Journal of
Chemical Engineering & Process Technology

Al-Shukrawi et al., J Chem Eng Process Technol 2017, 8:5
DOI: 10.4172/2157-7048.1000365

Research Article Open Access

Characteristics of Trimethoprim Adsorption on Attapulgite Iraqi Clay

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Abstract

Adsorption characteristics of trimethoprim medicine were determined using Attapulgite an Iraqi clay of 75 µm grain particle size in order to verify its capability as a poison antidote, adsorption process was very adequate within 30 minutes. Followed by minor and slow desorption process occurred after 60 minutes. Physical properties such as ΔH, ΔG, ΔS and K were determined and documented for this work.

Keywords: Clay adsorption; Tri methoprim adsorption by attapulgite clay; Poisons adsorption by clay; Medicine adsorption by clay; Clay characteristics for medicine

Introduction

Adsorption is the process of adhesion of atoms, ions, or molecules from a gas, liquid (adsorbate) on the surface of a solid material (adsorbent) which is occurred through Freundlich or Langmuir isotherms when forming a single layer film of adsorbate species, but multilayer of adsorbate different species formed on the adsorbent adsorbents, the process is going on basis of Brunauer, Emmett and Teller's models which is random distribution of molecules on the adsorbent surface [1,2] Since ancient times Clays and activated charcoals Clays were traditionally considered as natural poisons antidotes for their high adsorption capacities for hydrophobic materials [3,4], nowadays clays were practically used to detox the catastrophic radiation contaminants as principal components in the bury matrix over Chernobyl reactor (1986) for their known adsorption ability to reduce the escape of radioactive contaminants [5-7]. Activated charcoals and clays act via different complex formation ways such as an inert complex, detoxificant, eliminator for toxic effects via receptor site blockade and/or competition of toxins [7]. Nano clays are extensively used to produce reinforced polymers for automotive industry textile industry, paints, inks greases, cosmetics, medicines (as drug delivery vehicles) and waste water treatment technologies. Newly developed multifunctional nanoparticles for biomedical and biotechnological applications to improve cancer therapy and biomedical researches such as DNA transfection, directed drug delivery, intra vital imaging as well as enzymes bioresearches [8-16]. The aim of this scientific work is to test the capability of Attapulgite (an Iraqi clay) for treatment accidental poisoning cases by (2,4-Diamino-5-(3,4,5-trimethoxybenzyl) pyrimidine as seen in Figure 1 [17-21].

Attapulgite is a needle-like naturally occurring clay (Figure 2), it is a common magnesium-aluminum silicates mineral, it is also called Polygorskite which is salt gel, fuller's earth, and activated natural clay of the following chemical composition. Attapulgite has very good dispersion colloidal properties, high adsorbing capacities, alkaline resistant, de coloring abilities, temperature resistant and has adhesive and plastic characteristics of a molecular formula is: $Mg_5Si_8O_{20}$ (HO) $2(OH_2)_4.4H_2O$ (Figure 3).

Attapulgite is of a laminated chain structure as seen in Figure 4, it looks like a soil and compact texture strongly absorbs water and/or hydrophobic materials but, wet Attapulgite exhibit plastic and adhesive characteristics. Attapulgite is a crystalloid hydrous magnesium-aluminium silicate mineral, of laminated chain lattice structure in which lattice displacement might occur, thus it clay crystals may contain uncertain quantities of Na^+, Ca^+, Fe^{3+} and Al, thus present in different the shapes such as needles, fibers. Attapulgite is used to refine animal, vegetable, and mineral oils, insecticides and fungicides carrier, oil, grease and chemical absorber and also used for soil conditioner for many applications such as poultry, golf and green houses.

Experimental Part

Instruments

The instruments used were: RF1501 UV -Vis Spectrometer, Cuvette, Quartz cuvettes, Centrifuge. Thermo static shaker bath, digital pH meter, digital pH meter, Sartorius digital balance, Oven as well as variety of glassware tools.

Materials

Chemicals: HCl 36% w/w from BDH, England, Trimethoprim was obtained from (SDI), Iraq. Attapulgite, Bentonite Flint and Kaolin Iraqi clays were obtained from the (General Company for Geological Survey and Mining).

Methodology

Clays were purified to remove the soluble impurities by washing with demineralized water until clear water outwash, dried at 115°C for 3 hrs to constants weight, crushed using electrical crusher machine, sieved with the aid of standard molecular sieves to the mesh numbers of 75, 80,150, 200 and 250 µm then stored dry. Adsorption experiments for each purified dry clay (0.1 gm of 75 µm particle size) by contact with 10 ml of each of trimethoprim stock solutions with concentrations $5 \times 10E^{-3}$, $5 \times 10E^{-4}$, $5 \times 10E^{-5}$ and $5 \times 10E^{-6}$ M/L respectively using a thermostatic shaking water bath running at 150 rpm. Series of experiments were done to determine the best contact time for adsorption process at 25°C, then to determine the kinetic physical constants of Gibbs free energy (ΔG), enthalpy (ΔH) and entropy (ΔS). Samples were twice centrifuged at 5000 rpm for 2 minutes, clarified using paper filter no. 542 for nanoparticles separation to prepare ultra clear solution for accurate

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Received November 07, 2017; **Accepted** December 08, 2017; **Published** December 22, 2017

Citation: Al-Shukrawi AH, Al-Baitai AY, Fadhel DH (2017) Characteristics of Trimethoprim Adsorption on Attapulgite Iraqi Clay. J Chem Eng Process Technol 8: 365. doi: 10.4172/2157-7048.1000365

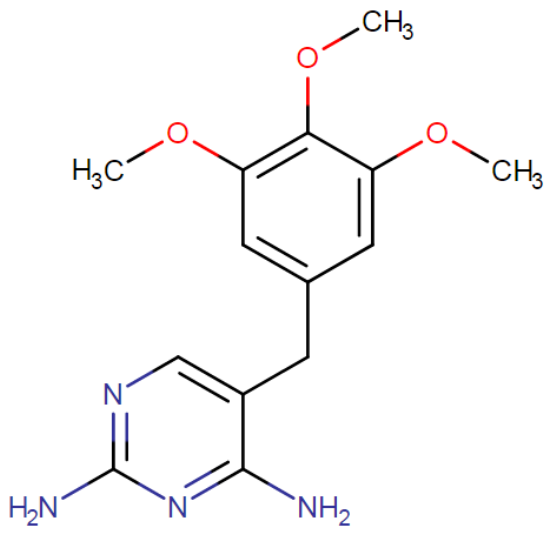

Figure 1: Structural formula of TM (2, 4-Diamino-5-(3,4,5-trimethoxybenzyl) pyrimidine).

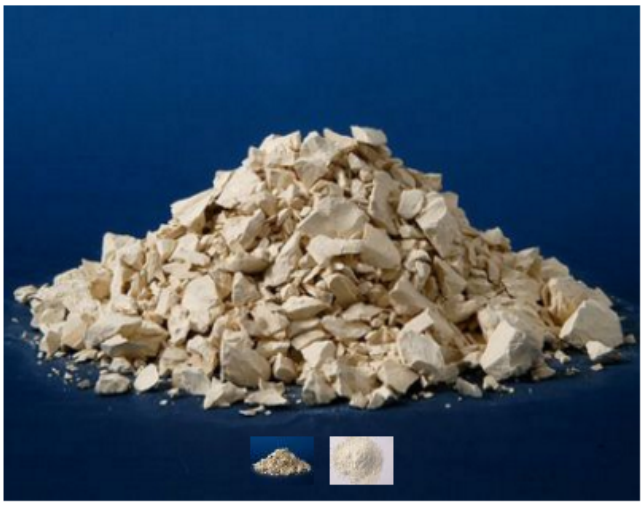

Figure 2: Attapulgite Clay Mineral.

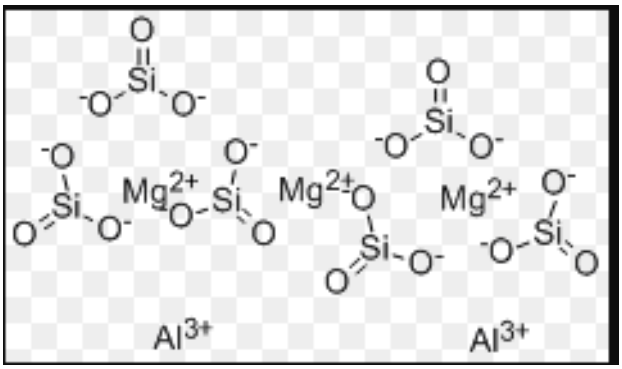

Figure 3: Structure of Attapulgite Clay.

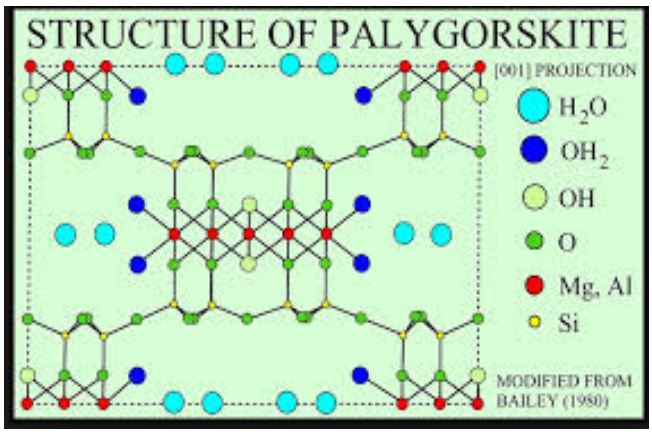

Figure 4: Photograph Showing the Structure of Attapulgite.

spectrophotometric analysis. Serious of adsorption experiments were done to determine the extinction coefficient for each adsorbate solution at different concentrations at each clay [22-24].

Results and Discussion

Extinction coefficient (ε) was determined accurately determined (Figure 5) for trimethoprim adsorption process at room temperature (25°C) for trimethoprim at serious molar concentrations and was found to equal to 1.4×10^4. Analytical data were determined according to Beer's - Lambert law as seen by equation 1:

$$A=\varepsilon b c=\varepsilon c \ (b=1) \quad (1)$$

Attapulgite clay proved to be very efficient adsorbent for the excess toxic dose of trimethoprim drug within 30 minutes contact time at 25°C and showing minor desorption characteristic (reverse reaction) during the time interval 60-120 minutes during adsorption process as seen in Figure 6, but the clay can adsorb much higher quantities than the other two clays (Bentonite and Flint) due to nature of Attapulgite molecular structure which thought to somewhat a helical form structure, therefore the adsorption process can occur on the both sides of the aminated helical structure.

Trimethoprim/Attapulgite adsorption (Figure 7) is showing Freundlich and Küster isotherm (1894) due to the relationship of Log Qe (mg of adsorbate /g of clay) against the equilibrium concentration (Ce (M / L) for the adsorption process which is a straight line for the clay which satisfies the following formula (Eq 1 and 2) which could be applied for the gaseous or solid adsorbates, where C is the molar concentration of the adsorbate, (x / m) the mass ratio of adsorbate to adsorbent (Qe), k and n are empirical constants:

$$6x/M=KC\ 1/n \quad (2)$$

$$\text{Log } Qe=\log Kf+1/n \log Ce \quad (3)$$

According to Figure 7 the empirical constants of the adsorption process of trimethoprim on Attapulgite clay were calculated to yield $Kf=1.66 \times 10^4$ and 1/n=1.076 which is approximated to one indicate that the degree of trimethoprim adsorption process on Attapulgite clay is one order. The empirical constants seen in Table 1 depicts that adsorption of trimethoprim depends greatly upon the active sites that are available on the surface of the clay particles such as Si ---O, Al---O. Mg----O, Fe-----O as well as their hydrogen atoms resulting in the formation of hydrogen bonding there with the higher electronegative

oxygen and nitrogen atoms of the active groups of the drug (Table 2).

According to Vant Hoff -Arrenheus equation:

$$\log X_m = \frac{-\Delta H}{2.303RT} + \frac{\Delta E}{R} \quad (4)$$

Enthalpy (ΔH) and entropy (ΔS) changes could be calculated using (Figures 5 and 6). Where: Xm=maximum adsorbed mass of the adsorbate (mg) in one gram of the adsorbent. Gibbs free energy change is also calculated using the following equation at 310.5 K:

$$G=\Delta H-T\Delta S \quad (5)$$

The values of the physical constants are shown in Table 3 conclude that the adsorption process occurred spontaneously for the negative values of ΔG, meanwhile ΔH is positive for attapulgite (Figure 8) and is negative for Bentonite, the overall free energy proceed the adsorption reaction readily spontaneously.

The reaction rate of the adsorption process can be calculated as follows:

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

Clay Type	1/n	Kf	n	r
Attapulgite	1.076	1.66×10^4	0.93	0.998

Table 1: The empirical constants for trimethoprim adsorption on attapulgite clay.

Component	%	%
CaO	2.02	6.2-7.94
MgO	9.05	6
K_2O	0.75	
Na_2O	0.83	
TiO_2	0.60	
Al_2O_3	8.76	13.6-16.4
SiO_2	53.64	42.2-50.8
Fe_2O_3	3.36	6.4-7.52
MnO	0.23	
H_2O	10.89	12

Table 2: Elemental Compositions of Attapulgite.

Clay type	ΔH, Kj/M	ΔG, Kj/M	ΔS, J/M	K
Attapulgite	16.053	20.275	-14.16	3×10^{-4}

Table 3: The physical constants values for adsorption of trimethoprim by attapulgite clay.

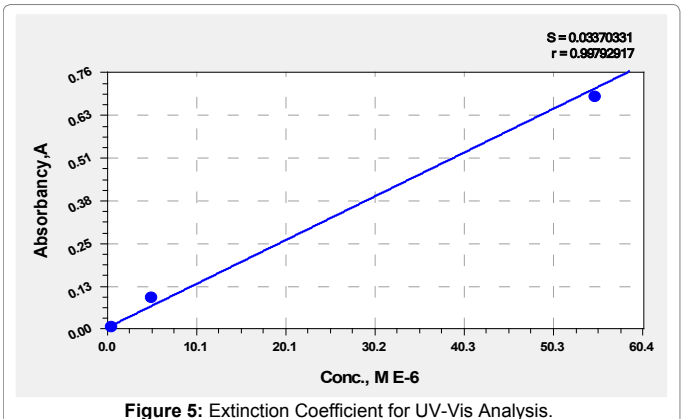

Figure 5: Extinction Coefficient for UV-Vis Analysis.

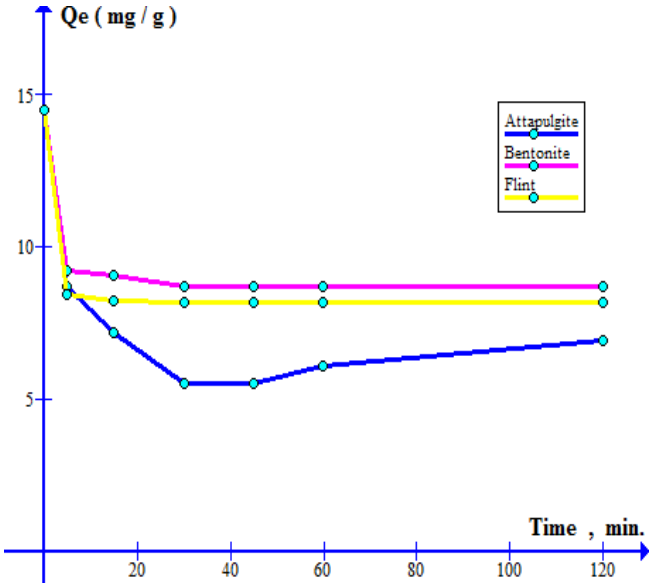

Figure 6: Residence Time for Trimethoprim Adsorption on Iraqi Clays.

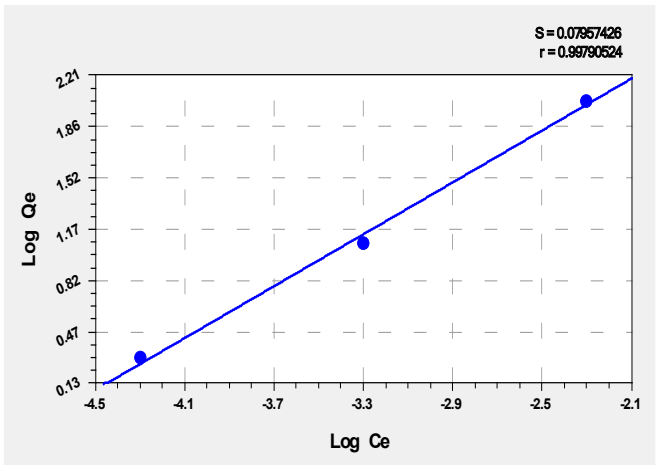

Figure 7: Attapulgite Adsorption Isotherm for Trimethoprim on Attapulgite.

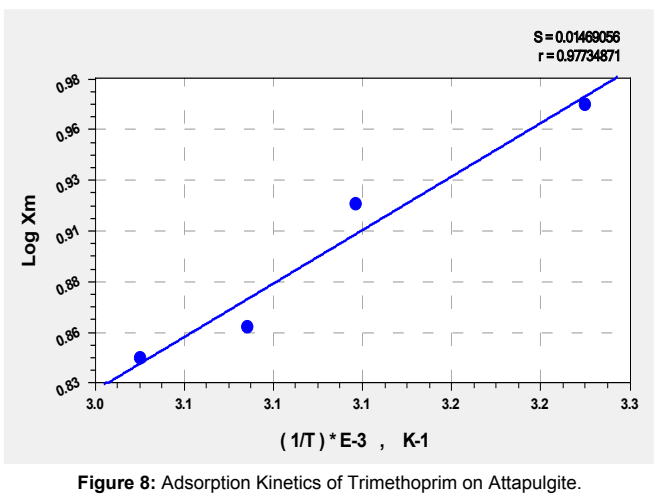

Figure 8: Adsorption Kinetics of Trimethoprim on Attapulgite.

$$K=3 \times 10^{-4} \quad (7)$$

Conclusion

The loading capacity of adsorbing Attapulgite is 2-3 times folds than the other clays (~47 mg) of trimethoprim/g of Attapulgite), this value will increase up to 5 times fold by converting the clays to nanoparticles for surfaces area increase thousands times fold therefore the adsorbing active sites of the clays which is responsible for attraction of adsorbate molecules and/or ions are greatly increased.

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