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# Removal of PCB from Residual Soil using Nanoalumina

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## Abstract

This paper presents the findings of a study on adsorption of poly chlorinated biphenyl (PCBs) nano-alumina. PCBs are of significant concern due their high toxicity and long environmental half-lives. Experiments were conducted using batch adsorption procedures at different PCBs concentrations, from 10 to 60 mg/L. The amounts of nano-alumina used were 0.25%, 0.50%, 0.75%, 1%, 2% and 10%. The adsorption of PCBs solution onto the nano-alumina was characterized by an initial rapid adsorption which eventually became constant within 17 hours. Results of this study indicated that nano-alumina was a good adsorbent material for PCB in this study. While at 10% of nano-alumina 54% PCB were removed by nano-alumina. The effect of pH and temperature were also investigated. The Freundlich isotherm adsorption and Langmuir isotherm adsorption were used to analysis the result of the study.

**Keywords:** Poly chlorinated biphenyl; Adsorption; Nano-alumina; Freundlich; Langmuir

## Introduction

In recent years, releases of different pollutants to the environment have drawn the attention of scientists due to their toxic effects [1]. Organochlorine (OC) compounds, such as dichloro diphenyl trichloroethane and its metabolites (DDTs) and polychlorinated biphenyls (PCBs) are particularly given much consideration because of their capability to degrade environmental quality and cause ecological risks. These compounds are hardly soluble in water and resistant to biological, chemical and photolytic degradation [2]. PCBs were utilized in several industrial fields such as electrical industry and hydraulic fluids, casting wax, carbonless carbon paper, compressors, heat transfer systems, pigments, fluoresecent light ballasts, etc. Figure 1 shows the PCB structure. Different systems can be considered for removing the environmental pollution. Adsorption is known as a popular method due to its accessibility and simplicity. Adsorption is increasingly employed for the elimination of both organic and inorganic contaminations found in water and soil. Features of adsorbent are significant in this process. The adsorbent must consist of influential traits in order to absorb the pollutants. Traditional remediation technologies have indicated confined efficacy in reduction of the concentration of contaminations in air, water, and soil.

According to Boehm F [3], nanomaterials can act more remarkably and influentially as filtration media in comparison with bigger particles with the same chemicals. Nanoparticles have the ability to appear in a wide range of morphologies, from spheres to flakes, platelets, dendritic structures, tubes and rods. The unique structure and compounds of some nanoparticles establishes them as strong adsorbents, particularly for organic substances [4]. Nano-alumina, as an effective adsorbent for the adsorption of different analytes, was characterized by strong adsorption capacity, which could be attributed to its high surface area, porosity, degree of surface reactivity, mechanical strength and low temperature modifications. However, hydrophilic naked alumina nanoparticles do not represent perfect capacity of adsorption for organic substances [5]. In fact, the adsorption capacity of naked alumina nanoparticles for organic contaminants was not effective, because of the weak interaction between organic compound and the hydrophilic surface of alumina. To overcome this problem, chemical or physical modifications of the surface y-Al<sub>2</sub>O<sub>2</sub> nanoparticles with certain functional groups containing some donor atoms were necessary, as introduced by [6]. According to Tchomgui-Kamga E, et al. [7], aluminum was an effective adsorbent for eliminating-fluoride from drinking water. Furthermore, Bhatnagar et al. 2010 highlighted that nano-alumina could remove nitrates from solutions.

## **Materials and Methods**

## Materials

Polychlorinated biphenyl was purchased from Sigma Adlrich, USA. The PCBs that was used in this study is 2-chlorobiphenyl. Its purity is 99.2%. The PCB that was used in this study was  $C_{12}H_9Cl$  (2-chlorobiphenyl). It is 188.65 g/mol in molecular weight. Nanoalumina (99.9%) was purchased as-ultra pure  $\gamma$ -Alumina ( $\gamma$ -Al<sub>2</sub>O<sub>3</sub>) powder from Inframat Advanced Material (USA). The trade name is Gamma Alumina. Some nano-alumina properties shown in Table 1. The centrifuge used was Kubota 5100. An-auto sampler Agilent Gas chromatography model 7890A equipped with 63Ni electron capture detector (ECD) was used to detect residues in the soil solution samples. All data were acquired and analyzed using Agilent CHEMSTATION' software (Agilent Corporation, MA, and USA). The limits of detection (LOD) for PCB were 0.09 µg/mL.

## Method

The PCB solutions with different concentrations were prepared in triplicates. The concentrations of PCB used in this study were 10, 20, 30, 40, 50, 60 ppm. Also the various amounts of nano-alumina, used in the experiments were 0.25%, 0.50%, 0.75%, 1%, 2% and 10% g of soil.



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Average diameter	200nm		
Purity	100%		
Form	Powder		
Vapor density(at 20)	3.5-3.9 g/cm <sup>3</sup>		
BET surface area	143.7037 m²/g		

 Table 1: Some typical properties of the nano-alumina used in this study.

The PCB solutions were added to the flask containing 12 g of soil and various amounts of selected nanomaterials. Each test experiment was conducted in 2 phases — one phase with soil and the other-without soil. Adsorption amounts were measured from the difference between the results of experiments on samples that included soil and those without soil. The mixture was shaken until equilibrium was reached using a reciprocating shaker at 150 rpm. The adsorption equilibrium time was determined from the above equilibrium time. This was followed by centrifugation at 3000 rpm for 20 minutes thus the separation between liquid phase and solid phase was obvious. A blank sample was also included. The concentrations of PCB in the clear supernatant were determined by GC. The test tubes were taken out of the reciprocating shaker bath at every interval time and centrifuged at 3000 rpm for 10 minutes, to separate a top supernatant layer from a bottom layer of soil. The amounts of PCB adsorbed on to nano-alumina were calculated from the difference between the initial concentration and the amount remaining in the supernatant. A set of controls (untreated soil) and blanks was included in all the experiments. The amount of adsorption at equilibrium, Q<sub>a</sub> (mg/kg), was calculated using the formula below:

$$Q_e = (C_o - C_e) \frac{V}{W}$$
<sup>(2)</sup>

Where  $C_o$  and  $C_e$  (mg/L) represent the liquid phase concentrations of PCB at the initial stage and at equilibrium, respectively. *V* is volume of solution and *w* is the mass of soil. The pH of the solution was adjusted (using diluted NH<sub>3</sub> and CH<sub>3</sub>COOH) to 2, 3, 4 and 9, 10 for PCB. The test tubes and their contents were shaken using a controlled environment incubator shaker (150 rpm) at a constant temperature of 28°C. To investigate the effect of temperature on adsorption, experiments were carried out at 28°C, 30°C, 32°C and 34°C of the shaking equilibrium time. The supernatants were centrifuged for 20 minutes at 3500 rpm. Subsequently, 1 ml of the supernatant extract was filtered through a 0.45 µm nylon syringe filter (Millipore<sup>\*</sup>, USA), and injected into the GC-ECD. A similar procedure was also carried out for PCB adsorptions with the soil samples. Three untreated controls (without soil and nanoparticles) for PCB were also maintained simultaneously [8].

#### **Results and Discussions**

The physicochemical properties of soil samples are enumerated in Table 2. Some of the soil properties that were investigated in this study were pH, CEC (cation exchange capacity) and water content. All soil data are expressed on a dry weight basis. The bulk density of the soils was also recorded.

In this study, FESEM was used to prepare the image of the nano-alumina. As shown in Figure 2, nano-alumina have high agglomerations and net uniform distributions. This agglomeration is shown clearly in the bottom image. The nano-alumina particles are shown to agglomerate together quite significantly. This value could be possibly lower than the actual because of particle agglomeration [9].

#### Effect of contact time

In order to determine the equilibrium (shaking) time for adsorption, the effect of contact time on adsorption was studied at room temperature. For initial concentrations of PCB in contact with nano-alumina in this study, the results are shown in Figure 3. It was found that the percentage of adsorption increased with time and slowly reaching equilibrium after about 24 hrs. As can be seen from the Figure 2, the percentage of adsorption increased with time and slowly reaching equilibrium after about 24 hrs.

### Adsorption percentage of contaminants by nano-alumina

The results of the adsorption test conducted in this study are shown in Figure 4. The initial concentrations of PCB were between 10 ppm and 60 ppm. In cleaning-up of PCB from soil and soil solutions, the amount of adsorbent used in removal process is very important for economic reasons. Thereby, the effect of adsorbent dosage on adsorption of PCBs was first carried out.

As shown in the Figure 4 the maximum percentage of adsorption of PCB on nano-alumina in this study was 54%.

The linearized Freundlich isotherm for all data is shown in Figure 5. As observed and predicted, the value of the constant K, i.e. the adsorption capacity increases with nano-alumina amounts. The K and n parameters for PCB are 5.2 and 1.07, respectively. These values are

Soil Properties	Unit
Soil Ph	4.89
Soil CEC-(meq/100 g)	8.9
Present clay fraction (%)	26.9
Present silt fraction (%)	20.5
Present sand fraction (%)	52 51

Table 2: Some soil properties.



Figure 2: Fesem images of nano-alumina.







greater than 1 indicating favorable adsorption.

Figure 6 show the experimental data fitted in the linearized Langmuir model for PCB, The  $\alpha$  and  $\beta$  values for PCB are 0.12 L/mg and 370.37 mg/kg at 0.25% nano-alumina while  $\alpha$  and  $\beta$  are 0.1 L/mg and 666 mg/kg 10% nano-alumina, respectively (Table 3).

## Effect of pH

In this study, the effect of pH on the adsorption of PCB on to nanoalumina surface was examined by varying the PCB-soil solution pH over the range of 3 to 10. The solution pH was one of the key factors that influenced the adsorption process on carbon materials by controlling the electrostatic interactions between the adsorbent and the adsorbate [10]. As illustrated in Figure 7 the amount of adsorption of PCB increased with the increase of solution pH. The initial concentration was 10 mg/L at 28°C. A maximum uptake (70%) was noted at pH 7. The electron donor acceptor (EDA) interactions were suggested by [11] as primary mechanisms for pH effects on adsorption. Chen W, et al. [11] explained that the effect of pH on organic chemical adsorption depended on the instability and electron-donor acceptor ability of adsorbent.

## Effect of temperature

Temperature also is an important parameter that can influence the equilibrium and rate of environmental process. The effects of temperature on the equilibrium adsorption of PCB on the nano-alumina were studied at different temperatures, i.e. 28°C, 30°C, 32°C, and 34°C using a temperature-controlled water bath. The initial concentration of PCB was 10 mg/L and the nano-alumina dosage was 10%.

As shown in Figure 8, the amount of PCB adsorbed onto

nano-alumina increased with increasing temperatures. When the temperature was increased from 28°C to 34°C, the trend exhibited a gradual improvement of adsorption of PCB onto nano-alumina. These phenomena could be explained by the fact that the kinetic energy of cation increased at higher temperatures, therefore, the contact between PCB and active site of adsorbent was sufficient, leading to increased adsorption efficiency [5]. In this study, it was observed that due to the small range of temperature (the average temperatures in Malaysia); it has small effect on adsorption. An increase in the amount of equilibrium adsorption of PCB with rise in temperature may be explained by the fact that the adsorbent sites were more active at the higher temperature [12]. The adsorption equilibrium depends on the temperature in two different ways. High temperature, generally, increases the rate of diffusion of the adsorbate molecules through the solution to the external and internal surface of the adsorbent and may change the equilibrium adsorption capacity of the adsorbent for the particular adsorbate [13]. Results showed that the increasing of temperature had no significant effect on PCB adsorption. High

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Figure 6: Linearized Langmuir	adsorption isotherm	n for PCB on nano-alumina.
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Freundlich isotherm parameters			Langmuir isotherm parameters			
	K	n	$R^2$	α	β	$R^2$
0.25%	5.91	1.09	0.99	0.12	370.37	0.99
0.5%	8	1.16	0.99	0.13	415.66	0.99
0.75%	6.35	1.09	0.99	0.11	526	0.99
1%	6.4	1.13	0.99	0.1	555	0.99
2%	5.92	1.12	0.99	0.1	626	0.99
10%	5.2	1.07	0.99	0.1	666	0.99

 Table 3: Some parameters of Freundlich and Langmuir isotherms for adsorption of PCB on nano-alumina.

![](_page_2_Figure_16.jpeg)

![](_page_3_Figure_1.jpeg)

temperature generally increases the rate of diffusion of the adsorbate molecules through the solution to external and internal surfaces of the adsorbent, and may change the equilibrium adsorption capacity of the adsorbent for the particular adsorbate [5]. Investigation of the results showed that the temperature has no significant effect on adsorption of PCB to nano-alumina. As the adsorption capacity increased at higher temperatures, it could be concluded that temperatures would lead-to higher chances for adsorbate to be adsorbed onto the adsorbents [14]. Since the adsorption was endothermic, higher temperatures would result in high adsorptions [14]. However, in this study it is noted that the effect of temperature on adsorption was not significant because the temperature change is small.

## Conclusion

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Comparisons among the result in this research showed that nanoalumina was a relatively good adsorbent for PCB. It was found that PCB concentrations reduced significantly in the soil samples, when the amounts of nanomaterials were increased from 0.25% to 10%. The effectiveness of the treatment depended not only on the properties of adsorbent, but also on environmental conditions and variables used for the adsorption process such as pH, temperature, contact time and initial concentration. Results illustrated that increasing of pH had positive effect on adsorption PCB on nano-alumina. Results also showed that increasing the temperature from 28°C to 34°C had small positive effect on adsorption of PCB by nano-alumina.

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