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Article *in* Advanced Materials Research · April 2014

DOI: 10.4028/www.scientific.net/AMR.925.105

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Advanced Materials Research Vol. 925 (2014) pp 105-109
Online available since 2014.Apr.17 at www.scientific.net

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Removal of benzene from aqueous solution using carbon nanotube synthesized from fuel oil waste

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Keywords: CNT; Benzene; Fuel oil; Adsorption

Abstract:

This investigation is dealing with adsorption of benzene compound from aqueous solution using a new carbon nanotube (CNT) synthesized from a fuel oil waste of power plants which identify by FE-SEM and TEM. It was found that a CNT has a very significant adsorption for benzene compared to that of non-activated carbon. The equilibrium adsorption data were analyzed using adsorption models of Langmuir, Freundlich and Temkin. The results showed that the model isotherms are fitting very well with the experimental data. Kinetic study was conducted and the results pointed out that a pseudo-first order model was represented the data. Values of the activation thermodynamic functions were calculated through equilibrium constants at different temperature. All values of Gibbs functions were negative with values of -1.6 and -13.0 $kJmol^{-1}$ for non-active and CNT respectively, while values of enthalpy and entropy were about -33$kJmol^{-1}$ and-65JK^{-1} mol^{-1} for CNT respectively. These results indicated that the adsorption process was feasible, spontaneous and exothermic.

1.Introduction

Carbon nanotubes have different characteristics of that of the macro-scale. Uses of carbon nanotubes are concern mostly on environmental aspects as unique sorbents for removal of pollutants [1-3]. Adsorption technology has been used widely for a removal of organic compounds from aqueous solution using activated and nanocarbon materials [4-6]. However, the removal of benzene has been investigated with highly significant approach using carbon nanotubes [1,2,7], graphitized carbon [8], carbon nano porous coated with polymers [4]. The main objective of this research is to remove the residual benzene using synthetic CNT manufactured from fuel oil wastes as the adsorbent.

2. Experimental

The samples of fuel oil residue were collected from the power plant in Al-Anbar area, Iraq, and refining according to the procedure mentioned elsewhere [9].

The CNT was synthesis from fuel oil waste using ultrasonic technology and identified using FE-SEM and TEM as shown in Figs. (1) and (2) [10].

2.1. Characterization

Surface area, density, ash content, pH and Moisture content of prepared CNT were determined as shown in table (1).

Table1: Characteristics of CNT and non-activated carbon samples.

Samples	*Surface area m^2/g*	*Density g/cm^3*	*Ash%*	*pH*	*Moisture %*
CNT	1050.4	0.4908	0.11	3.79	1.600
Non-active carbon	72.32	0.5834	2.61	6.15	0.773

2.2. Adsorption studies

Determination of equilibration time for benzene adsorption on CNT was done by shaking 5ml of benzene at concentrations of 50,200 and 300 ppm with 0.1gof adsorbate at constant temperature of 283,293,313 and 333 K for 120 min. Sub-sample were taken from each concentration at 10 to 120 min for kinetic studies and then filtrated. UV-Vis spectrophotometer has been used to measure the absorbance for benzene at equilibrated solutions at certain wavelength (204 nm).The quantity of adsorbate was calculated by using the following formula:

$$q_e = V_{Sol} (C_o - C_e)/M \quad (1)$$

Where q_e is quantity of adsorbate (mg/g), V_{Sol} is total volume of adsorbate solution (L), C_o is initial concentration of adsorbate solution (mg/L), C_e is concentration of adsorbate solution at equilibrium (mg/L) and M is weight of adsorbate (g). While the removal percentage of benzene and equilibrium adsorption q_e (mg/g) was uptake, using the formula 2.

$$\% \textit{ adsorption Efficiency} = (C_o - C_e)/ C_o \times 100 \quad (2)$$

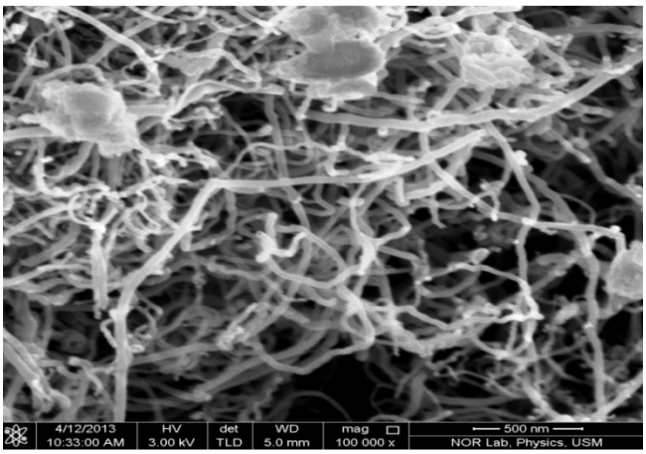

Fig.1: FE-SEM image of CNT

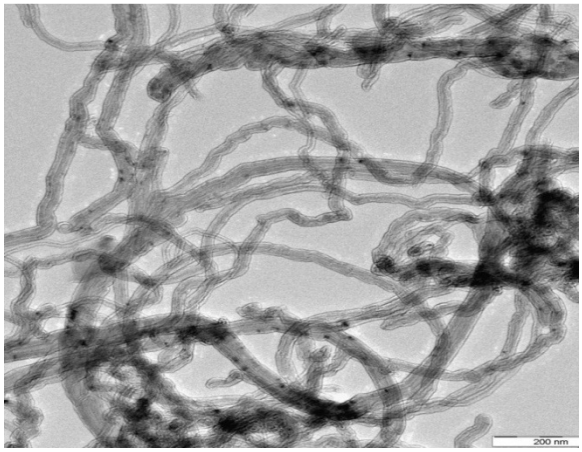

Fig.2: TEM image of CNT

3. Results and Discussions

3.1. Adsorption Equilibrium

The isotherm of the benzene adsorption by synthetic CNT was represented by applying the Langmuir, Freundlich and Temkin adsorption models, and it was found that the adsorption process on the synthetic CNT fits very well with the isotherm models table (2). The Freundlich linear isotherm is expressed as:

$$\text{Log } Q_e = \log K_f + 1/n \log C_e \quad (3)$$

Where Q_e is the equilibrium value of benzene adsorbed per unit weight of synthetic CNT powder, i.e. a liquid-phase sorbate concentration occurred at equilibrium and K_f as the Freundlich constant. Freundlich constants are shown in table (2) while the relations are clearly indicated that the Freundlich isotherm model fits the analyzed data according to its correlation coefficients (R^2).

The synthetic CNT powder takes up benzene on a heterogeneous surface by multilayer adsorption as described by Langmuir. However, the Langmuir linear isotherm is expressed as

$$C_e/Q_e = (1/K_L Q_o) + (1/Q_o) Ce \quad (4)$$

Where Q_o is the maximum amount of adsorption corresponding to complete monolayer coverage and K_L is the Langmuir constant. Table (2) shows the Langmuir factors and relations.

The linear form of Temkin isotherm is expressed as:

$$q_e = B \ln A + B \ln C_e \quad (5)$$

Table (2) shows the Temkin factors and relations.

Table 2: The values of Langmuir, Freundlich and Temkin constants at different temperatures.

	T K	Langmuir constants			Freundlich constants			Temkin constants		
		R^2	a	K_L	R^2	n	K_F	R^2	B	A
CNT	283	0.980	9.667	111.11	0.935	5.587	9.506	0.924	1.760	245.25
	293	0.935	2.399	28.571	0.981	2.222	5.916	0.974	4.899	2.463
	313	0.956	3.423	38.461	0.992	2.012	4.989	0.916	1.850	67.12
	333	0.948	2.256	25.641	0.985	1.767	4.335	0.986	5.662	1.357
Non-active carbon	283	0.806	- 0.013	0.016	0.976	0.337	2.68×10^{-5}	0.868	9.463	0.031
	293	0.795	- 0.012	0.014	0.984	0.325	1.44×10^{-5}	0.913	9.509	0.029
	313	0.580	- 0.013	0.007	0.907	0.237	8.9×10^{-8}	0.960	11.626	0.024
	333	0.385	- 0.013	0.002	0.802	0.165	2.917×10^{-11}	0.931	13.718	0.022

The adsorption rate of benzene increased with benzene concentration and decreased with temperature. (Tables 3, and 4). A complete removal of benzene concentration was obtained for initial concentration of 50 ppm and temperature of 283 K. Hence it is clearly proved that benzene adsorption by synthetic CNT agrees fair enough with the Langmuir, Freundlich and Temkin adsorption models. It was poorly fit with Langmuir isotherm model using non-active carbon. The correlation coefficient was very high throughout the experimental range of benzene concentrations studied.

Table3: The adsorption percentage of benzene at concentration 50 mg/L for different temperature.

Temp. / *Time*	*adsorption efficiency %*							
	283K		293K		313K		333K	
	CNT	Non-active carbon	CNT	Non-active carbon	CNT	Non-active carbon	CNT	Non-active carbon
0	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
10	90.74	3.88	90.33	2.87	89.67	1.68	84.55	0.49
20	91.53	6.42	90.74	5.92	90.51	4.90	87.67	1.34
30	93.26	7.10	92.09	6.42	91.67	5.58	89.81	1.68
40	93.95	8.12	93.63	7.78	93.44	6.42	92.74	1.85
50	96.33	12.45	95.72	11.00	95.40	8.80	95.07	2.19
60	98.14	17.78	97.67	14.90	96.65	9.14	96.33	2.53
70	99.02	24.90	98.56	21.34	97.49	16.59	97.02	2.70
80	99.63	34.90	99.44	32.36	98.05	17.44	97.58	3.03
90	99.91	35.24	99.81	32.53	99.63	17.44	97.86	3.54
100	100.00	36.09	100.00	32.53	100.00	17.61	99.21	4.73
110	100.00	36.09	100.00	32.70	100.00	18.29	100.00	5.07
120	100.00	36.09	100.00	32.70	100.00	18.29	100.00	5.41

Table 4: The adsorption percentage of benzene at concentration 300 mg/L for different temperatures.

Temp. / *Time*	*adsorption efficiency %*							
	283K		293K		313K		333K	
	CNT	Non-active carbon	CNT	Non-active carbon	CNT	Non-active carbon	CNT	Non-active carbon
0	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
10	95.03	69.46	88.97	69.29	87.24	65.62	81.52	62.09
20	95.73	70.96	91.49	70.03	90.18	68.78	85.58	66.75
30	95.86	71.13	92.03	70.65	91.52	70.51	87.06	69.69
40	95.97	74.43	93.00	71.44	92.49	71.33	88.49	71.01
50	97.73	74.74	94.18	71.86	93.49	71.47	90.27	71.35
60	97.93	75.17	95.03	72.71	94.12	72.20	90.85	71.75
70	98.17	76.10	95.79	73.02	94.49	72.77	92.24	72.00
80	98.42	76.38	96.36	73.36	95.30	72.79	93.30	72.65
90	98.55	76.58	98.05	73.50	96.36	72.91	94.76	72.94
100	98.79	76.58	98.19	73.56	97.45	73.05	95.64	72.99
110	98.98	76.58	98.33	73.61	97.89	73.08	97.79	73.02
120	99.09	76.64	98.45	73.61	98.17	73.10	98.11	73.05

3.2. Thermodynamic parameters

The thermodynamic parameters ΔG^o, ΔS^o, and ΔH^o for adsorption processes are determined by using following equations [11].

$$\Delta G^o = -RT \ln K \quad (6)$$

Where K is the thermodynamic equilibrium constant. The effect of temperature on thermodynamic constant is determined by:

$$\text{Log } K = \Delta S^o/ 2.303\ R - \Delta H^o/2.303\ RT \quad (7)$$

Where ΔG^o is the free energy change (kJ/mol); R is the universal constant (8.314 J/mol K) and T the absolute temperature (K); ΔH^o change in enthalpy; ΔS^o is the change in entropy. The ΔH^o and ΔS^o values were calculated from slope and intercept of the linear plot, of log K vs. 1/T as shown in Fig. (3). The corresponding values of thermodynamic parameters are presented in Table 5. The negative values of ΔG^o indicate that the benzene adsorption process is spontaneous and feasible. The negative value of ΔH^o shown the adsorption process is exothermic in nature. The negative ΔS^o indicated the decrease in randomness at the solid-liquid interface during adsorption of benzene on carbon.

Table 5: Thermodynamic functions of benzene adsorption process.

	Samples	Initial Conc.(mg/l)	T(K)	K	ΔH^o kJ.mol^{-1}	ΔS^o J.mol^{-1}.k^{-1}	ΔG^o kJ.mol^{-1}
	CNT	200	283	572.066	-33.148	-64.343	-14.939
		200	293	270.396		-66.577	-13.641
		200	313	99.770		-67.636	-11.978
		200	333	69.497		-64.282	-11.742
	Non-active carbon	200	283	2.065	-2.580	-3.088	-1.706
		200	293	2.002		-3.037	-1.690
		200	313	1.906		-2.881	-1.678
		200	333	1.744		-3.123	-1.540

3.3. Adsorption kinetics

The adsorption data of benzene by CNT was fitted through kinetic model including pseudo-first order kinetic [12] as shown in following equation.

$$\text{Ln } (q_e - q_t) = \ln q_e - k_1 t \quad (8)$$

Where q_e (mg/g) is the amount of benzene at equilibrium time while q_t (mg/g) is the amount of benzene at any time (t), k (min^{-1}) is the pseudo-first order rate constant k and q_e were determined from the slope and intercept of the linear plot of ln (q_e-q_t) against t, respectively Fig. 4.

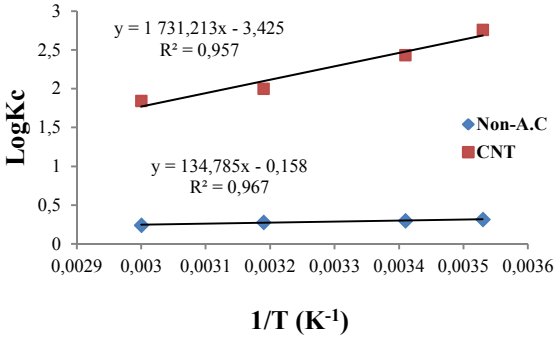

Fig. 3: The plot of Log Kc vs. 1/T with CNT and Non-active carbon

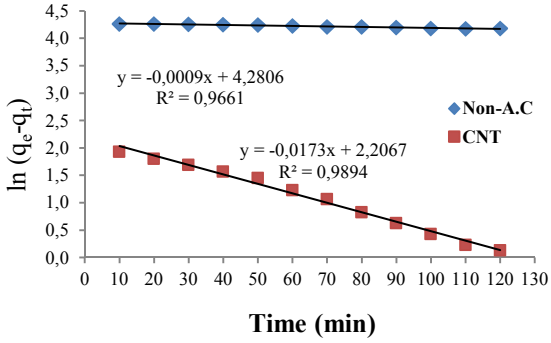

Fig.4: Relationship between Ln (Ce) and Time for benzene solutions at temperature of 283K and Conc. (200mg/L) for CNT and Non- active carbon.

4. Conclusions

The addition of CNT powder as an absorbent to a sample of benzene reduces, the concentration dramatically. About 100% reduction in concentration of benzene was obtained. The adsorption process of CNT powder fit the Langmuir, Freundlich and Temkin isotherm equilibrium adsorption models.

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