Adsorption Kinetics and Thermodynamics Characteristics of Expanded Graphite for Polyethylene Glycol

XIU-YAN PANG

College of Chemistry and Environmental Science,
Hebei University, Baoding 071002 People's Republic of China.

pxy833@163.com

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Abstract: In the present study, expanded graphite (EG) was prepared with 50 mesh crude graphite through chemical oxidation and its adsorption kinetics and thermodynamic characteristics for polyethylene glycol (PEG) with different molecular weight (MW) in aqueous solution was investigated. We studied the influence of initial PEG concentration, temperature, pH and ionic strength on adsorption capacity. Langmuir constants and Gibbs free energy change (ΔG°) were calculated according to experimental data, respectively. Thermodynamic study indicates that all the equilibrium adsorbance increase with the rise in ionic strength. However, solution acidity does not have an obvious effect. Adsorption of EG for PEG with different MW is all types and PEG molecule lies flat on EG surface. Adsorption processes are all spontaneous. Kinetic studies show that the kinetic data can be delineated by pseudo second-order kinetic model. Second-order rate constants and the initial adsorption rate rise with the increasing of temperature and half-adsorption time decreases with the increasing of temperature. The adsorption activation energy of each PEG is less than 30 kJ mol⁻¹, physical adsorption is the major mode of the overall adsorption process.

Keywords: Expanded graphite, Polyethylene glycol, Adsorption kinetics, Adsorption thermodynamics, Ionic strength.

Introduction

Polyethylene glycol (PEG) is widely applied in industries, such as medicament, metal forming, cosmetics and food. However, the wasted medicament solution may become one of the major wastewater sources in industry because the main components of the some minor additives such as PEG, which is a neutral surfactant and acts as a drug stabilizer¹. The principal treatment methods of PEG wastewater are biodegradation², chemical oxidation³, and adsorption treatment such as adsorption into activated carbon^{4,5} or hydrophobic zeolite⁶.

In wastewater treatment, it is well known that adsorption process has been considered available method for eliminating organic pollutants. In the adsorption of PEG with active carbon as adsorbent, Zhao *et al.*¹ reported the adsorbed molecules lay flat on active carbon surface and isotherms are all Langmuir type. Chang *et al.*^{5,6} indicated a high adsorption capacity of 303 mg·g⁻¹ for PEG with an average MW of 6000 from copper electroplating solutions at 288-313 K. While a long equilibrium period of 14 days needed.

Activated carbons are the most widely used adsorbent in the removal or recovery of organic compounds from liquid-phase streams due to their large surface area and nearly non polar surface⁷, they still present some disadvantages such as flammability, difficult to regenerate high-boiling organics and promoting polymerization of some polymerizable mixtures.

Expanded graphite (EG) is a kind of new adsorbent; it can be prepared with chemical method or electrical chemical method. The pore in EG ranges from several nm to hundreds μm and it can be described using a 4-level model⁸. EG has been attracting attentions of scientists and engineers as an absorbent with a high adsorption capacity for organic materials, such as heavy oil and biomedical molecules⁹⁻¹⁴. The research group of Pang has studied the adsorption capacities of EG for oil, dyes, aromatic sulfonates¹⁵⁻¹⁷, results indicate EG show definite adsorption capability for these organic substances, especially for oils. Both adsorbate molecular weight and molecular structure affected sorption type, saturation adsorbance.

Contrast to the adsorption on activated carbon, basic study of PEG on EG is scarce. Therefore, aim of this work is to study the adsorption equilibrium and adsorption kinetics of EG for PEG with different MW in water solution and do further evaluation of applicability of common isotherm models (*i.e.*, Langmuir and Freundlich) and pseudo-second-order rate model.

Experimental

EG is prepared according to literature¹⁸ and its pore distribution was detected by with Auto Pore II 9220 Mercury Porosimeter (Micromeritics Inc. USA) under the condition of 0.58~1301PSIA. Results given in Table 1 show pores in EG are mainly micron pore and the detected total pore area appear too higher than that of BET method¹⁹.

Table 1. Structural parameter of EG

Bulk density, g/mL	Total pore area, m ² /g	Total volume, cm ³ /g	Distribution of pore volume, mL/g			
			~1037 nm	1037~ 10072, nm	10072~ 112689, nm	112689~ 313584, nm
0.0308	1044.99	30.11	1.3693	7.4544	15.8525	3.6583

Adsorbates characteristic

Adsorbates used in experiment were PEG with different MW of 1000, 4000, 10000, 20000, respectively. Simulated PEG wastewaters were prepared by dissolving PEG in distilled deionized water at various concentrations. In quantitative analysis^{20,21}, Dragendoff was used as colored reagent of PEG and absorbance of the colored complex (color reaction lasted 10 min) was detected with T6 New Century UV spectrophotometry (Puxi Tongyong Instrument Limited Company of Beijing). Absorbance values were recorded at the wavelength for maximum absorbance (λ_{max}) (as listed in Table 2) and its solution was initially calibrated for concentration in terms of absorbance units.

Table 2. MW and maximum absorbance wavelength of PEG

PEG	1000	4000	10000	20000
λ_{max} , nm	512	508	512	510

Static adsorption of PEG

0.20 g of EG is mixed in different conical glass flasks with 100.0 mL solution at the desired PEG concentration, pH and ionic strength. pH was adjusted with dilute HCl or NaOH solutions and ionic strength was adjusted with NaCl or Na₂SO₄. Mass of EG to volume of solution was standardized as $M/V = 0.200 / 0.1 = 2.0$ g/L. PEG concentration changes were recorded with spectrophotometer at $\lambda_{\max}$ and adsorbance was determined according to equation (1):

$$q_e = V(C_0 - C_e) / M \quad (1)$$

q_e Equilibrium adsorbance of adsorbate on EG; mg/g; C_0 Initial concentration of PEG in solution; mg/L; C_e Equilibrium concentration of PEG in solution; mg/L; V Volume of solution; L; M Mass of EG; g.

Adsorption kinetics of PEG

Adsorption kinetics experiments were carried out using a HZS-D shaking water bath (Donglian Haerbin, China) with a shaking speed of 100 rpm/min. A series of desired PEG concentration and fixed 100.0 mL were placed in vessels, where they were brought into contact with EG at 5 °C, 25 °C and 45 °C, respectively. Amount of PEG captured by EG at different time was determined as equation (2):

$$q_t = V(C_0 - C_t) / M \quad (2)$$

q_t Accumulative adsorbance of adsorbate on EG at the moment of t ; mg/g; C_t Concentration of PEG in solution at the moment of t ; mg/L;

Results and Discussion

Investigation of adsorption isotherm and thermodynamic parameters

Static adsorption capacities of EG for PEG (1000, 4000, 10000, 20000) were measured. Figure 1 illustrates a typical I type isotherm just as the adsorption of linear herring sperm DNA on EG⁹. The planar structure and large molecules of PEG might form certain kinds of conformation on the surface of EG, which might reduce the adsorbed sites and make the further adsorption difficult. As shown in Figure 1(b), adsorbance decreases with the increase of PEG MW. Similar result was obtained as the adsorption of active carbon for PEG⁴. But adsorption capability of EG is lower than that of active carbon, the results testify the sieve effect.

Figure 1. Adsorption isotherm of PEG (1000, 4000, 10000, 20000) at 25 °C, (a) Adsorbance is defined as mg/g; (b) Adsorbance is defined as mmol/g

A considerable number of studies have been made on the polymer adsorption theory, providing a description of the polymer concentration and possible conformation in the interfacial region. For the adsorption of PEG on EG, Langmuir and Freundlich isotherm equations (3) and (4) were used to treat the isotherm data. The molecule area (a) of PEG was calculated according to q_0 and total pore area as shown in Table 3.

$$\text{Langmuir equation}^{22}: 1/q_e = 1/q_0 + A/(q_0 \times C_e) \quad (3)$$

$$\text{Freundlich equation: } \ln q_e = \ln K_F + (1/n) \ln C_e \quad (4)$$

q_0 Maximum adsorption amount of PEG in forming complete monolayer coverage on EG pore surface; mg/g, A Equilibrium concentration of PEG corresponding to half saturation adsorbance; mg/mL, K_F Freundlich equation constant; $1/n$ adsorption intensity for Freundlich equation.

Table 3. Langmuir and Freundlich isotherm constants at 25 °C

MW	Langmuir			Freundlich			
	q_0	a	A	r	K_F	$1/n$	r
	mmol/g	nm ² /molecule	mmol/mL				
1000	0.045	38.53	0.0837	0.9991	1.0945	0.4538	0.9846
4000	0.012	144.5	0.0341	0.9995	11.1799	0.2078	0.9525
10000	0.005	361.2	0.00988	0.9975	6.9114	0.2786	0.9567
20000	0.001	1445	0.00628	0.9941	2.60833	0.4314	0.9056

^aArea of PEG molecule, (nm)²/molecule; r Linear related coefficient

As shown in Table 3, Langmuir isotherm gives a better fit than Freundlich isotherm. It's just the same as the adsorption of active carbon for PEG. There is an almost linear relationship between PEG MW and PEG molecule area (Figure 2). Results suggest PEG molecule lies flat on the EG surface⁴. PEG with high MW has a small A and strong appetency with EG.

Figure 2. Relationship between PEG MW and molecule area

At the same time, adsorption free energy change (ΔG°) of the adsorption process was calculated according to equation (5)²³, negative ΔG° (Table 4) indicates that adsorption of these reference compounds on EG are all spontaneous.

$$\Delta G^\circ = \Delta RT \ln b \quad (5)$$

b Langmuire equation constant; ΔG° Free energy change in the adsorption; kJ/mol.

Table 4. Adsorption constants and thermodynamic parameter of PEG

PEG	1000	4000	10000	20000
ΔG° kJ/mol	-4.510	-4.853	-5.639	-3.74

Influence of ion strength on adsorption capacity

Influence of ion strength on adsorption capacity is investigated using a 100 mg/L PEG (10000, 20000) solutions which contain NaCl or Na₂SO₄ with concentration ranging from 0 to 50 mg/L. The results are shown in Figure 3. It briefly indicates that the presence of NaCl, Na₂SO₄ can improve the adsorption capacity of EG for PEG and the influence of Na₂SO₄ is more obvious than that of NaCl. The results may be caused by the following reasons: the electrostatic interaction between PEG and adsorbent decreased with the increase in ionic strength for the suppression of the electric double layer²⁴ and hydrophobic attraction of PEG increases due to the “salting-out” effect.

Figure 3. Influence of ion strength on adsorption capacity of EG for PEG (10000) (a), PEG (20000) (b) at 25 °C

Influence of pH on absorbency and adsorption capacity

pH of solution is adjusted with HCl or NaOH. Investigation results show pH has no obvious influence on both PEG absorbency and adsorbance.

Adsorption kinetic

Equilibrium time

Influence of PEG (4000, 10000, 20000) concentration and temperature on adsorption equilibrium time was detected and shown in Figure 4-6. Results suggest that adsorbance is the function of PEG concentration, temperature and adsorption time. Adsorption rate increases with the increasing of temperature, which might be caused by the change of solution viscosity under different temperature. In kinetic experiment, different adsorption equilibrium times were used for different PEG under different temperature.

Figure 4. Influence of PEG (4000) concentration and temperature on adsorption kinetics. (a) 50 mg/L, (b) 200 mg/L, (c) 500 mg/L; (■) 5 °C, (●) 25 °C, (▲) 45 °C

Figure 5. Influence of PEG (10000) concentration and temperature on adsorption kinetics. (a) 50 mg/L, (b) 200 mg/L, (c) 500 mg/L; (■) 5 °C, (●) 25 °C, (▲) 45 °C

Figure 6. Influence of PEG (20000) concentration and temperature on adsorption kinetics. (a) 300 mg/L, (b) 500 mg/L, (c) 700 mg/L; (■) 5 °C, (●) 25 °C, (▲) 45 °C

Adsorption kinetic models

Both pseudo first and second order adsorption models were used to describe the adsorption kinetics data²⁵. In both models, all the steps of adsorption such as external diffusion, internal diffusion and adsorption were lumped together and it is assumed that the difference between the average solid phase concentration and the equilibrium concentration is the driving force for adsorption and the complete adsorption rate is proportional to either the driving force (as in the pseudo first order equation) or the square of the driving force (as in the pseudo second order equation).

$$\text{First-order model: } \ln(q_e - q) = \ln q_e - kt \quad (6)$$

$$\text{Second order model: } t/q = 1/(k q_e^2) + t/q_e \quad (7)$$

k Adsorption rate constant (min^{-1} for first order adsorption, $\text{g} \cdot \text{mg}^{-1} \cdot \text{min}^{-1}$ for second-order adsorption); t Adsorption time (min)

Since q reaches q_e at equilibrium, q values smaller than $0.9q_e$ were used for analysis. Plots of $\ln(q_e - q)$ versus t and t/q versus t were used to test the first and second order models and the fitting results are given in Table 5-7. As for the line curve fit, second order model gives higher correlation coefficients than first order model. And $q_{e,\text{cal}}$ corresponding to second order model, agrees more well with experimental data except at 5°C or lower PEG concentration. Thus, second order model is more suitable to describe the adsorption kinetics data. Similar results were observed in the adsorption of EG for Basic fuchsin and Auramine lake yellow O²⁶ and zeolite for PEG¹³.

Table 5. Comparison of the adsorption kinetic models of PEG (4000) on EG

C_0 , mg/L	T , $^\circ\text{C}$	$q_{e,\text{exp}}$, mg/g	First order			Second order		
			$q_{e,\text{cal}}$, mg/g	k , 1/min	r	$q_{e,\text{cal}}$, mg/g	k , g/mg·min	r
50	5	8.046	7.51 ±1.04	0.039±0.020	-0.996	11.92±0.332	0.002±6.89E-05	0.999
	25	8.330	8.222±1.057	0.051±0.002	-0.995	10.99±0.199	0.004±7.93E-05	0.999
	45	11.01	8.483±1.247	0.065±0.016	-0.943	13.95±0.418	0.006±1.76E-04	0.998
200	5	20.61	28.20±1.128	0.047±0.005	-0.982	26.59±2.38	0.001±1.33E-04	0.996
	25	31.62	39.67±1.112	0.072±0.005	-0.990	37.38±0.974	0.002±4.55E-05	0.999
	45	39.24	32.55±1.201	0.059±0.006	-0.974	44.66±1.300	0.002±7.15E-05	0.999
500	5	21.46	6.866±1.042	0.072 0.002	-0.999	22.05±3.2	0.023±8.55E-05	0.999
	25	33.32	8.217±1.054	0.059±0.003	-0.993	32.76±2.03	0.024±4.34E-05	1.000
	45	37.83	20.38±1.227	0.136±0.009	-0.988	40.65±0.361	0.010±9.24E-05	1.000

Table 6. Comparison of the adsorption kinetic models of PEG (10000) on EG

C_0 , mg/L	T , $^\circ\text{C}$	$q_{e,\text{exp}}$, mg/g	First order			Second order		
			$q_{e,\text{cal}}$, mg/g	k , 1/min	r	$q_{e,\text{cal}}$, mg/g	k , g/(mg·min)	r
50	5	6.71	7.530 ±1.11	0.065±0.010	-0.986	7.664 ±0.787	0.007±7.15E-04	0.990
	25	14.20	12.41 ±1.09	0.039±0.003	-0.984	12.80±0.022	0.012±2.07E-05	1
	45	14.84	7.943±1.319	0.089±0.020	-0.952	15.92±0.832	0.017±0.001	0.997
200	5	11.28	10.49±1.035	0.044±0.002	-0.997	12.92±0.299	0.004±0.261	0.999
	25	17.56	17.38±1.038	0.033±0.002	-0.993	16.46±0.296	0.003±0.421	0.991
	45	35.18	24.59±1.203	0.121±0.014	-0.988	39.11±0.692	0.007±1.15E-04	0.999
500	5	21.41	20.59±1.175	0.085±0.010	-0.983	25.08±1.571	0.005±1.313	0.995
	25	39.97	15.90±1.072	0.090±0.004	-0.989	39.15±1.813	0.008±4.93E-04	0.997
	45	52.10	9.718±1.093	0.070±0.008	-0.998	47.96±2.286	0.0185±0.001	0.999

Table 7. Comparison of the adsorption kinetic models of PEG (20000) on EG

C_0 , mg/L	T, °C	$q_{e,exp}$ mg/g	First order			Second order		
			$q_{e,cal}$ (mg/g)	k, 1/min	r	$q_{e,cal}$ (mg/g)	k (g/(mg·min))	r
300	5	12.96	10.39±1.018	0.0259±0.001	-0.997	13.73±0.662	0.004±1.75E-04	0.993
	25	14.75	9.700±1.066	0.057±0.004	-0.993	16.77±0.942	0.007±3.81E-04	0.997
	45	26.14	17.78±1.149	0.090 ±0.013	-0.990	29.16±0.537	0.008±1.51E-04	0.999
500	5	10.78	9.863±1.044	0.045 ±0.001	-0.991	13.56±0.294	0.004±8.07E-05	1
	25	13.25	10.43±1.082	0.068 ±0.004	-0.993	14.99±0.133	0.008±7.51E-05	1
	45	29.64	10.10±1.041	0.051 ±0.003	-0.997	29.45±0.380	0.013±0.382081	1
700	5	16.31	14.22±1.046	0.034 ±0.002	-0.994	18.93±0.466	0.003±6.58E-05	0.999
	25	28.15	20.98±1.112	0.047±0.006	0.989	24.50±1.08	0.007±1.02E-03	0.999
	45	35.75	15.75±1.069	0.045 ±0.003	-0.989	33.81±0.184	0.012±6.56E-05	1

Based on the second order model, initial adsorption rate and half-adsorption time were estimated according to equations (8) and (9):

$$u=kq_e^2 \quad (8)$$

$$t_{1/2}=1/(kq_e) \quad (9)$$

u Initial adsorption rate, mg/(g·min); $t_{1/2}$ half-adsorption time (min).

Half-adsorption time $t_{1/2}$ is often used as a measure of the adsorption rate. As shown in Table 8-10, u is found to increase with the increase of initial PEG (4000, 10000, 20000) concentration and temperature and $t_{1/2}$ decrease with the increase of temperature. Second-order rate constants are used to estimate activation energy of PEG adsorption on EG using Arrhenius equation²⁷:

$$\ln k = \ln A - E_a/(RT) \quad (10)$$

A The re-exponential factor, (g·mg⁻¹·min⁻¹); E_a Activation energy, (kJ/mol)

Slope of $\ln k$ versus $1/T$ is used to evaluate E_a , which is found less than 40.0 kJ·mol⁻¹ (Table 8-10). So, the adsorption is mainly physical adsorption. Similar results were observed in adsorption of EG for dyes of Basic fuchsine and Auramine lake yellow O²⁶.

Table 8. Kinetic parameters for the second order adsorption model of PEG (4000)

C_0 , mg/g	T, °C	u mg/g·min	$t_{1/2}$, min	E_a , kJ/mol	r
50	5	0.351	50.316	16.08±2.31	0.990
	25	0.528	27.442		
	45	1.139	15.511		
200	5	1.065	32.190	9.11±2.42	0.967
	25	2.441	18.108		
	45	4.911	10.351		
500	5	2.345	27.100	4.11±0.00	1.0
	25	3.870	18.301		
	45	12.241	9.024		

Table 9. Kinetic parameters for the second order adsorption model of PEG (10000)

C ₀ , mg/g	T, °C	u, mg/g·min	t _{1/2} , min	E _a , kJ/mol	r
50	5	0.409	21.406	16.92±0.95	0.998
	25	1.945	5.926		
	45	4.393	3.888		
200	5	0.749	19.764	7.0±1.47	0.979
	25	0.891	9.690		
	45	9.980	4.356		
500	5	1.159	9.905	24.02±4.97	0.979
	25	2.926	2.993		
	45	23.770	1.040		

Table 10. Kinetic parameters for the second order adsorption model of PEG (20000)

C ₀ , mg/g	T, °C	u, mg/g·min	t _{1/2} , min	E _a , kJ/mol	r
300	5	0.683	21.293	15.20±4.02	0.967
	25	1.908	9.999		
	45	6.976	4.664		
500	5	0.701	23.340	22.03±2.61	0.993
	25	1.906	8.900		
	45	11.128	2.629		
700	5	0.958	22.936	28.02±3.95	0.990
	25	4.331	5.657		
	45	13.759	2.324		

Internal diffusion analysis

Adsorption process on a porous adsorbent generally involves three stages: (i) external diffusion; (ii) internal diffusion (or intra-particle diffusion); (iii) actual adsorption²³. Quantitative treatment of experimental data may reveal the predominant role of a particular step among the three that actually governs the adsorption rate. Adsorption step is usually very fast for the adsorption of organic compounds on porous adsorbents in comparison to the external or internal diffusion step²⁷ and it is known that the adsorption equilibrium is reached within several minutes in the absence of internal diffusion²⁸. Thus, the long adsorption equilibrium time of PEG on EG (40~180 min corresponding to adsorption temperature of 5~45 °C) suggests that the internal diffusion may dominate the overall adsorption kinetics.

To provide definite information on the rate-limiting step, an internal diffusion model based on Fick's second law is used to test if the internal diffusion step is the rate-limiting step²³:

$$q = k_{id}t^{1/2} \quad (11)$$

k_{id} Internal diffusion constant, mg/(g·min^{1/2}).

According to the internal diffusion model, a plot of q versus $t^{1/2}$ should give a straight line with a slope k_{id} and an intercept of zero if the adsorption is limited by the internal diffusion process. The relationships between q of PEG (4000, 10000, 20000) and $t^{1/2}$ at different temperature are shown in Figure 7-9. In the range of the tested temperature, a linear relationship between q versus $t^{1/2}$ with a zero intercept is found when the temperature is not high, which suggests internal diffusion step dominates the adsorption process before the equilibrium is reached.

Figure 7. Plot of q vs. $t^{1/2}$ in internal diffusion model of PEG (4000), (a) 50 mg/L, (b) 200 mg/L, (c) 500 mg/L; (■) 5 °C, (●) 25 °C, (▲) 45 °C

Figure 8. Plot of q vs. $t^{1/2}$ in internal diffusion model of PEG (10000). (a) 50 mg/L, (b) 200 mg/L, (c) 500 mg/L; (■) 5 °C, (●) 25 °C, (▲) 45 °C

Figure 9. Plot of q vs. $t^{1/2}$ in internal diffusion model of PEG (20000). (a) 300 mg/L, (b) 500 mg/L, (c) 700 mg/L; (■) 5 °C, (●) 25 °C, (▲) 45 °C

Conclusion

Adsorption of EG for PEG with different MW has been investigated. The results are summarized as follows:

- (1) The same as active carbon, adsorption isotherms of EG for PEG can be well described with Langmuir equation. But the adsorption equilibriums are faster than active carbon, and adsorption capacity of EG is lower than that of active carbon.
- (2) Adsorption of EG for PEG is spontaneous, adsorption isotherm (1000, 4000, 10000, 20000) is type I, and PEG molecule lies flat on EG surface.
- (3) Adsorption kinetics of EG for PEG can be described by pseudo-second-order model equation. Equilibrium time and half-adsorption time $t_{1/2}$ decreases with the increase of temperature. The adsorption belongs to physical adsorption, and internal diffusion is tested to be the rate-limiting step of the complete adsorption process.

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