Available online on www.ijddt.com
International Journal of Drug Delivery Technology 2019; 9(2); 125-129
doi: 10.25258/ijddt.9.2.3

ISSN: 0975 4415

Research Article

Removal of Vitamin B6 (Pyridoxine) Antibiotics Pharmaceuticals From Aqueous Systems By ZnO

Aseel M. Aljeboree

Department of Chemistry, College of Science for Women, University of Babylon, Iraq

Received: 7th Jan, 19; Revised: 19th Feb, 19, Accepted: 14th Mar, 19; Available Online: 25th Jun, 2019

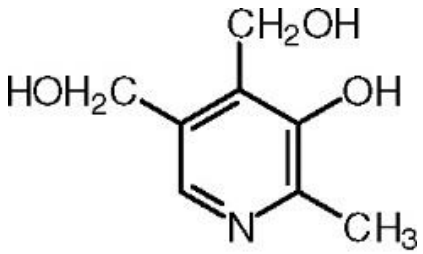
ABSTRACT

Pharmaceutical products are being increasingly discovered, in the environment. However, traditional treating systems do not provide an adequate remedy, for pharmaceutical drug elimination and still there is not a regulated criterion for their limitation in water. Adsorption is one of the most efficient and practical techniques to remove pollutants from water. ZnO to be used as adsorbent being abundant and cheap has removal capabilities for certain pollutants from water such as drug,drug, metal ions, phenol, and different anions. In this paper the potential of ZnO for removal of vitamin B_6 has been discussed. Different parameters like, initial drug concentration (5-80 mg/L), temperature (15, 25, and 50 ºC), adsorbent dosage (0.001-0.15gm), were investigated. The adsorption of drug was best at neutral pH. The adsorption uptake decrease with increase in initial drug concentration, but increase with the amount of adsorbent and temperature. The equilibrium was evaluated using Langmuir, Freundlich isotherms. The maximum capacity of adsorption obtained from the Freundlich model was 108.556mg/g.

Keyword: Pharmaceuticals, antibiotics, ZnO, Adsorption, vitamin B6, Adsorption isotherms

INTRODUCTION

Pharmaceuticals and antioxidants are natural or synthetic substances are environmental pollutants because of their low biodegradability, facile bioaccumulation and high persistence [1, 2]. And they have received increasing worry for their pollutions [3] contain varied groups, such as anti-inflammatory agents,antibiotics, steroidal hormones and blood-lipid regulators [4, 5]. As a result of repeated use, enormous amounts of antioxidant substances are accumulated in the aquatic environments [6-8] and the pollution of ground water and surface has emanate as grave problem in recent years [1, 9] result entry of Pharmaceuticals through diverse waste streams like hospitals, households and pharmaceutical companies, raise an emerging ecological issue that need to be concerned [10]. Thus, these pollutants must be eliminated from wastewater. Several treating processes were reported to remove pharmaceuticals from water, like activated sludge systems [11, 12] photo catalytic oxidation processes oxidative degradation [13, 14], bio membrane, membrane bioreactors [14], ozonation,biological filtration [15], reverse osmosis [4], membrane filtration [15] and adsorption processes [16-19]. Regardless of technology applied, adsorption is always involved [20]. Many researchers have investigated the adsorptive removal of pharmaceuticals by carbon nanotubes [21, 22] activated carbon [17, 23], Graphene [24], ZnO, TiO_2 [25], BN based materials [26], layered hydroxides [27] natural aquifer materials [28], soils [29], and sediment [30]. Although adsorption is a simple technology, the adsorbed Vitamin B_6 on the adsorbent need further handling to reduce its risk [31, 32]. The goal of this study was to esteem



vitamin B_6

Figure 1: Chemical stretcher Vitamin B6

the efficacy of ZnO and new adsorbent to removing the drug Vitamin B_6 from aqueous solutions and wastewater. Study affect different parameters such as, temperature, adsorbent dose and the drug Vitamin B_6 concentration were evaluated. Also adsorption isotherms and Thermodynamic were also analyzed**.**

MATERIALS AND METHODS

Preparation of Calibration Curve

ZnO was supplied from Sigma-Aldridge/Germany with purity 99%. Freshly prepared aqueous solution of pure drugs. Vitamin B_6 standard solutions (100mg/L) was prepared by dissolving 0.1gm of drug in distilled water, the solution was made up to 1000mL with distilled water transferred into a series of (5-80 mg/L) in 100 mL

**Author for Correspondence: annenayad@gmail.com*

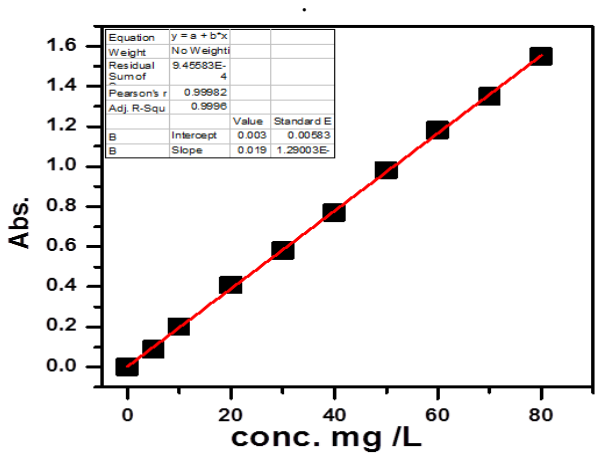


Figure2 . The calibration curve of Drug vitamin B_6

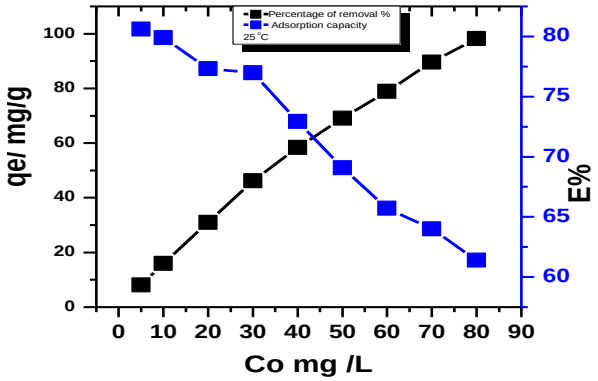


Figure 4: The effect of initial concentration on adsorption of B6 using ZnO.

calibrated volumetric flasks. The absorbance determined spectrophotometric ally by using UV-Visible Spectrophotometer as shown in Figure 2 Commercial ZnO powder was purchased from (sigma-Aldrich, Germany). Drug vitamin B_6 (Samara Company of drug manufacturing, Iraq). All chemicals were of analytical grade.

The varying the quantity of ZnO was studied in the adsorption of vitamin B_6 in range of (0.01, 0.03, 0.05, 0.075, 0.1, and 0.15 g/100mL).

The effect of initial drug vitamin B_6 concentration was studied with different concentrations in the range of (5-80 mg/L), 120 rpm, 0.05gm of ZnO at room temperature.

The temperature effect on ZnO adsorption capacity was conducted at (15, 25, and 50°C) in water bath at 120rpm, 0.05gm of ZnO and 100mL of drug concentration (5–80mg/L.

All the experiments were conducted at optimum values of parameters like initial drug concentration 50 mg/L, ZnO mass (0.05 g/100mL) and temperature at 25°C.

The solution was shaken for 24h then the concentration of residue was measured to determine the optimum adsorbent value. The amount of adsorption (q_e) and removal percentage (E%) were calculated according to the drug concentration before and after adsorption, using the equations mentioned in [33]

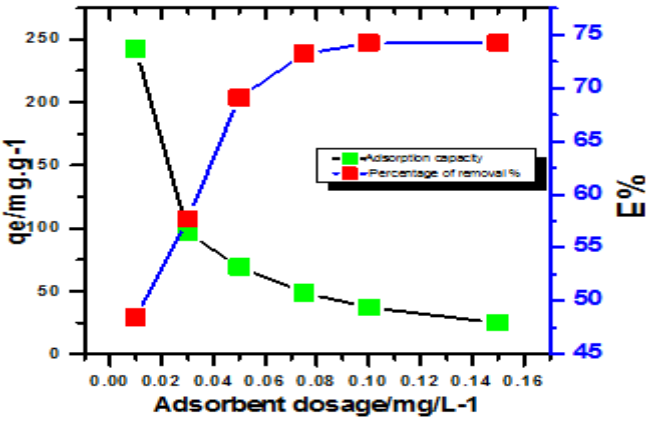


Figure 3: The effect of initial adsorbent dose on B6 adsorption using ZnO surface

RESULTS AND DISCUSSION

Adsorbent concentration

Proper selection is required for Initial adsorbent dose because it depends on surface area and binding site availablity [18]. The adsorption efficiency and capacity is related inversely with adsorbent dose (Figure 3). Drug uptake increased from 48.453% to 74.226%, while the capacity of adsorption decreased from 242.268mg/g to 24.478 mg/g against the dose of adsorbent (0.01–0.15 g/100mL), respectively [34]. This result attributed to as that increasing the active sites caused by increasing amount of adsorbent [35].

The adsorption capacity (qe) is proportional with the initial drug concentration increase. But a percentage decrease of drug removal in the initial concentrations. The drug removal by adsorption on ZnO strting rapid with low drug concentrations then slow with increase in drug concentration. This is due to Van der Waals forces and electrostatic attractions [34, 36-38] (Figure 4).

The temperature effect

The B6 adsorption on ZnO was studied at 15 to 50ºC. The adsorption efficiency of drug onto the adsorbent surface of ZnO is approved. The efficiency of adsorption increased from 6.59 mg/g (44.97% removal) to 70.20 mg/g (67.77 % removal), 8.06 mg/g (61.40% removal) to 98.24 mg/g (80.61% removal) and 8.40 mg/g (67.84 % removal) to 108.55 mg/g (84.02 % removal) for initial concentration 5-80 mg/L of B6, when the temperature increased from 15-50°C at the same order [35], results are shown in Figure 5, the adsorption process found to be endothermic in nature. By the temperature increase, the diffusion rate of the adsorbate molecules and the internal pores of adsorbent particle increases with decreasing solution viscosity [38].

Adsorption isotherms

The adsorption isotherm was conducted at 25ºC in different initial Vitamin B6 concentrations. The adsorption of Vitamin B6 increases until equilibrium with no further change (Figure 6). High initial concentration of Vitamin B6 provides the strong driving force against mass transfer resistances [39, 40]. The equilibrium for the adsorption of

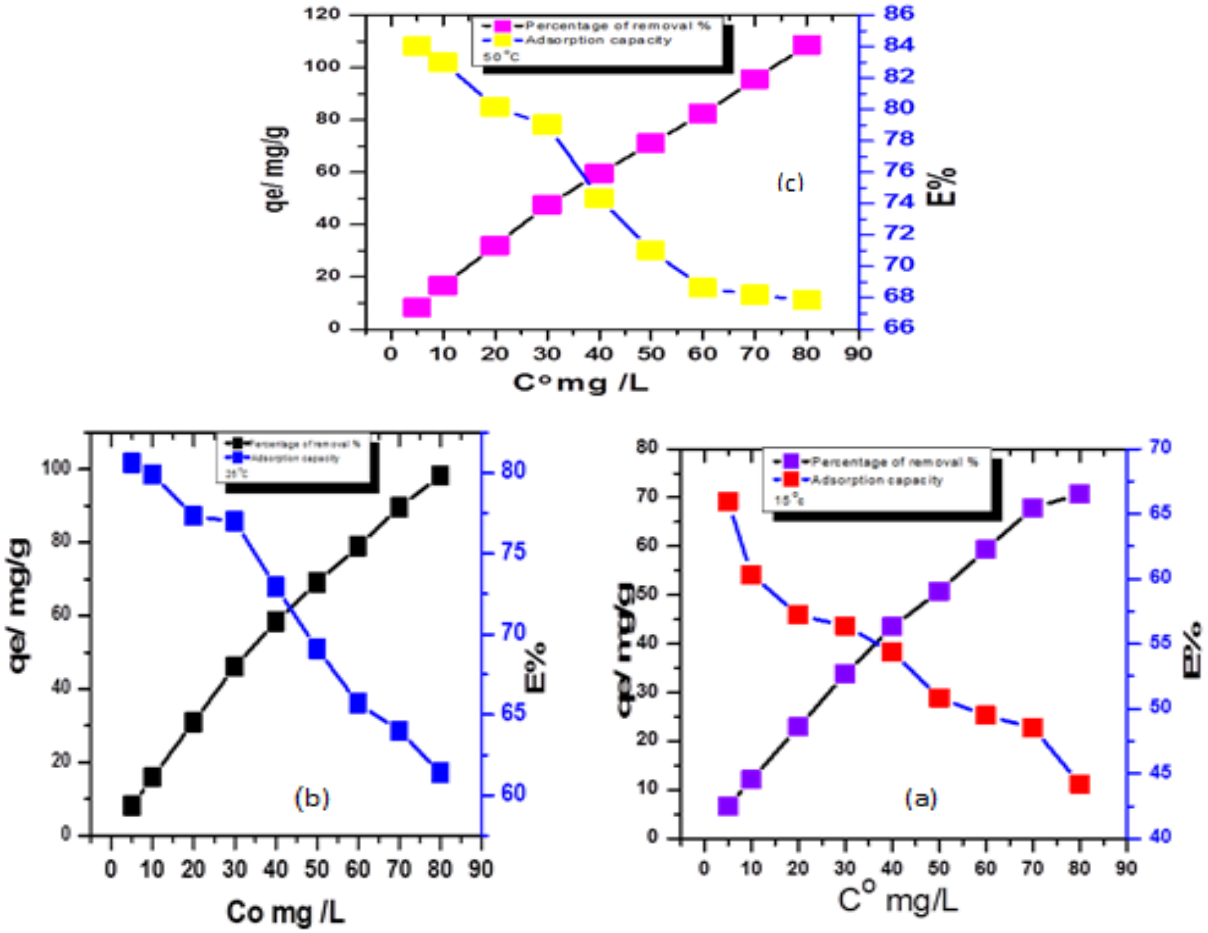


Figure 5: The effect of temperature on adsorption of B6 using ZnO (a)15ºC , (b)25ºC , (c)50ºC

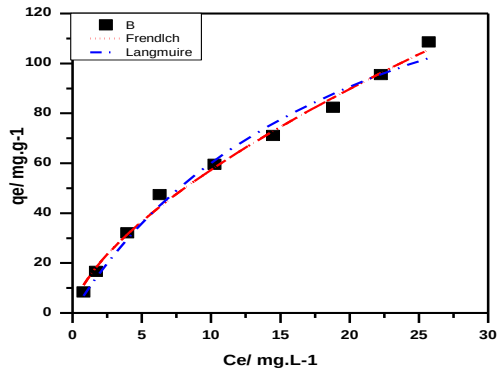


Figure 6: The adsorption isotherm of B6 drug (pH 6, Temp. 298 K, mass dosage 0.05 gm/100 ml, t = 24 h, agitation speed = 120 rpm)

Vitamin B6 was analyzed by the Langmuir and Freundlich isotherm equations mentioned in [41] The parameters of Langmuir and Freundlich equations are summarized in Table 1. Temperature has a positive influence on the amount of Vitamin B_6 adsorbed and is proportional with the adsorption capacity.

The Freundlich isotherm model showed the best fit for Vitamin B_6 adsorption on ZnO; as seen in the R^2 values

Table 1: The Langmuir, Freundlich, the isotherms parameters for B6 adsorbed on ZnO at 25°C

Isotherm model	Parameter	B6	Standard Error
Langmuir	qm ($mg.g^{-1}$)	184.582	±21.5822
	K_L($L.mg^{-1}$)	0.0423	±0.01013
	R^2	0.9845	
Freundlich	K_F	12.9633	±1.11423
	1/n	0.64591	±0.02960
	R^2	0.9924	

and K_F increase with increased adsorption temperature (Table 1).

CONCLUSION

The equilibrium between Vitamin B_6 and ZnO surface was achieved in 24h. The Freundlich (R2 = 0.9924) is the best isotherm that described the adsorption process and then followed Langmuir isotherms. Isothermal results showed that Vitamin B_6 adsorption was favorable on ZnO and followed multilayer adsorption. The capacity of adsorption (qe) is proportional with the initial drug concentration, while the removal percentage decrease with initial drug concentrations. The maximum adsorption capacity of Vitamin B_6 on ZnO was found to be 108.55 mg/g. There

for, Vitamin B6 can be removed effectively from aqueous solution using ZnO as an economic adsorbent.

REFERENCES

1. Aseel Musthaq Aljeboree, A.N.A., Colorimetric Determination of phenylephrine hydrochloride drug Using 4-Aminoantipyrine: Stability and higher sensitivity J. Pharm. Sci. & Res., 2018, 10(7): p. 1774-1779
2. Zhang, S., et al., Adsorption of pharmaceuticals on chitosan-based magnetic composite particles with core-brush topology. Chemical Engineering Journal. 304: p. 325-334.
3. Peng, G., et al., Adsorption and catalytic oxidation of pharmaceuticals by nitrogen-doped reduced graphene oxide/Fe3O4 nanocomposite. Chemical Engineering Journal. 341: p. 361-370.
4. Aseel M. Aljeboree, A.N.A., Adsorption of Pharmaceuticals as emerging contaminants from aqueous solutions on to friendly surfaces such as activated carbon: A review J. Pharm. Sci. & Res., 2018, 10 :(9)p. 2252-2257
5. Sun, J., et al., Occurrences of pharmaceuticals in drinking water sources of major river watersheds, China. Ecotoxicology and Environmental Safety. 117: p. 132-140.
6. Nazari, G., H. Abolghasemi, and M. Esmaieli, Batch adsorption of cephalexin antibiotic from aqueous solution by walnut shell-based activated carbon. Journal of the Taiwan Institute of Chemical Engineers. 58: p. 357-365.
7. ASEEL M ALJEBOREE, ABBAS NOOR ALSHIRIFI, Spectrophotometric Determination of phenylephrine hydrochloride drug in the existence of 4- Aminoantipyrine: Statistical Study. International Journal of Pharmaceutical Research, 2018, 10(4.(
8. Liu, J.-L. and M.-H. Wong, Pharmaceuticals and personal care products (PPCPs): A review on environmental contamination in China. Environment International. 59: p. 208-224.
9. Song, J.Y. and S.H. Jhung, Adsorption of pharmaceuticals and personal care products over metal-organic frameworks functionalized with hydroxyl groups: Quantitative analyses of H-bonding in adsorption. Chemical Engineering Journal. 322: p. 366-374.
10. Sarmah, A.K., M.T. Meyer, and A.B.A. Boxall, A global perspective on the use, sales, exposure pathways, occurrence, fate and effects of veterinary antibiotics (VAs) in the environment. Chemosphere, 20 :(5)65 .06p. 725-759.
11. P.N. Carvalho, A.P., M. Basto, P. Clara, Activated sludge systems removal efficiency of veterinary pharmaceuticals from slaughterhouse wastewater. Environ. Sci. Pollut. Res, 2013, 20(12): p. 8790-8800.
12. Watkinson, A.J., E.J .Murby, and S.D. Costanzo, Removal of antibiotics in conventional and advanced wastewater treatment: Implications for environmental discharge and wastewater recycling. Water Research, 2007, 41(18): p. 4164-4176.
13. Chen, W., et al., Synthesis of MnOx/SBA 15-for Norfloxacin degradation by catalytic ozonation. Separation and Purification Technology. 173: p. 99-104.
14. Mart-ínez, F., et al., Coupling membrane separation and photocatalytic oxidation processes for the degradation of pharmaceutical pollutants .Water Research. 47(15): p. 5647-5658.
15. Ziylan, A. and N.H. Ince, The occurrence and fate of anti-inflammatory and analgesic pharmaceuticals in sewage and fresh water: Treatability by conventional and non-conventional processes. Journal of Hazardous Materials. 187(1): p. 24-36.
16. Dom-ínguez, J.n.R., et al., Removal of common pharmaceuticals present in surface waters by Amberlite XAD-7 acrylic-ester-resin: Influence of pH and presence of other drugs. Desalination. 269(1): p. 231-238.
17. Aseel M Aljeboree, A.N.A., Ayad F Alkaim, Kinetics and equilibrium study for the adsorption of textile drugs on coconut shell activated carbon. Arabian journal of chemistry, 2017, 10: p. 3381-3393.
18. Aljeboree, A.M., Adsorption of methylene blue drug by using modified Fe/Attapulgite clay Research Journal of Pharmaceutical, Biological and Chemical Sciences, 2015, 6(4): p. 778.
19. Liu, W., et al., Sorption of norfloxacin by lotus stalk-based activated carbon and iron-doped activated alumina: Mechanisms, isotherms and kinetics. Chemical Engineering Journal. 171(2): p. 431-438.
20. K.A. Landry, T.H.B., Diclofenac removal in urine using strong-base anion exchange polymer resins. Water Res., 2013, 47(17): p. 6432-6444.
21. Ayad Fadhil Alkaim, Z.S., Dunia Kamil Mahdi, Saif Mohammed Alshrefi, Abdulkareem Mohammed Al-Sammarraie, Faisal Muhammad Alamgir, Preet Mohinder Singh, Aseel Mushtaq Aljeboree, Preparation, structure and adsorption properties of synthesized multiwall carbon nanotubes for highly effective removal of maxilon blue drug. Korean Journal of Chemical Engineering, 2015, 32(12): p. 2456-2462.
22. H.H. Cho, H.H., K. Schwab, Effects of solution chemistry on the adsorption of ibuprofen and triclosan onto carbon nanotubes Langmuir, 2011, 27(21): p. 12960-12967.
23. Guedidi, H., et al., The effects of the surface oxidation of activated carbon, the solution pH and the temperature on adsorption of ibuprofen. Carbon. 54: p. 432-443.
24. W. Song, T.Y., X. Wang, Y. Sun, Y. Ai, G. Sheng, et al., Experimental and theoretical evidence for competitive interactions of tetracycline and sulfamethazine with reduced graphene oxides. Environ. Sci. Nano, 2016, 3 p. 1318-1326.
25. Alkaim, A.F., Alrobayi, Enas M,Algubili, Abrar M and Aljeboree, Aseel M, Synthesis, characterization, and photocatalytic activity of sonochemical/hydration–dehydration prepared ZnO rod-like architecture nano/microstructures assisted by a biotemplate.

Environmental technology, 2017, 38(17): p. 2119-2129.
26. S. Yu, X.W., H. Pang, R. Zhang, W. Song, D. Fu, et al., Boron nitride-based materials for the removal of pollutants from aqueous solutions: a review. Chem. Eng. J., 2017, 333: p. 343-360.
27. W. Yao, S.Y., J. Wang, Y. Zou, S. Lu, Y. Ai, et al., Enhanced removal of methyl orange on calcined glycerol-modified nanocrystallined Mg/Al layered double hydroxides. Chem. Eng. J., 2017, 307: p. 476-486.
28. A.C. Hari, R.A.P., D.A. Sabatini, T.C.G. Kibbey and Effects of pH and cationic and nonionic surfactants on the adsorption of pharmaceuticals to a natural aquifer material. Environ. Sci. Technol., 2005, 39(8): p. 2592-2598.
29. Sukul, P., et al., Sorption and desorption of sulfadiazine in soil and soil-manure systems. Chemosphere, 2008, 73(8): p. 1344-1350.
30. K. Stein, M.R., G. Fink, M. Sander, T.A. Ternes, Analysis and sorption of psychoactive drugs onto sediment. Environ. Sci. Technol., 2008, 42(17): p. 6415-6423.
31. Bui, T.X. and H. Choi, Influence of ionic strength, anions, cations, and natural organic matter on the adsorption of pharmaceuticals to silica .Chemosphere. 80(7): p. 681-686.
32. Goyne, K.W., et al., Sorption of the antibiotic ofloxacin to mesoporous and nonporous alumina and silica. Journal of Colloid and Interface Science, 2005, 283(1): p. 160-170.
33. ALJEBORI, A.M.K.A., ABBAS NOOR, Effect of Different Parameters on the Adsorption of Textile Drug Maxilon Blue GRL from Aqueous Solution by Using White Marble. Asian Journal of Chemistry, 2012, 24(12): p. 5813-5816.
34. Aljeboree, A.M., Adsorption of crystal violet drug by Fugas Sawdust from aqueous solution. International Journal of ChemTech Research, 2016, 9: p. 412-423.
35. Ahmed M. Kamil, H.T.M., Ayad F. Alkaimand Falah H. Hussein Adsorption of Congo red on multiwall carbon nanotubes: Effect of operational parameters Journal of Chemical and Pharmaceutical Sciences 2016, 9(3.(
36. Aseel M Aljeboree, A.F.A., Ammar H Al-Dujaili, Adsorption isotherm, kinetic modeling and thermodynamics of crystal violet drug on coconut husk-based activated carbon. Desalination and Water Treatment, 2015, 53(13): p.3667-3656 .
37. A F Alkaim, M.B.A., Adsorption of basic yellow drug from aqueous solutions by Activated carbon derived from waste apricot stones (ASAC): Equilibrium, and thermodynamic aspects. international journal of chemical sciences, 2013, 11(2): p. 797.814-
38. Alqaraguly, A.F.A.A.M.B., Adsorption of basic yellow drug from aqueous solutions by activated carbon derived from waste apricot stones (asac): equilibrium, and thermodynamic aspects. Int. J. Chem. Sci., 2013, 11(2): p. 797-814.
39. H.F. Xiong, D.L.Z., D.D. Zhou, S.S. Dong, J.W. Wang, B.E. Rittmann, Enhancing degradation and mineralization of tetracycline using intimately coupled photocatalysis and biodegradation (ICPB). Chem. Eng. J., 2017, 316: p. 7-14.
40. W.P. Xiong, J.T., Z.H. Yang, G.M. Zeng, Y.Y. Zhou, D.B. Wang, P.P. Song, R. Xu, C. Zhang, M. Cheng, Adsorption of phosphate from aqueous solution using iron-zirconium modified activated carbon nanofiber: performance and mechanism. J. Colloid Interface Sci., 2017, 493: p. 17-23.
41. H.P. Wu ,C.L., G.M. Zeng, J. Liang, J. Chen, J.J. Xu, J. Dai, X.D. Li, J.F. Liu, M. Chen, L.H. Lu, L. Hu, J. Wan, The interaction of composting and biochar and its implication for soil amendment and pollution remediation-a review. Crit. Rev. Biotechnol., 2016, 6 : p. 1-11.