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Research paper

# Equilibrium and kinetic adsorption of drugs on bentonite: Presence of surface active agents effect

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ABSTRACT

The adsorption of five drugs: promethazine (PM), triflupromazine (TFP), trimethoprim (TM), carbamazepine (CM), and ibuprofen (IBU) onto bentonite (Bent) was studied spectrophotometrically in aqueous solutions and in the presence of different type of surfactants i.e. anionic sodium dodecyl sulfate (SDS) and cationic dodecyl trimethyl ammonium bromide (DTAB) . The adsorption experiments were carried out as a function of time, initial concentration and temperature. Adsorption kinetic data were modeled using the Lagergren first order and the pseudo-second order kinetic equations. Interlayer diffusion graphics were also plotted. The data obtained from all kinetic studies are fitted to the pseudo-second order kinetic equation better than the Lagergren first order kinetic equation, except the adsorption of IBU onto Bent. The Giles isotherms were used to understand the adsorption mechanism of drugs. Isotherms plotted appear to fit L-type according to Giles isotherm classification. The Langmuir and Freundlich isotherms were used to model the equilibrium data obtained. The Langmuir model appears to fit the isotherm data better than the Freundlich model. The adsorption capacity values found for the adsorption of drugs in aqueous solutions followed the order: PM > TFP > TM > CM > IBU. It was observed that the presence of surfactants had significant effects on the adsorption capacities, although it did not affect the equilibrium time of the adsorption processes.

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## 1. Introduction

One of the emerging issues in environmental chemistry in recent years is the occurrence of pharmaceutically active compounds in the aquatic environment and the investigations showed that substances of pharmaceutical origin are often not eliminated during waste water treatment and also not biodegraded in the environment (Heberer, 2002). Pollution by pharmaceuticals can occur in concentrations of parts per billion (ppb), but more often in parts per trillion (ppt) (where 1 ppt equates to 1 ng/L). Although these concentrations are very low and are difficult to detect, they have the potential for environmental effects (Jones et al., 2004).

Pharmaceuticals occur in combination with other compounds in the environment. These compounds are not only other pharmaceuticals. They are also their own metabolites of the pharmaceuticals or other environmental pollutants, e.g., industrial chemicals, pesticides, or personal care products. Thus, possible mixture effects are also important (Escher et al., 2005). Little work has been reported examining environmental interactions with amphiphilic compounds such as surfactants (Hari et al., 2005).

Amphiphiles like surfactants are widely used in pharmaceutical products in order to stabilize emulsions and enhance the delivery of drugs in the body. In addition, surfactants from detergents and other products enter the environment with pharmaceuticals through wastewater discharge. Although amphiphiles found in the environment are present at low concentrations, amphiphiles entering the environment with pharmaceuticals will be present at significantly higher concentrations than the pharmaceuticals themselves (Hari et al., 2005).

The adsorption method becomes an attractive alternative for the waste water treatment, if the adsorbent is inexpensive and does not require an additional pretreatment step (such as activation) before the application (Janos and Smidova, 2005). In addition to being low cost adsorbents, clay minerals are fundamental components in several medicinal products where they are used as excipients and fulfill some technological function. Clay minerals also have potential to be used in the development of new drug delivery systems (Özcan and Özcan, 2004; Viseras et al., 2010).

In this context, the objective of this study is to investigate the adsorption kinetics of five different drugs selected onto bentonite and disclose the effects of the temperature and the presence of surface active agents on this adsorption process.

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**Table 1**
Textural characterization of Bent.

| $S_{BET}$ ($m^2/g$) | $V_{TOTAL}$ ($cm^3/g$) | $V_{MICROPORES}$ ($cm^3/g$) | $V_{MESOPORES}$ ($cm^3/g$) |
|---|---|---|---|
| 23 | 0.062 | ≈0.000 | 0.029 |

## 2. Materials

Bentonite (Bent) was obtained from Sigma and used as received without pretreatment, with the exception of washing to eliminate soluble impurities. Bent was washed 3 times with 1 L distilled water for each 10 g of adsorbent using magnetic stirrer and dried at 378 K for 24 hours to remove moisture. The $pH_{PZC}$ value was determined as 8.1 by using mass-titration method (Noh and Schwarz, 1989). Textural characterization (Table 1) was carried out by measuring the $N_2$ adsorption isotherms at 77 K in an automatic apparatus (Micromeritics ASAP 2010 M). Before the experiments, the samples were outgassed under vacuum at 393 K overnight. The isotherms obtained were used to calculate the specific surface area ($S_{BET}$) and pore size distribution. Total pore volume ($V_{TOTAL}$) is evaluated at relative pressure 0.99 and volume of micropores ($V_{MICROPORES}$) and mesopores ($V_{MESOPORES}$) is evaluated by density functional theory method. Chemical analysis of Bent was done by Istanbul University Advanced Analysis Laboratory. The main chemical components of the Bent were (mass, %): $SiO_2$ = 65.8, $Al_2O_3$

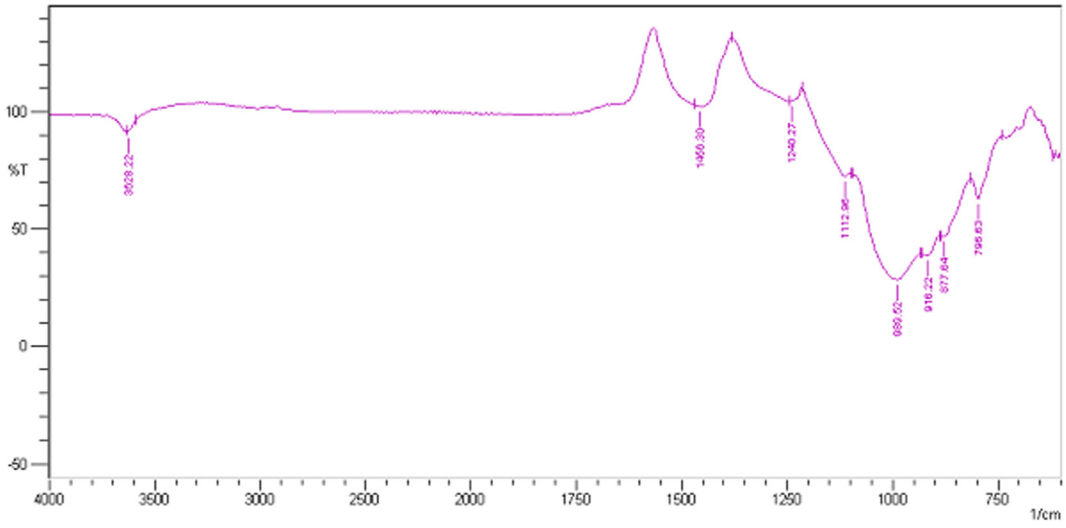


**Fig. 1.** FTIR analysis of Bent.

**Table 2**
Chemical structures and characteristics of drugs used.

| Molecular structure | Molecular formula | Molecular weight (g/mol) | Water solubility at 298 K | p$K_a$ at 298 K | $\lambda_{max}$ (nm) |
|---|---|---|---|---|---|
| PM | $C_{17}H_{20}N_2S \cdot HCl$ | 320.88 | Very soluble[a] | 9.1[b] | 298 |
| TFP | $C_{18}H_{19}F_3N_2S \cdot HCl$ | 388.88 | Soluble[c] | 9.2[d] | 306 |
| TM | $C_{14}H_{18}N_4O_3$ | 290.32 | 400 mg/L[e] | 7.3[f] | 278 |
| CM | $C_{15}H_{12}N_2O$ | 236.27 | 170 mg/L[g] | 7.0[g] | 285 |
| IBU | $C_{13}H_{18}O_2$ | 206.28 | 143 mg/L[h] | 4.5–5.2[h] | 264 |

[a] WHO, 2006.
[b] Sayem Alam et al., 2007.
[c] Kasture and Wadodkar, 2008.
[d] Florence and Attwood, 2006.
[e] Windholz, 1983.
[f] Pitarresi et al., 2004.
[g] Bhise and Rajkumar, 2010.
[h] Kokot and Zmidzinska, 2001.

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