The adsorptive capacity of activated carbon and its nano-particles in removal of organophosphorus malathion from aqueous solution

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Abstract

The Date Stones (DS) is an agricultural by-product, which contributes to large quantities in Egypt. The present study consists of the preparation of activated carbon (AC) from Date stones (DS), activated chemically by H_3PO_4 and preparation nano-Particles Activated Carbon (NPAC) by ball milling techniques for the adsorption of organophosphorus malathion from aqueous solution. Physicochemical parameters for these materials were calculated through the surface area, elemental analysis, and texture porosity. The effects of environmental factors on capacity of adsorbents were conducts. The results reveal that used nano-particales activated carbon lead to minimize time and dose as well as increasing the adsorptive capacity. Nano-particales activated carbon was more effective than activated carbon in removal malathion pesticide from aqueous solution.

Keywords: Activated Carbon, Nano-particales, Malathion, Adsorption

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1. Introduction

Annual production of date stone in Egypt is about 1.1 million tons representing 16% of total world production, approximately 10-15% of this amount represents the total possible crop of date stone is 10000-165000 tons annually [11]. The adsorption process using activated carbon is becoming an important process in the treatment of industrial wastewater and purification of drinking water. The organic chemicals such as malathion are often responsible for taste, odor and color problems. Therefore, the adsorption by activated carbon can generally be used to improve such impurities. Adsorption is a surface phenomenon in which adsorbate is held onto the surface of activated carbon by Van der Waals forces and saturation is

represented by equilibrium point. It is one of the well-known methods used in the removal of such hazardous compounds from polluted waters [1].

Activated carbon becomes versatile adsorbents with a wide range of applications that concerns principally with the removal of impurity species by adsorption from the liquid or gas phase. Activated carbon adsorption has proved to be the least expensive treatment option, particularly in treating low concentrations of wastewater streams and in meeting stringent treatment levels [3]. The characteristic of activated carbons itself with their large surface area, microporous character and chemical nature of their surface have made them potential adsorbents for the removal of heavy metals from industrial wastewater.

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Adsorption is a physical phenomenon, depending largely upon surface area and pore volume. Surface area of activated carbon typically is in the range of 300–2500 m^2/g [2]. The adsorption performance of organic pollutants found in aqueous conditions depends on the microspores in activated carbon for which adsorbent is concern, activated carbon is common versatile adsorbent used for removing organic compounds and it is usually regenerated by thermal reactivation or caustic washing. Activated carbon becomes versatile adsorbents with a wide range of applications that concerns principally with the removal of impurity species by adsorption from the liquid or gas phase. It is also used as catalyst support, in waste water treatment, in sugar syrup purification, in air pollution control as well as in the pharmaceutical and chemical industries. Almost any carbonaceous material can be used as precursor for the preparation of activated carbons [1]. For instance, adsorption performance of activated carbon originated from oil palm shell[7], coconut shell [9] wood and dust coal activated carbons [20], rice husk carbon [17], fly ash [15], nut shell [14]. Wood, peat, and lignite are commonly used for the decolorizing materials.

The large pore size permits the water pollutants to enter the carbon particle where they are adsorbed. The particle size of activated carbon affect the performance of adsorption of activated carbon; the smaller the particle size of activated carbon, the faster toxic substances are removed and the smaller the particle size of activated carbon, the larger the contact surface area to adsorb. Therefore, it is assumed that the increase of contact surface area accelerated the removal of pesticide onto activated carbons. However, the large pores cause a large reduction in the internal surface of carbon compared to others which means a loss in adsorption potential. Accordingly, in this study we prepared the nano-particles activated carbon by ball milling techniques, whereas increasing the surface area related with increasing efficiency.

Malathion is a broad spectrum organophosphate insecticide and miticide first registered in 1956. Malathion has numerous commercial agricultural, industrial, governmental, and homeowner uses.

In 2000, approximately 11–13 million pounds of malathion were used annually; currently, approximately 15 million pounds are used annually [10]. Many studies reported that the main pesticide residues were organophosphate [4,8]. Organophosphorous pesticides (OPPs) are a group of highly toxic agricultural chemicals widely used in plant protection. Common members of the family are methylparathion, malathion, dimethoate, phosphamidon, phorate, fienitrothion and monocrotophos [19]. There are many methods to treat raw water for its possible harmful organics caused by malathion contamination. This includes destroying ozonization, oxidation, disinfection, photocatalysis or removal by adsorption. Among these techniques, adsorption has proved to be the most appropriate method to treat effluents that offering advantages over conventional process [16].

The objective of this study is to determine the efficiency of normal particles and nano-particles of activated carbon that derived from date stones in removing malathion from aqueous solution. The characterization of normal and nanoparticales was conducted. The effectiveness of activated carbon to remove pesticide can be used as an alternative medium to treat the contamination from a water environment or wastewater. At the early stage of the study, the batch tests were conducted to investigate the adsorption capacity of Malathion onto normal particles and nano-particles of activated carbon. The effects of the environmental factors (temperature and pH) on efficiency of normal particles and nano-particles of activated carbon were also investigated.

2. Materials and methods

2.1. Chemical and reagents.

All the solvents (acetonitrile, petroleum either) used for ML extraction were HPLC grade and purchased from Merck, Darmstadt., Germany.

2.2. Malathion.

Malathion; 95% of active ingredient was supplied by Sigma Aldrich. Solutions were prepared with distilled water passed through a Mili-Q water purifier (Millipore Corporation) to reach concentration at (1µg/ml)..

2.3. Preparation of activated carbon (AC) and its nano-particales(NPAC) derived from dates stones.

Dates stones as one of agricultural by-products was chosen as a precursor for the production of activated carbons. The following chart explains the main steps which carried out for the production of AC derived from dates stones.

The activated carbon was prepared from dates stones collected from dates factories in Upper Egypt.

The dates stones was dried at 70 ℃ in a drying oven for 1 day then crushed and sieved to semi fine particles (500- 850 μm). These particles were used for the production of activated carbon through a single-step chemical activation using H_3PO_4 according to Seif *et al.* (2011) [18]. In brief: One hundred grams of crushed dates stones was soaked in phosphoric acid (85 wt. %) at ratio of 1:3 (w/w). Slightly agitated to ensure penetration of the acid throughout. The mixture was heated to 70 °C for 2 h and left overnight at room temperature. The impregnated solid was activated into muffle furnace. The temperature was raised to 500 °C for 2 h at rate of 5°C /1 min. The acid was removed by washing with distilled water till the pH reached 6.8. The carbon product was dried at 110 °C for 24 h using electric oven. Nano-Particles Activated Carbon (NPAC) was prepared by ball milling techniques after preparation of activated carbon (Fig. 1).
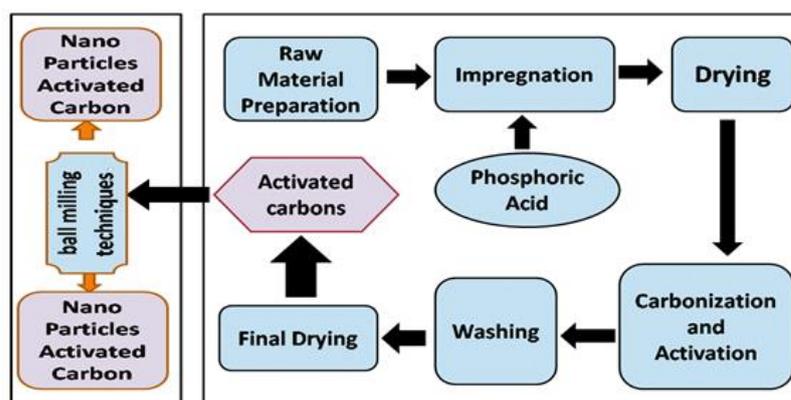

Figure 1: Steps involved with the production of activated carbon and Nano-particales from dates stone

2.4. Characterization of (AC) and nano-particales (NPCA).

The content of carbon, hydrogen and nitrogen in activated carbon (AC) and Nano-particles activated carbon (NPAC) deviated from date stone using elemental analyzer (Thermo Electron Flach EA 1112).

The surface area and pore size and pore distribution of AC and NPAC were determined from the adsorption- desorption isotherm of nitrogen at 77K. The surface area (S_{BET}) was calculated by Brunaur- Elemmet-teller (BET) equation, the pore size distribution was calculated on the basis of desorption data by employing the barrett-joyner- halenda (BJH) methods [23].

Sample was measured and taken in a crucible. It was then heated to 750 C for 1.5 hr. During this test the crucible was left open. After the required heating, the crucible was cooled in a descicator and then weighed to determine the ash content.

The surface topography of the AC and NPAC prepared was observed by scanning electron microscope (GSM-6480, Japanese Corporation of Electron).

2.5. Batch kinetic experiments.

AC and NPAC were characterized and tested for their potential application for the removal of ML from aqueous solution as follow: An amount of 0.1, 0.2 and 0.5g of AC and 0.05, 0.1 and 0.2g of NPAC was incubated with a 100 ml of ML (1mg/L). The mixture was agitation at 100rpm using water-bath shaker at 25° C for 15, 30, 60, 120, 180, 240 and 300 min. After the agitation period, the samples were filtrated using filter paper Whatman No. 40.

The amount of ML adsorbed at each time (Qt, mg ML/g adsorbent) was calculated based on the following equation:

$$Qt = (Co − Ct)\ V/m$$

Where: Co and Ct are the liquid-phase concentrations (mg/l) of ML at initial and at any given time respectively; V is the solution volume (L) and m is the adsorbent mass (g).

The equilibrium condition was considered when no further decrease in the ML concentration was occurred.

2.6. Effect of pH and temperature on the adsorption capacity of AC and it nano-particales (NPAC).

To investigate the effect of pH on the adsorption capacity of AC for ML from aqueous solutions, an initial amount of AC (0.5g/100ml) was incubated with concentration level of ML (1mg/L) in 30 ml of aqueous solution. The mixtures were agitated using a thermo stated shaker water-bath at 100 rpm for 120 min at 25 ± 1 °C. The pH was adjusted using a NaOH (0.1 M) or HCl (0.1 N) and measured by pH meter (Genway, Model 3505).

In the context to study the effect of pH on the adsorption capacity of NPAC for ML from aqueous solutions, an initial amount of NPAC (0.1g/100ml) was incubated with concentration level of ML (1mg/L) in 30 ml of aqueous solution. The mixtures were agitated using a thermo stated shaker water-bath at 100 rpm for 60 min at 25 ± 1 °C. The pH was adjusted using a NaOH (0.1 M) or HCl (0.1 N) and measured by pH meter (Genway, Model 3505).

To study the influence of temperature on the adsorption capacity of AC and NPAC for ML from aqueous solutions, the same procedures were followed under an additional temperature (37°C).

2.7. Extraction of malathion from water samples.

Extraction of malathion was carried out based on the procedures described by Chuanhong (2004) [6]. The procedures can be summarized in brief as follows: All sample filtered through filter paper, then 10 ml of each sample was transferred to a 100-ml reparatory funnel. After adding 2ml of methylene choloride the separatory funnel was

sealed and then shaken vigorously for 1 to 2 minutes with periodic venting to release excess pressure. Once the funnel was still for 10 minutes, the extract for organic layer was collected. The extraction was repeated twice, using fresh portion of solvent. The resulting three portion of the extract were combined and dried with anhydrous sodium sulfate, then evaporated to near dryness. The residue was dissolved with 1 ml of acetone and transferred into the sample vial for GC analysis.

2.8. Determination of malathion by gas chromatography (GC).

Malathion was determined with GC (hp 5890 series II) equipped with Electron capture detector (ECD). The separation was accomplished on CP-Sil 8CB (30m x 0.25 mm i.e. 0.25 flim thichness). Splitless Injection was used at 250 °C and the temperature of detector was 300°C. Nitrogen was used as a carrier gas at flow rate of 1 ml/min. The temperature program was isothermal temperature program, it performed as following initial temperature 160 °C help for 0 min. the same temperature (160 °C) held constant for 25 min. the identification of malathion was performed by the retention time and compared with certified standards at the same condition. The quantities of malathion were calculated on peak area basis. The retention for malathion was 14.910 min.

2.9.Statistical analysis

All Data were expressed as means ± standard error of mean (SEM). One-way analysis of variance (ANOVA) was performed to test for differences between the groups mean. Significant differences between the means were determined by Duncan's multiple range test and P values < 0.05 were regarded as significant [22].

3. Results and Discussion

The removal ability of malathion using adsorbent agent was studied on aqueous solution. Activated carbon (AC) and Nano-particales activated carbon (NPAC) were used as an effective adsorbent agent. Both of adsorbents were added in three levels (i.e.0.05, 0.1 and 0.2 % v/v). These levels succeeded to remove the malathion from aqueous solution since it has the ability to adsorb the toxin from aqueous solutions.

3.1. Production and characterization of Activated carbon (AC) and developed nano-particales activated carbon (NPAC).
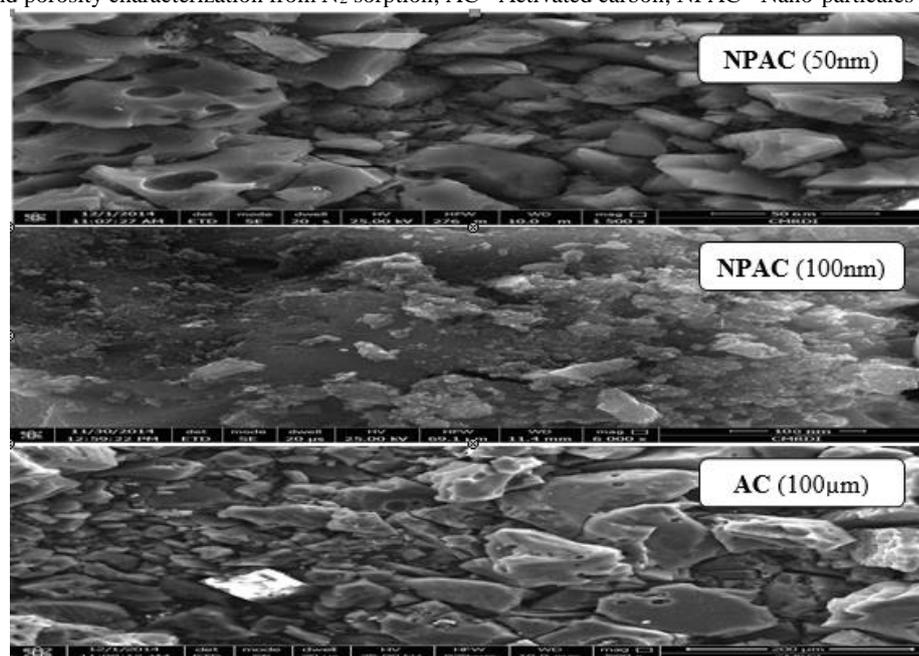
The specific surface area and porosity measurements were achieved using Burunauer-emnett-tellrr nitrogen adsorption technique (BET).

Characteristics of the prepared activated carbon are presented in Table (1). The size distribution of mciropores of activated carbon is understood to be one of the critical factors determining its applicability.

Table 1. The characteristics of the activated carbon and its nano-particales.

Parameter		Items	
		AC	NPAC
Surface Area S_{BET}		916 (m^2/g)	2020 (m^2/g)
Porosity (%)		48 %	69%
Particles size		10-100 μm	50-200 nm
Ash content (%)		4.3%	3.1 %
Elemental Content (%)			
	C	74.9 ± 0.3	76.7 ± 0.4
	N	74.9 ± 0.3	76.7 ± 0.4
	H	74.9 ± 0.3	76.7 ± 0.4

Surface area and porosity characterization from N_2 sorption; AC= Activated carbon; NPAC= Nano-particales activated carbon.

Figure 2. Scanning Electronic Microscope images of AC and its NPAC

Data located in Table (1), showed that the porosity and specific surface area (S_{BET}) of AC and its nano-particales, the surface area (S_{BET}) and porosity of nano-particales AC was higher than AC, moreover the capacity of adsorption of nano-particales of was higher than normal particles AC.

Particles size of AC and its nano-particales was shown in figure 2.

3.2. Effect of contact time and carbon dosage on adsorbtion of malathion from aqueous solution.

Data presented in Figures (3 and 4) showed the kinetic of sorption for each sorbent agent (AC and

NPAC) against ML in aqueous solution. The results indicated that the maximum adsorption of ML from aqueous solution was achieved within 120 min for AC at dose 0.1 g/ 100ml, which 7.46 mg ML/g AC. However, the percentages of reduction of ML reached 31, 95.6 and 78 % at sorbent dose 0.05, 0.1 and 0.2g respectively (Table 2).
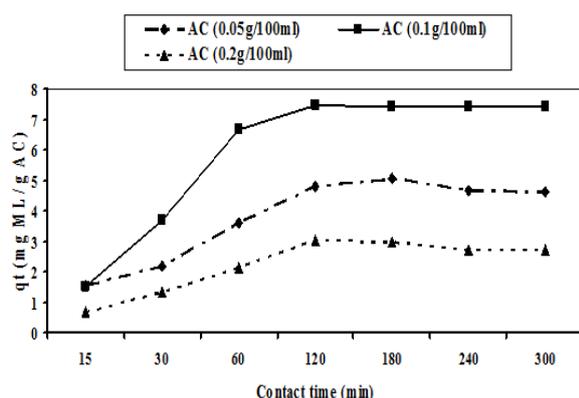

Figure 3. Effect of contact time on the adsorption of malathion by activated carbon at different carbon doses.
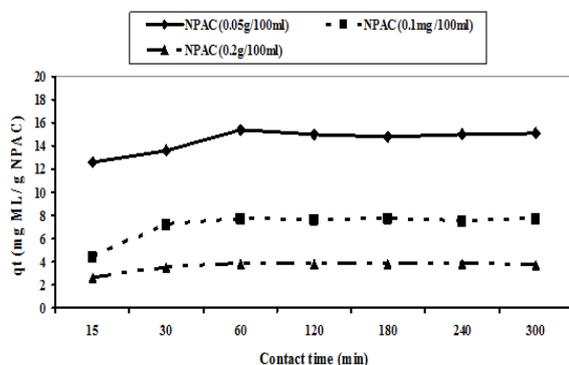

Figure 4. Effect of contact time on the adsorption of malathion by Nano-particales activated carbon at different carbon doses.

In addition, the results indicated that the maximum adsorption of ML from aqueous solution was achieved within 60 min for NPAC which recorded 15.38, 7.65 and 3.76 mg ML/ g NPAC using 0.05, 0.1 and 0.2g of NPAC respectively. However, the percentages of reduction reached 98.6, 98.1 and 96.3% using 0.05, 0.1 and 0.2gof NPAC respectively (table 3).

Table 2. Effect of contact time on the % reduction of ML by AC at different carbon doses.

Contact time (min)	Carbon doses		
	AC (0.05g/100ml)	AC (0.1g/100ml)	AC (0.2g/100ml)
15	10	19.45	17
30	14	47.5	34.2
60	23	85.66	55.2
120	31	95.6	78
180	32.6	95.31	76
240	30.1	95.1	69.3
300	29.8	95.1	69.1

AC= activated carbon. ML= Malathion; min= minutes

Table 3. Effect of contact time on the % reduction of ML by NPAC at different carbon doses

Contact time (min)	Carbon doses		
	NPAC (0.05g/100ml)	NPAC (0.1mg /100ml)	NPAC (0.2g/100ml)
15	81	56.1	67.45
30	87.2	92.24	89.2
60	98.6	98.1	96.3
120	96.2	98	96.72
180	95.14	98.14	96.31
240	96.24	98.24	96.74
300	96.86	98.3	96.1

NPAC= Nano-particales activated carbon; ML= malathion; min= minutes

3.3. Effect of pH on the adsorption capacity of adsorbent agents towards ML

Since the surface charge of the adsorbent could be modified by changing pH of the solution, pH considers one of the most important parameters affecting the adsorption capacity of carbon towards organic substances. The effect of pH on adsorption of ML onto the adsorbent agents (AC and NPAC) was conducted in the pH ranged from 3 to 9. The equilibrium adsorption of AC against ML with initial level concentration of 78μg/100ml was found to increase from 6.8 to 7.5mg ML/g AC when the initial pH of solution was increased from 3 to 7, respectively. However at pH 9, the adsorption capacity decreased again and reached 7.1mg ML /g AC (Fig. 4, A).

For NPAC, the same trend was found since the maximum adsorption equilibrium (max qt) was increased from 14.5 to 15.8 mg ML/g NPAC with the increase of pH from 3 to 7. While, at pH 9, the max qt decreased again and recorded 14.1mg ML/g NPAC (Fig.4, B).
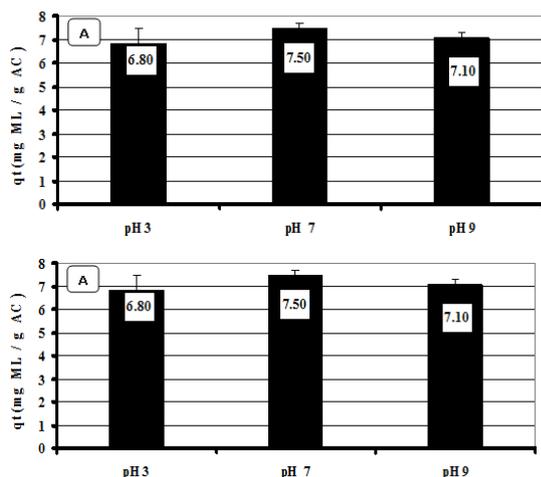

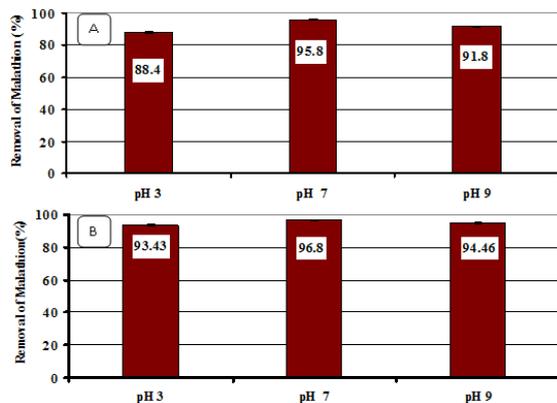**Figure 5.** Effect of pH on the adsorption capacity of AC (A) and NPAC (B) against ML.

Figure 6. The effect of pH on % removal of ML from aqueous solutions using AC (A) and NPAC (B).

3.4. Effect of temperature on the adsorption capacity of AC and its nano-particales towards ML.

The results presented in Figs (6 A,B and 7 A, B) indicated that as the temperature increased from 25 °C to 37 °C, the adsorption of ML onto AC and NPAC was slightly increased. The results presented in Fig. (6 A and B) indicated that the removal percentage of ML from aqueous solution using AC at initial level of 78µg/100ml increased from 95.6 % to 98.8 % when the temperature

increased from 25 to 37 °C (Fig 6 A), since the maximum adsorption capacity was increased only from 7.46 to 7.85mg ML/g AC (Fig 7 A). In the same respect, at higher temperature (37 °C) the maximum adsorption capacity was increased and recorded 17.62 mg ML/g NPAC. Whereas, the maximum adsorption capacity (max qt) was found to be 15.41 mg ML/g NPAC at temperature 25 °C as shown in (Fig. 7 B).

The current results represented in Fig (8 B) indicated that the percentage removal of ML was increased from 96.8 to 98.9 % as the temperature increased from 25 to 37 °C.

The results presented in Fig (6A, B) revealed that the removal percentage of ML from aqueous solutions at pH 3 was found to be 88.4% for AC. However, this percentage reached 95.8% when solution pH reached 7.0. Similarly, the percentage of reduction for NPAC reached 93.43% at pH 3, while this percentage was increased at pH7 and recorded 96.8% at the same concentration level.
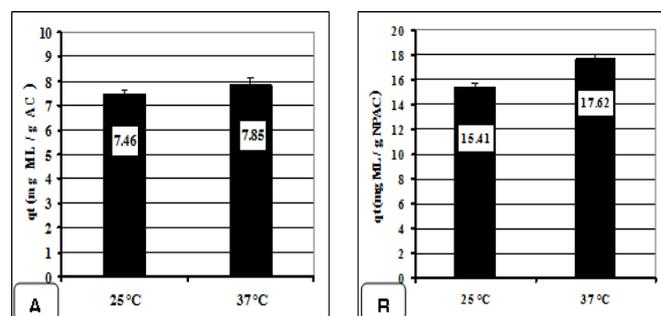

Figure 7. The effect of temperature on % removal of ML from aqueous solutions by AC (A) and NPAC (B)
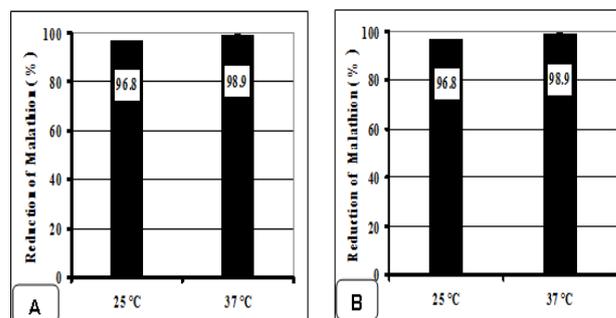

Figure 8. The effect of temperature on the adsorption of AC (A) and NPAC (B) against ML.

The adsorption reached to equilibrium stage where the amount of ML being adsorbed to adsorbent agents (AC or NPAC) was equal to the amount of

ML desorbing from these agents. From the result in Tables (2 and 3) and Figures (3 and 4) above consideration, it could be observed that NPAC was more effective in the removing of ML from aqueous solution than AC due to the highest surface area of NPAC.

Whereas the max qt (maximum adsorption at equilibrium) for NPAC was reached 15.38 mg/g NPAC at dose of 0.05g/100ml at equilibrium time (60 min). While, the max qt for AC was reached 7.46mg ML/ g AC at dose of 0.1g/100ml at equilibrium time (120 min), it's due to the highest surface area and % of porosity of NPAC. Also, the present results illustrated that surface charge of the adsorbent could be modified by changing pH of the solution. Generally pH is considered one of the most important parameters affecting the adsorbtion capacity of carbon towards malathion. The effect of pH on the adsorbent capacity of AC and NPAC was tested at 100 mg of AC and at 50 mg of NPAC. It is well known that malathion is relatively stable under neutral and slightly acidic condition.

The present results indicated that the retention of malathion onto AC and NPAC had the same behavior and the percentage removal was increased as pH elevated from 3.0 to 7.0 and decreased again with pH 9.0 for AC and NPAC. Thus, AC succeeded to remove 95.8 % of malathion at pH= 7.0, and initial concentration 78µg/100ml and the maximum removal percentage of malathion at the same condition onto NPAC was 96.8 percent. Also, the results presented (fig. 5) indicated that the maximum adsorption of malathion reached a highest value at pH 7.0 was 7.50 mg ML/g AC and 15.8mg Ml/ g NPAC. Those results were found to be consistent with the results obtained by Chatterjee *et al.,* (2010*)* [5] who studied the adsorbtion of malathion by Rhizopus oryzae biomass at pH range from 2 to 8. They concluded that the adsorbtion capacity increased at higher pH values. Furthermore, the findings of the present study were found to be in agreement with the results obtained by Singh *et al.,* (2010) [21] who found that the adsorbtion of malathion by flay ash was increased from 96 to 99 % at the initial concentration of of 1 ppm and increased from 89 to 90% at 10 ppm at a high range of pH.

The data illustrated in figures (7 and 8) show the effect of temperature on the percentage removal and the adsorbtion capacity of adsorbtion agents (AC and NPAC) against malathion at equilibrium times. The present results showed that the maximum adsorption of malathion onto AC increased from 7.46 mg ML/g AC to 7.85 mg ML/g AC when the temperature increased from 25 to 37 °C. The removal percentage of malathion was increased from 96.8 to 98.8 with increased the medium temperature from 25 to 37 °C. It was well documented that temperature has three major effects on the adsorption process since (1) increasing the temperature is known to increase the diffusion rate of the adsorbate molecules across the external boundary layer and in the internal pores of adsorbent particles, which was found to increase by increasing the temperature, (2) the viscosity of the solution is deceased by increasing the temperature, and (3) the solubility of pesticide in water is increased as the temperature increased [13]. The current were agreement with those obtained by Gupta *et al.,* (2001) [12] who studied the adsorption isotherms of lindane and malathion using bagasse fly ash at three different temperatures and suggested that the adsorption is almost unaffected whish rising temperature at lower concentration of lindane and malathion, however, it decreased at higher concentration for both the pesticides. The effect of temperature can be explained on the basis of solubility. With the increasing temperature, the solubility of malathion increases. Therefore, exhibits lower tendency to go to the adsorbent surface and get adsorbed.

From the results it can be concluded that activated carbon and its nano-particales success in removal malathion from aqueous solution. Using nano-particales activated carbon lead to minimize time and dose as well as increasing the adsorptive capacity. Nano-particales activated carbon was more effective than activated carbon in removal malathion pesticide from aqueous solution.

Compliance with Ethics Requirements. Authors declare that they respect the journal's ethics requirements. Authors declare that they have no conflict of interest and all procedures involving human / or animal subjects (if exist) respect the specific regulation and standards.

References

1. Acharya, J.; Sahub, J.N.; Sahoob, B.K.; Mohantyc, C.R. and Meikapb, B.C. (2009). Removal of chromium (VI) from wastewater by activated carbon developed from tamarind wood activated with zinc chloride. Chemical Engineering Journal, 150:25–39.

2. Ansari, R. and Sadegh, M., Application of activated carbon for removal of arsenic ions from aqueous solutions. *E-Journal of Chemistry*, **2006**, *4*(1), 103–108.

3. Babel, S. and Kurniawan, T.A. Low-cost adsorbents for heavy metals uptake from contaminated water: a review. Journal of Hazardous Materials., **2003**, *97*, 219–243.

4. Bai, Y.H.; Zhou, L. and Wang, J. Organophosphorous pesticides residues in market foods in Shaanxi area, China. Food Chemistry, **2006**, *98*, 240–242.

5. Chatterjee, S.K.; Das, R.; Chakravarty, A.; Chakrabarti, S. and Guha, A.K., Interaction of malathion, an organophosphorus pesticide with Rhizopus *oryzae*, *J. Hazard. Mater.*, **2010**, *174*, 47-53.

6. Chuanhong, T. U., Analysis of organophosphorus pesticides with Agilent technologies publication, **2004**, 5989-1335, http://www.agilent.com/cs/library/applications/5989-1335EN.pdf

7. Collin, G.J.; Hasnul, F.M.Z. and Siti, F.D., Treatment of landfill leachate in Kayu Madang, Sabah: textural and chemical characterization (part 1). Malaysian., *Journal of Analytical Sciences*, **2005**, *10*, 1–6.

8. Dalal, M.; Dani, R.G. and Kumar, P.A., Current trends in the genetic engineering of vegetables crops. *Scientia Horticulyuria.*, **2006**, *107*, 215–225.

9. Dwivedi, C.P.; Sahu, J.N.; Mohanty, C.R.; Raj Mohan, B. and Meikap, B.C., Column performance of granular activated carbon packed bed for Pb(II) removal. *Journal of Hazardous Materials*, **2008**, *156*, 596–603.

10. Edwards, D., Registration eligibility decision for malathion. Prevention Pesticides and Toxic substances., **2006**, Case No. 0248 US EPA.

11. FAO (Food and Agriculture Organization of the United Nations), Fishery Statistics 2002. Rome: Capture Production, **2004**, FAO.

12. Gupta, R. C. (2006). Toxicology of Organophosphate and Carbamate Compound. Elsevier Academic Press.

13. Moreno-Castilla C., Adsorption of organic molecules from aqueous solutions on carbon materials. *Carbon*, **2006**, *42*, 83–94.

14. Nomanbhay, S.M. and Palanisamy, K., Removal of heavy metal from industrial wastewater using chitosan coated oil palm shell charcoal. *Electronic Journal of Biotechnology*, **2005**, *8*, 43–53.

15. Rastogi, K.; Sahu, J.N.; Meikap, B.C. and Biswas, M.N., Removal of methylene blue from wastewater using fly ash as an adsorbent by hydrocyclone. *Journal of Hazardous Materials*, **2008**, 158, 531–540.

16. Ravikumar, K.; Krishnan, S.; Ramalingam, S. and Balu, K., Optimization of process variables by the application of response surface methodology for dye removal using a novel adsorbent. *Dyes Pigments*, **2007**, 72, 66–74.

17. Sahu, J.N.; Agarwal, S.; Meikap, B.C. and Biswas, M.N., Performance of a modified multi-stage bubble column reactor for lead(II) and biological oxygen demand removal from wastewater using activated rice husk. *Journal of Hazardous Materials*, **2009**, *161*, 317–324.

18. Seif, M.M.; Khalil, F.A.; Abdel-Aziz, A.M.; El-Kady, A.A. and Abdel-Galil, M.M., Application of Egyptian adsorbent agents for the removal of Deoxynivalenol mycotoxin from aqueous solution. *J. of Food Industries and Nutr. Sci.*, **2011**, *1*, 75-86.

19. Senthilkumaar, S.; Krishna, S.K.; Kalaamani, P.; Subburamaan, C.V.N. and Subramaniam, G., Adsorption of organophosphorous pesticide from aqueous solution using ''waste'' jute fiber carbon. *Modern Applied Science*, **2010**, *4*(6), doi: http://dx.doi.org/10.5539/mas.v4n6p67

20. Singh, C.K.; Sahu, J.N.;Mahalik, K.K.; Mohanty, C.R.; Raj Mohan, B. and Meikap, B.C. (2008). Studies on the removal of Pb(II) from wastewater by activated carbon developed from tamarind wood activated with sulphuric acid. *Journal of Hazardous Materials*, **2008**, 153, 221–228.

21. Singh, V.K.; Singh, R.S.; Ravi, S.; Tiwari, P.N.; Singh, J.K.; Gode, F. and Sharma, Y.C. (2010). Removal of malathion from Aqueous Solutions and Waste Water Using Fly Ash. *J. Water Resource and Protection*, **2010**, *2*, 322-330.

22. Sokal, R.R. and Rohlf, F.J., Biometry: *The Principles and Practice of Statistics in Biological Research*, 3rd Edn., W.H. Freeman and Co., New York, **1995**, pp: 887.

23. Tang, Y.; Liu, Q. and Chen, F. (2012). Preparation and characterization of activated carbon from waste ramulus mori. *Chemical Engineering Journal*, **2012**, *203*, 19–24.