

Comparative Study of the Adsorption and Desorption Behavior of Single and Multi-Ring Aromatics in Sediment Fractions

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ABSTRACT

The sorption behaviour of benzene, toluene, ethyl benzene, xylene and naphthalene using clay and sand sediments under ambient conditions is examined in this study. Experimental results showed that, the time taken to attain adsorption equilibrium for naphthalene, and BTEX were 28, 30, 30, 32, 28 hrs and 20, 22, 22, 24, 22 hrs while the desorption equilibrium time were 10, 13, 12, 15, 12 hrs and 9, 9, 9, 11, 10 hrs in clay and sand respectively. All of the naphthalene, and BTEX were adsorbed at the different equilibrium times, using clay while the amount of naphthalene and BTEX adsorbed by sand, at different equilibrium times were 117, 121, 127, 123 and 134 mg. Following the results of the adsorption/desorption experiments, quantitative measurements showed that sand exhibited higher affinity for the solute as retained more chemicals (as high as between 58% - 66%) within its pores while nearly all the chemicals adsorbed by the clay were released at the attainment of equilibrium. The implication of this is that occlusion within the sand particles may likely be the resultant effect of continued sand-chemicals contact. The amount of contaminant solute adsorbed and desorbed affirmed that clay has a better capacity to retain naphthalene and BTEX than sand and this may not be unrelated to its large surface area, high porosity and higher hydraulic conductivity for the solutes arising from its good binding sites (small pore sizes) that tend to hold the adsorbates to its particles.

Keywords: Equilibrium; Kinetic Models; Pollutants; Pore Spaces; Surface Area; Permeability

1. Introduction

The major environmental concern in urban and industrial areas can be attributed to the increasing level of pollution particularly by substances of organic origin. Various toxic chemicals such as polycyclic aromatic hydrocarbons (PAHs): naphthalene, anthracene, benzo(a)pyrene, phenanthrene, benzene, toluene, ethyl benzene and xylene (BTEX), heavy metals and dyes are continuously discharged into the environment as industrial waste, causing water, air and soil pollutions. These chemicals due to their recalcitrant persistent nature have relatively low solubilities in water, but are highly lipophilic. The four and more condensed aromatic rings are considered to be more dangerous than the two and three rings PAHs [1-3]. The presence of these compounds which are listed as priority pollutants [4-7] in the environment is of considerable public health and ecological concern due to their toxicity to a wide range of biological systems.

These effects apart from the degradation of the ecosystem, also results in commodity loss, loss to the communities and economic loss due to spill clean up cost.

Studies have shown that solute transport with linear equilibrium is an integral component of the degradation and/or mineralization of these toxic chemicals [8-12]. Contaminant transport is significantly viewed from two possible scenarios; fast sorption/desorption and slow sorption/desorption. Sorption tends to separate the direct contact between microorganisms and contaminants, which is necessary for biodegradation to occur. The practical effect of the adsorption and desorption rates, is that it controls the overall reaction rate of degradation process. They are one of the primary factors which affect availability, mobility and toxicity of contaminants in the soil [3-17]. They determine the measured concentration and the mechanism of distributing the contaminants into surfaces and into pores of individual soil particles [18-22] and are thus counteractive to efficient biodegradation.

Adsorption is a physical separation process in which

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certain compounds of a fluid phase are transferred to the surface of a solid adsorbent [23]. The separation is dependent on one component in a mixture being more readily adsorbed than the other components. The adsorption process takes place in three steps: macrotransport, microtransport and sorption. Macrotransport involves the movement of the organic material through the water to the liquid-solid interface by advection and diffusion. Microtransport involves the diffusion of the organic material through the macropore system of the soil particle to the adsorption sites in the micropores and submicropores of the soil particle. Adsorption results from the influence of Van der Waals forces which is essentially physical in nature. Due to the fact that the forces are not strong, the adsorption may be easily reversed. However, in some systems, additional forces bind adsorbed molecules to the solid surface. These are chemical in nature involving the exchange or sharing of electrons, or possibly molecules forming atoms or radicals. In such cases, the term chemisorption is used to describe the phenomenon [24]. This is less easily reversed than physical adsorption, and regeneration may be a problem.

The present study seeks to compare the processes of adsorption and desorption of naphthalene and BTEX in clay and sand as sediments with a view to determining the capacity and extent to which each sediment retains solutes at equilibrium as these primary processes form the basis for their availability for microbial uptake and mineralization. This is significant because in the long run results from the study will serve as a benchmark for the removal of these recalcitrant compounds which compromise the integrity of the environment. Suitable kinetic models which best describes the sorption-desorption mechanisms of the organic chemicals on the sediments will also be determined.

2. Materials and Methods

2.1. Materials

The sediments used in this study were obtained from Ikpoba River, Benin City, Edo State, Nigeria. On collection, the sediments were stored in a black polythene bag. Pretreatment of the sediments was carried out via removal of stones and other heavy particles. A 220 μm mesh was used to remove the large non clay fractions from the clay and a 2.5 mm mesh was used to sieve the sand. Finally, they were then dried overnight at 383 K in a vacuum oven and stored in an air-tight container covered with a black polythene bag prior to the adsorption and desorption experiments described by [14].

The adsorbates, naphthalene, benzene, toluene, ethyl benzene and xylene were obtained from an auto-mechanic workshop at Ojota, Lagos state, Nigeria while the distilled water used for sample preparation, dilution and

solution was obtained from the Department of Chemical Engineering, University of Benin, Benin City, Edo State, Nigeria.

2.2. Methods

2.2.1. Adsorption-Desorption Kinetics Experiments

The rates of adsorption by the sediments (clay and sand) were determined from the uptake levels of PAHs and BTEX from aqueous solution in batch experiments before and after contact until adsorption equilibrium was attained in the clay and sand. 100 g of each of the sediments (clay and sand) were mixed with 500 ml of aqueous solution of the adsorbates at room temperature following the procedure of Owabor *et al.* [25]. The slurry suspensions were sampled at intervals of 2 hours and UV spectrophotometer was used for sample analysis.

2.2.2. Adsorption Kinetics Models

The adsorption kinetics which describes the mechanism of the adsorption process in a given system were determined in this study using pseudo-first order model as defined by Lagergren and described by Chang *et al.* and Sivaraj *et al.* [14,25], pseudo-second order model [14], Elovich Model [26], intraparticle diffusion model [27] and the power function technique [28].

3. Results and Discussion

The results of the experimentation and computation analysis of the kinetic modeling of the adsorption of naphthalene, benzene, toluene, ethylbenzene and xylene contaminated clay and sand are presented. The potential for the two sediments to adsorb and subsequently release the solutes for eventual mineralization by microbes have been exploited. The importance of this study was to explore the possible effects of sorption and bioavailability on biodegradation rates and the prediction of risk likely to occur from prolonged exposure of the soil to the contaminant solutes. The information on the mobility and hence availability of the chemicals is crucial in remediation studies as it determines the extent of partitioning and sequestration of the chemicals with sediments. A comparative analysis of the predictions from the kinetic modeling data obtained from experiments provided the basis for the choice of the applicable and suitable mechanisms for describing the adsorption and desorption processes.

3.1. Equilibrium Time for Adsorption

From **Figure 1**, the adsorption and desorption results showed that equilibrium was achieved at 28 and 20 hrs for naphthalene in the clay and sand fractions respectively. Similarly, for benzene, toluene, ethyl benzene and

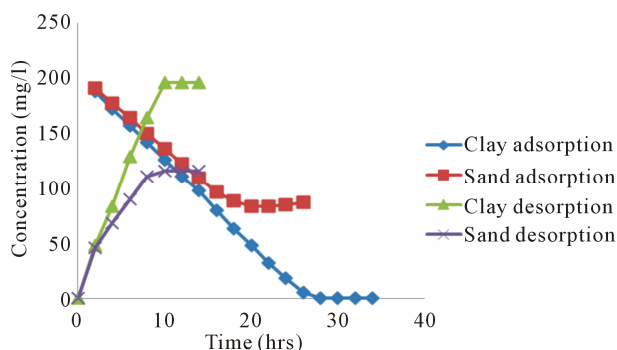


Figure 1. Variation of naphthalene concentration with time for adsorption and desorption.

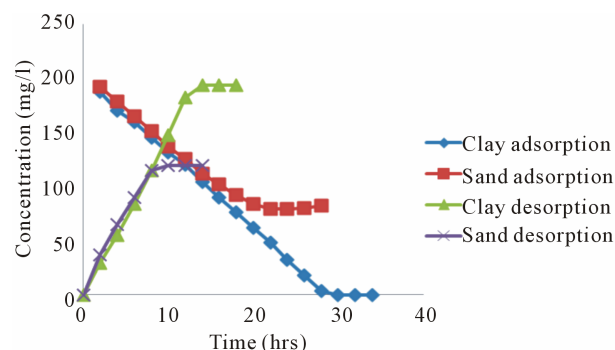


Figure 2. Variation of benzene concentration with time for adsorption and desorption.

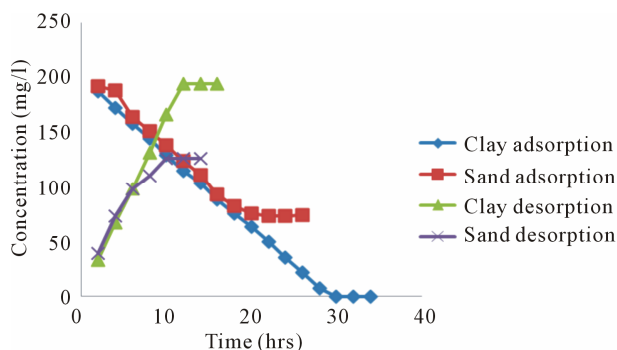


Figure 3. Variation of toluene concentration with time for adsorption and desorption.

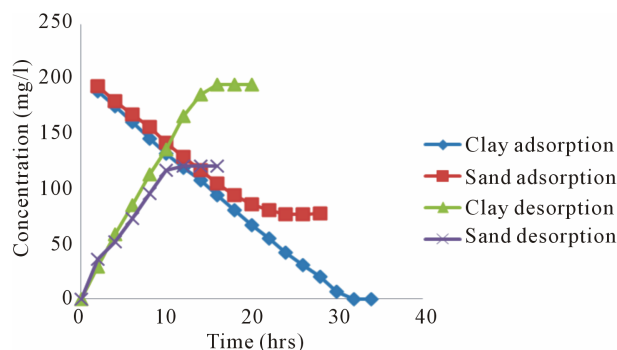


Figure 4. Variation of ethyl benzene concentration with time for adsorption and desorption.

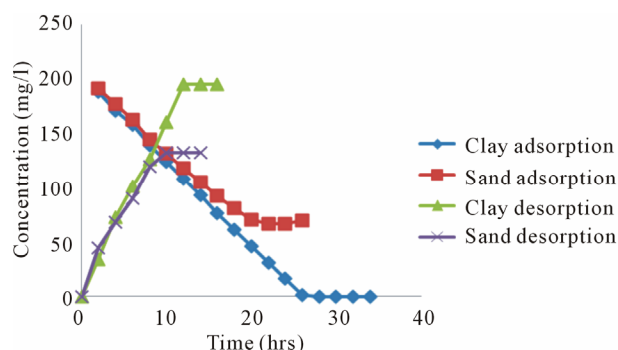


Figure 5. Variation of xylene concentration with time for adsorption and desorption.

xylene, equilibrium was attained at 30, 30, 32 and 28 hrs and 22, 22, 24 and 22 hrs as shown in **Figures 2-5** respectively. From the equilibrium time, sand was observed to reach equilibrium faster than clay for all of contaminant solutes used in this study. This can be attributed to its large pore spaces, intra-porous nature of sand (its pore spaces are connected to one another) and its higher permeability.

However, it is worthy to note that before equilibrium was attained for both clay and sand, the amount of naphthalene adsorbed in the clay sample was 102 mg while in sand, the amount was 91 mg. Similarly, for benzene, toluene, ethyl benzene and xylene using clay, the amount adsorbed were 96, 97, 92 and 107 mg while in sand, the amount adsorbed were 88, 89, 83 and 95 mg. Based on the amount adsorbed given equal time before equilibrium, clay would be described as a better adsorbing agent of naphthalene and BTEX than sand because it holds more of naphthalene, benzene, toluene, ethyl benzene and xylene. This property of clay over sand can be attributed to its larger surface area and higher porosity than sand [29, 30]. Sand has low porosity though not as many pore spaces because its grains are very large such that in a unit of sand, the fraction of soil volume that consists of holes is a lot less than for clay soil. Clay has many small pore spaces in which water containing the contaminant solutes remains clinging to the clay particle surfaces. Porosity is an important consideration when evaluating the potential volume of water or amount of hydrocarbons sediments may contain. Sediments with higher porosity typically have higher hydraulic conductivity, a property of sediments that describes the ease with which water can move through pore spaces.

3.2. Equilibrium Time for Desorption

Desorption equilibrium, for clay sediment, was achieved at 10 hrs with 197 mg of naphthalene desorbed, while using sand, equilibrium was achieved at 9 hrs with 116 mg of naphthalene desorbed. For benzene, toluene, ethyl benzene and xylene, in clay, equilibrium was achieved at

13 hrs with 193 mg of benzene desorbed, 12 hrs with 194 mg of toluene desorbed, 15 hrs with 195 mg of ethyl benzene desorbed and 12 hrs with 195 mg of xylene desorbed respectively. While in sand, equilibrium was achieved at 9 hrs with 119 mg of benzene desorbed, 9 hrs with 126 mg of toluene desorbed 11 hrs with 121 mg of ethyl benzene desorbed and 10 hrs with 132 mg of xylene desorbed respectively.

Laboratory results obtained from this study affirmed that the desorption process was slow for both sediment types. The implication of this is that the organic chemicals are very slowly released for uptake or mineralization. The amount desorbed by the sediments may have been retarded as a result of their hydrophobicity. The solutes have a high affinity to sediments and great tendency to bind with organic carbon, mineral surfaces and interstitial voids within the micropores and submicropores of the sediment fraction [17,22,31]. The observed slowly desorbing fraction can therefore be attributed to the effect of intraorganic matter and hindered pore diffusion mechanisms.

3.3. Mechanism of Adsorption and Desorption Processes

Tables 1-4 show the results of the kinetic modeling of the adsorption and desorption of naphthalene, benzene, toluene, ethyl benzene and xylene onto clay and sand

used in this study. While the value of the reaction rate constant k predicted for the adsorption kinetics by the pseudo-first order and power function technique model equations closely approximated, there were however, wide deviations for the pseudo-second order, Elovich and intra-particle models. In contrast, for the desorption kinetics only the pseudo-first order and Elovich equations gave good fits as the intraparticle and power function equations were characterized by negative rate constants. The pseudo-second order rate constant was found to be ambiguous. However, estimations from the coefficient of regression indicate that the power function model best described the mechanism of adsorption of naphthalene, benzene, toluene, ethyl benzene and xylene while the intra-particle model gave the best description for desorption of naphthalene, benzene, toluene, ethyl benzene and xylene. They closely approximated to unity. Interestingly, the observed good approximations of the correlation coefficients obtained from the power function technique and intra-particle model for the contaminant solutes were consistent for both clay and sand sediments. This further affirms the suitability of the two models.

4. Conclusion

This study showed that adsorption and desorption of PAHs (naphthalene) and BTEX occurred in clay and sand with clay adsorbing more of the naphthalene, ben-

Table 1. Kinetic parameters and correlation coefficient (R^2) values for the adsorption of naphthalene and BTEX using clay sediment.

Kinetic Models	Parameters	Naphthalene	Benzene	Toluene	Ethyl benzene	Xylene
Pseudo-First Order	k_1	0.1210	0.116	0.136	0.095	0.150
	q_e	3.7547	3.8729	7.0781	3.2904	4.8211
	R^2	0.8202	0.7510	0.8440	0.8550	0.6842
Pseudo-Second Order	k_2	4×10^{-5}	9×10^{-5}	1.3×10^{-5}	1.259×10^{-5}	0.00242
	q_e	43.1034	27.7778	23.6407	-714.2857	30.8642
	R^2	0.0348	0.2087	0.3178	0.0002	0.056
Elovich	α	1.2458	1.2933	1.3063	1.3317	1.2430
	β	0.2723	0.2572	0.2581	0.2454	0.2775
	R^2	0.9179	0.8974	0.9075	0.9051	0.9241
Intra-Particle	k_p	0.496	0.483	0.477	0.469	0.496
	C	-0.737	-0.746	-0.723	-0.739	-0.718
	R^2	0.978	0.977	0.983	0.985	0.978
Power Function	ν	1.013	0.981	0.981	1.022	1.009
	k	0.0684	0.0714	0.0684	0.0607	0.0702
	R^2	0.989	0.995	0.996	0.996	0.986

Table 2. Kinetic parameters and correlation coefficient (R^2) values for the adsorption of naphthalene and BTEX using sand sediment.

Kinetic Models	Parameters	Naphthalene	Benzene	Toluene	Ethyl benzene	Xylene
Pseudo-First Order	k_1	0.1730	0.176	0.293	0.18	0.168
	q_e	2.1706	2.6013	1.6023	3.0465	2.7020
	R^2	0.9240	0.914	0.934	0.917	0.909
Pseudo-Second Order	k_2	0.0016	0.0003	0.0033	4.996×10^{-4}	2×10^{-5}
	q_e	6.3776	13.3333	-3.6657	-9.9404	-53.195
	R^2	0.1407	0.0436	0.1661	0.1045	0.0154
Elovich	A	2.419	1.9873	1.7778	1.9066	1.7794
	B	0.2054	0.2106	0.2120	0.2002	0.2325
	R^2	0.6067	0.9534	0.934	0.945	0.9472
Intra-Particle	k_p	0.330	0.327	0.380	0.340	0.379
	C	-0.389	-0.395	-0.543	-0.461	-0.488
	R^2	0.958	0.964	0.920	0.974	0.974
Power Function	V	1.008	1.019	1.176	1.081	1.045
	k	0.056	0.0522	0.0374	0.0428	0.0555
	R^2	0.966	0.968	0.969	0.979	0.979

Table 3. Kinetic parameters and correlation coefficient (R^2) values for the desorption of naphthalene and BTEX using clay sediment.

Kinetic Models	Parameters	Naphthalene	Benzene	Toluene	Ethyl benzene	Xylene
Pseudo-First Order	k_1	0.275	0.207	0.255	0.207	0.202
	q_e	-1.033	-0.882	-0.695	-0.757	0.453
	R^2	0.922	0.852	0.796	0.877	0.893
Pseudo-Second Order	k_2	14.100	6.416	10.576	7.363	9.423
	q_e	0.018	0.035	0.029	0.027	0.025
	R^2	0.660	0.644	0.673	0.607	0.611
Elovich	A	2.558	2.646	2.604	2.688	2.688
	B	1.585	1.610	1.556	1.602	1.722
	R^2	0.937	0.883	0.904	0.912	0.913
Intra-Particle	k_p	-0.387	-0.352	-0.363	-0.327	-0.354
	C	1.300	1.379	1.351	1.335	1.327
	R^2	0.983	0.969	0.975	0.983	0.982
Power Function	V	-1.607	-1.240	-1.363	-1.302	-1.355
	k	0.979	0.977	0.993	1.112	0.989
	R^2	0.677	0.634	0.653	0.641	0.617

Table 4. Kinetic parameters and correlation coefficient (R^2) values for the desorption of naphthalene and BTEX using sand sediment.

Kinetic Models	Parameters	Naphthalene	Benzene	Toluene	Ethyl benzene	Xylene
Pseudo-First Order	k_1	0.361	0.383	0.279	0.298	0.397
	q_e	1.533	-4.200	1.234	4.075	-1.382
	R^2	0.885	0.826	0.992	0.825	0.763
Pseudo-Second Order	k_2	35.270	34.908	33.385	26.800	31.607
	q_e	8.67×10^{-3}	8.67×10^{-3}	8.91×10^{-3}	9.43×10^{-3}	8.82×10^{-3}
	R^2	0.729	0.719	0.669	0.660	0.719
Elovich	α	5.076	4.329	4.237	4.484	4.065
	β	2.862	2.552	2.671	2.778	2.518
	R^2	0.948	0.958	0.974	0.921	0.948
Intra-Particle	k_p	-0.200	-0.234	-0.237	-0.214	-0.241
	C	0.627	0.740	0.740	0.751	0.797
	R^2	0.980	0.981	0.984	0.978	0.978
Power Function	V	-2.192	-1.800	-1.992	-1.626	-1.788
	k	0.600	0.429	0.624	0.549	0.610
	R^2	0.657	0.715	0.644	0.661	0.699

zene, toluene, ethyl benzene and xylene than sand while sand desorbed more of the naphthalene, benzene, toluene, ethyl benzene and xylene than clay. The adsorptive property of clay over sand was attributed to its large surface area, higher porosity and high hydraulic conductivity of the adsorbates while the slow desorption can be attributed to its good binding sites (small pore sizes) that tend to hold the adsorbates to its particles. For both adsorption and desorption, equilibrium was attained faster for sand than for clay as a result of the higher permeability of sand sediment. The power function model gave the best description for the adsorption mechanism of naphthalene, benzene, toluene, ethyl benzene and xylene, while the intraparticle model was most suitable for describing the mechanism of desorption of the contaminant solutes.

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