experiment

The adsorption capacities for formaldehyde vapor of DE and EDA-modified DE were examined at equilibrium condition using five concentrations of formaldehyde vapor. Moreover, two isotherms, Langmuir and Freundlich, were used to explain the equilibrium characteristics. The Langmuir isotherm takes monolayer coverage of an adsorbate over a homogeneous adsorbent surface. On the other hand, the

Freundlich isotherm assumes a heterogeneous surface with a possibility of multilayer adsorption (Vukovic *et al.*, 2010). The plots of the two isotherm models are shown in Figs. 10 and 11.

Tables 1 and 2 summarize the Langmuir and Freundlich isotherms for the adsorption of formaldehyde using DE and EDA-modified DE (DE<sub>0.25</sub>, DE<sub>0.5</sub> and DE<sub>0.75</sub>). For the Langmuir Isotherms, the  $q_{max}$  is the maximum adsorption

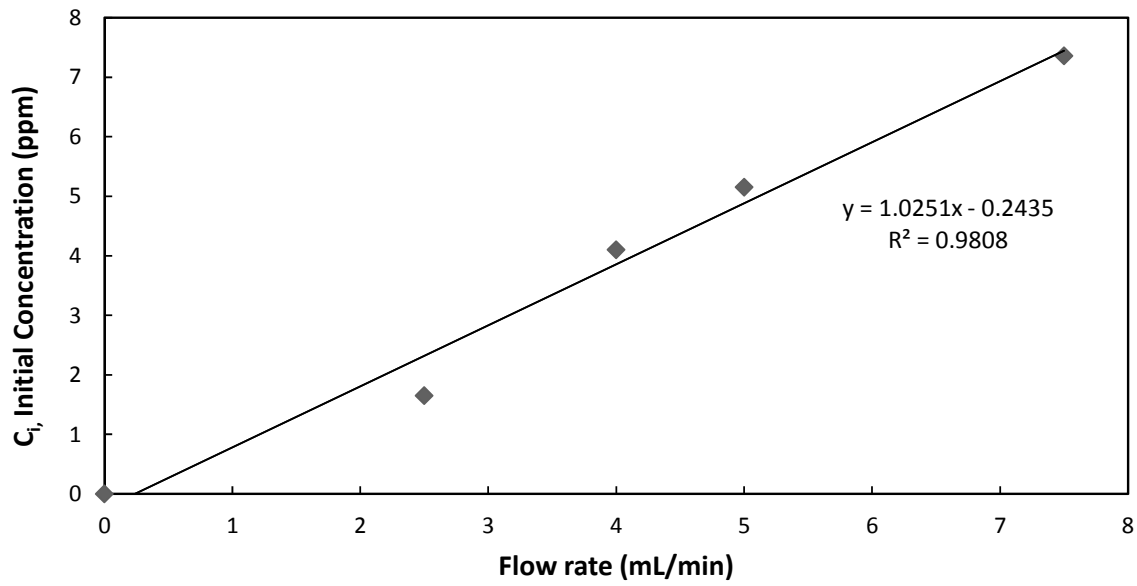


Fig. 6. Plot of nitrogen gas flow rate against formaldehyde vapor concentration.

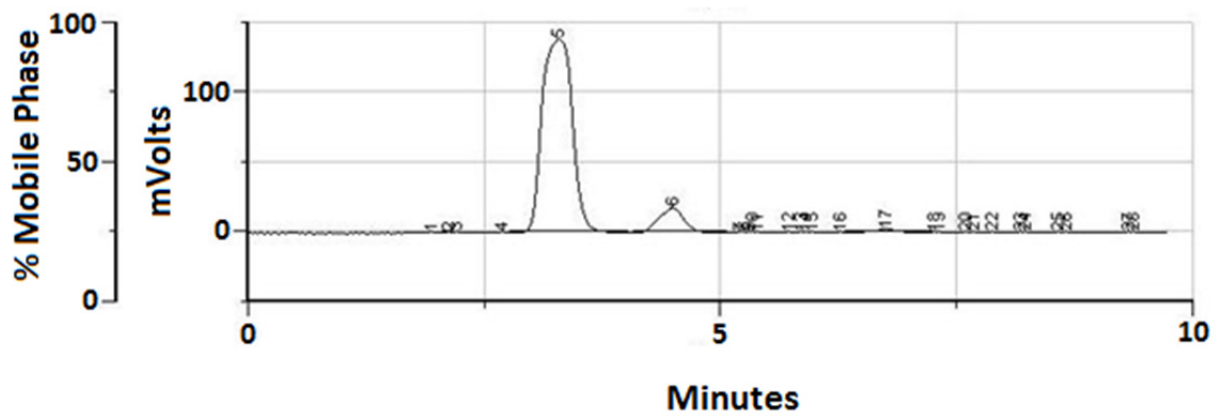


Fig. 7. Sample chromatogram which illustrates DNPH and HCHO peaks.

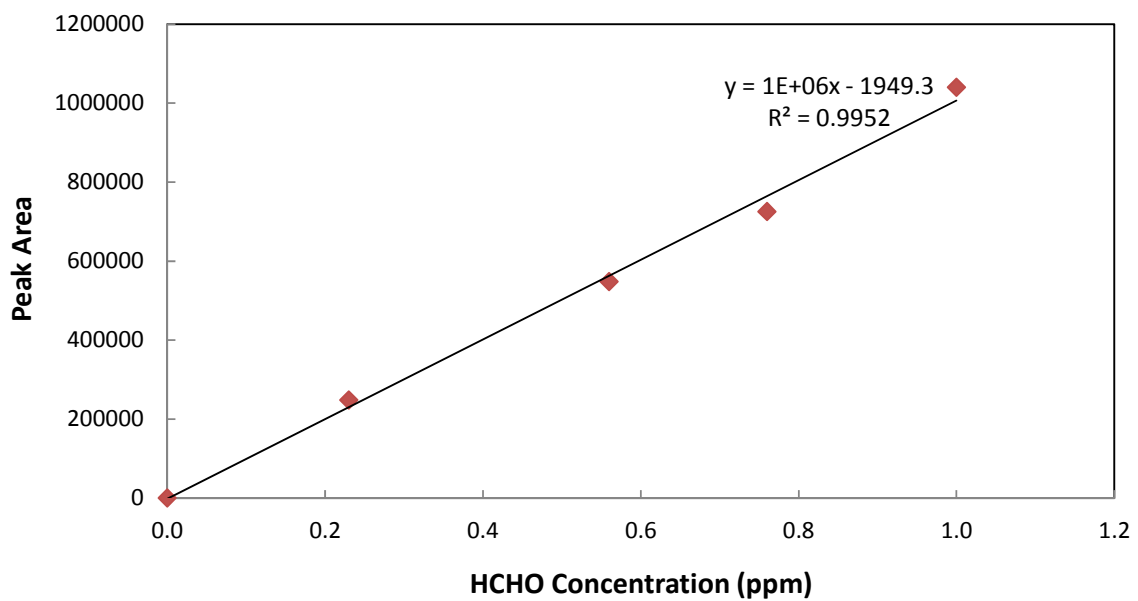


Fig. 8. Calibration curve from 0 to 1 ppm.

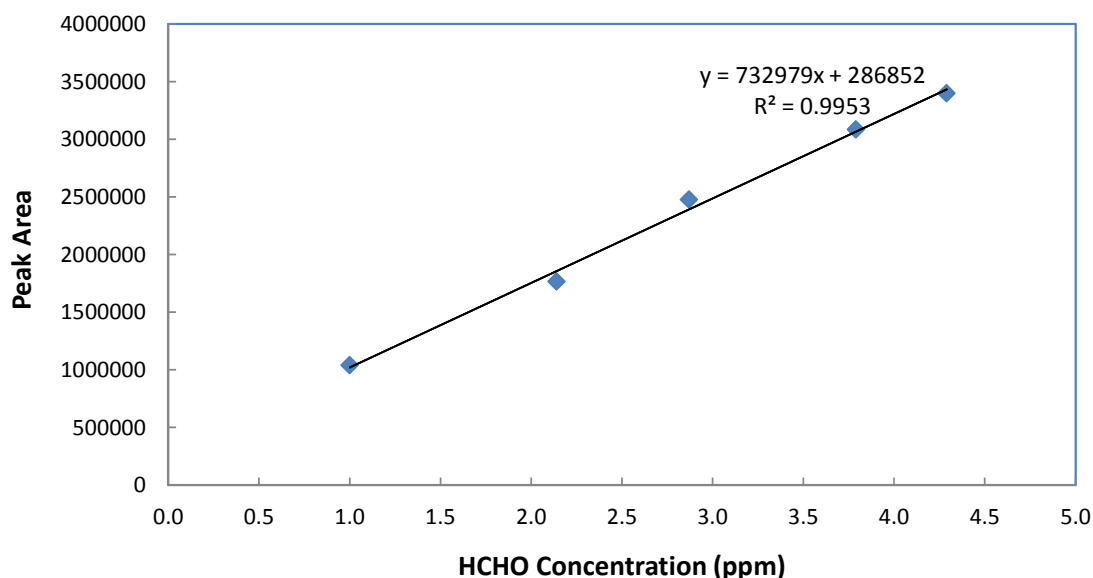


Fig. 9. Calibration curve from 1 to 4 ppm.

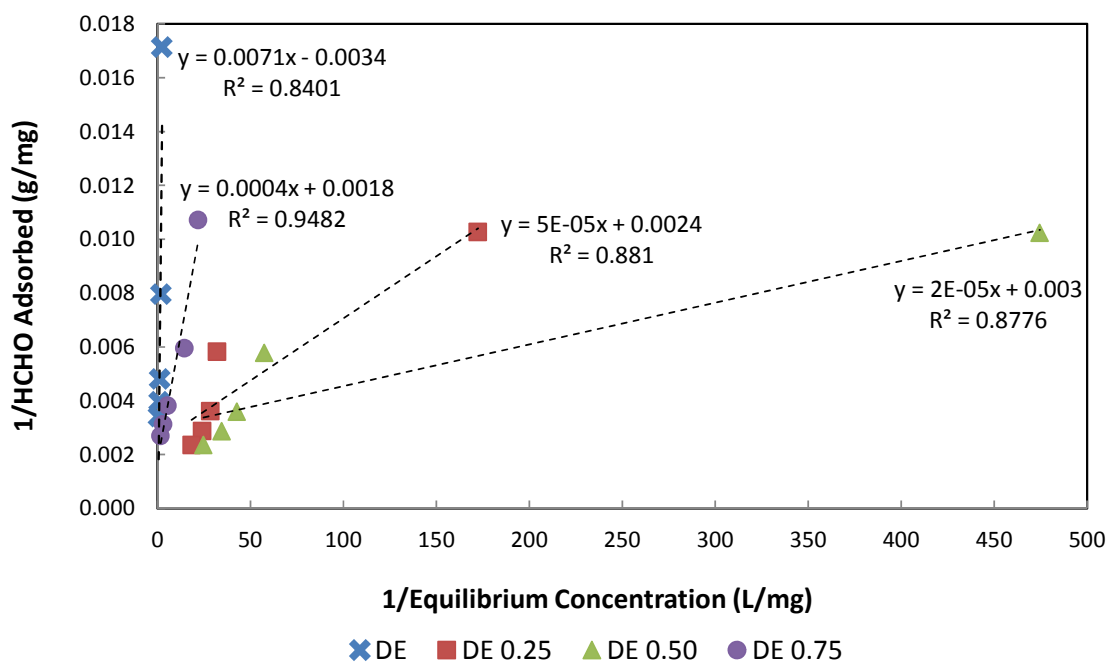


Fig. 10. Langmuir adsorption isotherm trend lines for DE and EDA-modified DEs.

capacity or the maximum amount of formaldehyde adsorbed at monolayer coverage and the value of  $K_L$  is the constant which represents the adsorption capacity at the equilibrium concentration. For the Freundlich isotherm, the  $n$  value shows the degree of dependence of the adsorption on the equilibrium concentration and  $K_F$  is similar to that of its counterpart in Langmuir,  $K_L$  (Vukovic *et al.*, 2010).

As seen from the tables, the correlation coefficients ( $R^2$ ) for the Langmuir model for DE<sub>0.25</sub> and DE<sub>0.75</sub> are higher than that of the Freundlich model while the values are almost the same for DE<sub>0.50</sub>. This data suggests that the Langmuir isotherm best describes the adsorption of formaldehyde on the modified adsorbents. On the contrary, for the adsorption

using DE, Freundlich isotherm predominates.

Aside from the monolayer coverage, the fit in the Langmuir isotherm was also indicative of chemisorptions, which may have resulted from the reaction between the amine groups and formaldehyde producing imine. In a study conducted by Srisuda and Virote (2008) where formaldehyde is adsorbed on amine functionalized mesoporous silica, the FTIR spectrum showed changes in peaks. After adsorption, the  $\text{NH}_2$  and  $\text{NH}$  bands, which were previously present, disappeared; and new imine peaks appeared. A similar mechanism in the study by Le *et al.* (2010) reacted amine groups with formaldehyde producing imine group trapped in a form of an imine compound.

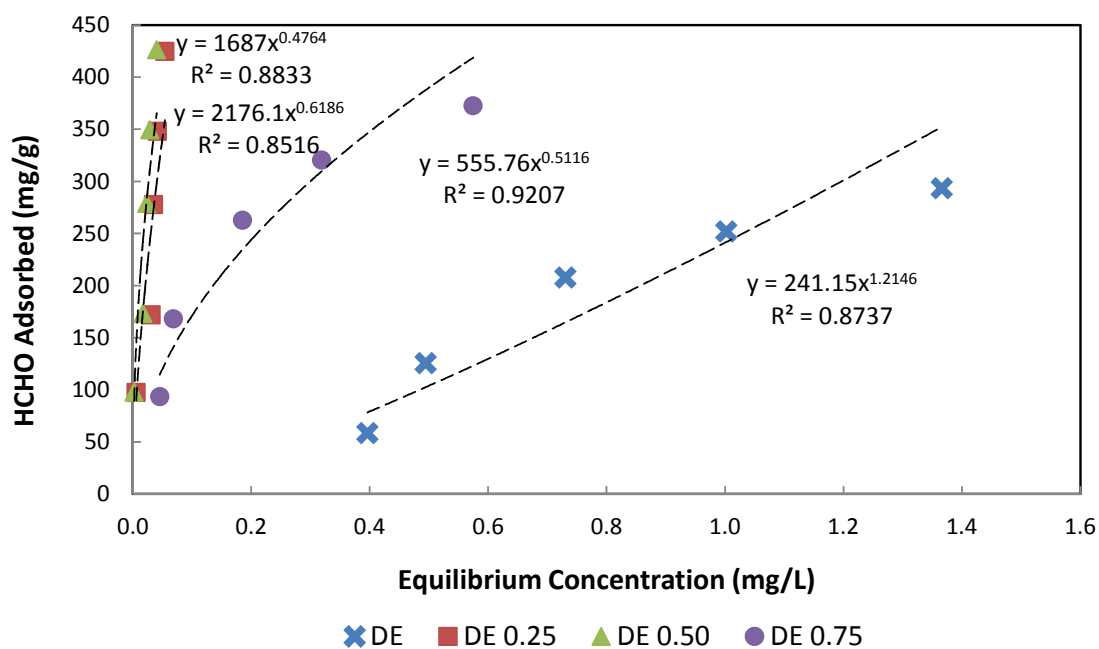


Fig. 11. Freundlich adsorption isotherm trend lines for DE and EDA-modified DEs.

**Table 1.** Langmuir isotherm parameters for the adsorption of formaldehyde in DE and EDA-modified DE.

	Langmuir Parameters		
	$q_{max}$	$K_L$	$R^2$
DE	298	0.474	0.8401
DE <sub>0.25</sub>	410	52.8	0.8810
DE <sub>0.50</sub>	334	193	0.8776
DE <sub>0.75</sub>	565	4.75	0.9482

**Table 2.** Freundlich isotherm parameters for the adsorption of formaldehyde in DE and EDA-modified DE.

	Freundlich Parameters		
	$n$	$K_F$	$R^2$
DE	0.823	241	0.8737
DE <sub>0.25</sub>	1.617	2176	0.8516
DE <sub>0.50</sub>	2.099	1687	0.8833
DE <sub>0.75</sub>	1.955	556	0.9207

For the investigated adsorbents (DE, DE<sub>0.25</sub>, DE<sub>0.50</sub> and DE<sub>0.75</sub>) where the Langmuir model fits, the maximum adsorption capacity,  $q_{max}$ , increased from 298 (mg/g) to 565 (mg/g) as the loading amount of EDA was increased. The highest value of  $q_{max}$  was obtained from the sample DE<sub>0.75</sub>, resulting to 89.5% increase in the adsorption capacity, indicating that functionalization with amine groups increased the adsorption capacity of DE. The trend in the adsorption capacity for the unmodified and modified DE (DE < DE<sub>0.50</sub> < DE<sub>0.25</sub> < DE<sub>0.75</sub>) proved that the amine groups provided greater interaction between the adsorbent (DE) and the adsorbate (formaldehyde). The adsorption capacities of the DE and EDA-modified DE are better than those of the other adsorbents used in previous studies as shown in Table 3.

#### Breakthrough Point Determination

Fig. 12 shows that DE reached its breakthrough point before the 10-minute mark. Meanwhile the modified DE samples, DE<sub>0.25</sub>, DE<sub>0.5</sub> and DE<sub>0.75</sub>, reached a  $C_o/C_i$  (ratio of outlet and inlet concentrations) of 0.23, 0.38 and 0.41, respectively, with almost similar breakthrough points at around the 45-minute mark. This data suggests that the modified adsorbents are less easily saturated than the unmodified DE proving that the amine groups have not only increased the interaction between formaldehyde and DE, but also provided adsorption sites that prevent the adsorbent from getting easily saturated with the adsorbate. The same effect was noticed by Ma *et al.* (2011) when they studied the effects of amine groups on the adsorption of formaldehyde using activated carbon (AC) concluding that the introduction of amine groups, particularly hexamethylene diamine (HMDA) improved the adsorption performance of AC by providing chemical adsorption sites.

#### CONCLUSIONS

The presence of amine in EDA increased interaction on the surface between the adsorbent and formaldehyde: the modified adsorbent loaded with the largest amount of EDA, DE<sub>0.75</sub>, had the highest adsorption efficiency.

The amine groups also provided adsorption sites that prevent the adsorbent from getting easily saturated with the adsorbate: results of the breakthrough point determination showed that, compared to the unmodified DE, the EDA-modified DE was less saturated by formaldehyde.

Overall, DE could be an excellent adsorbent for the removal of indoor air formaldehyde due to its high silica content and high porosity. Its high silica content also made possible functionalization with EDA. EDA-modified DE is an even better adsorbent because of the reaction of amine



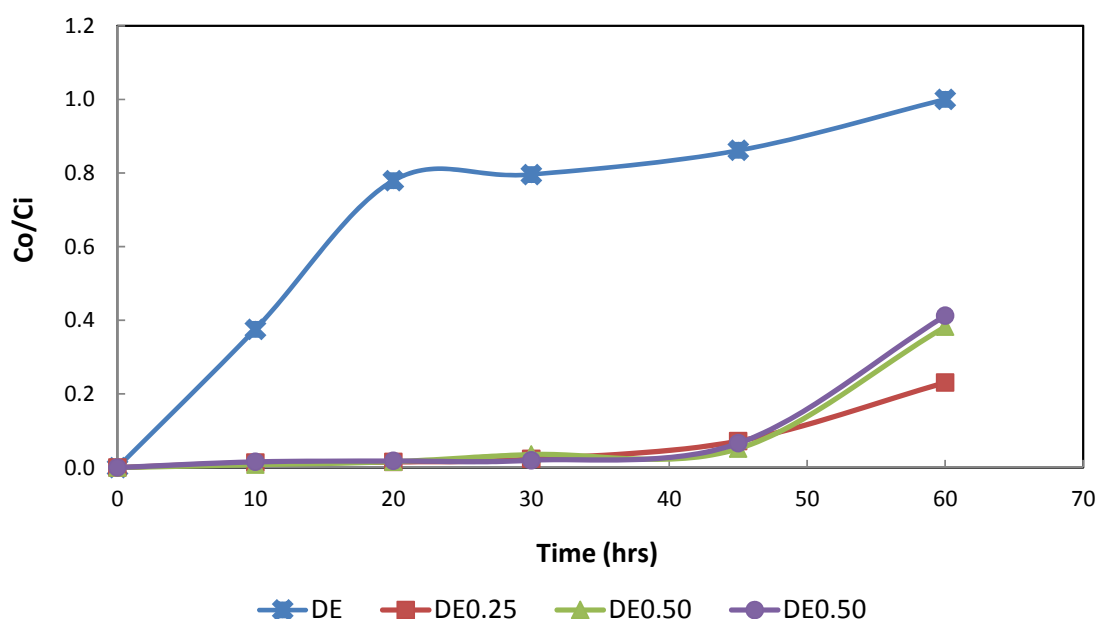


Fig. 12. Breakthrough point curve for DE and EDA-modified DE samples.

and HCHO producing imine. The presence of amine not only improved the adsorptive capacity of EDA-modified DE, but also provided adsorption sites that prevent the adsorbent from getting easily saturated with the adsorbate.

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