Contents lists available at ScienceDirect

Process Safety and Environmental Protection

journal homepage: www.elsevier.com/locate/psep

# The elimination of xylene from aqueous solutions using single wall carbon nanotube and magnetic nanoparticle hybrid adsorbent

*Hamidreza Pourzamani*[a,b], *Saeed Parastar*[a,b], *Majid Hashemi*[a,b,c,*]

[a] *Environment Research Center, Student Research Committee, Research Institute for Primordial Prevention of Non Communicable Disease, Isfahan University of Medical Sciences, Isfahan, Iran*
[b] *Department of Environmental Health Engineering, School of Health, Isfahan University of Medical Sciences, Isfahan, Iran*
[c] *Department of Environmental Health Engineering, School of Health, Kerman University of Medical Sciences, Kerman, Iran*

ARTICLE INFO

*Article history:*
Received 15 November 2016
Received in revised form 28 April 2017
Accepted 20 May 2017

*Keywords:*
Xylene
Single-wall carbon nanotubes
Iron nanoparticle
Adsorption
Isotherm
Hybrid adsorbent

ABSTRACT

Hybrid adsorbent of single wall carbon nanotubes- magnetic nanoparticle (SWCNT-MN) has been used for the elimination of a variety of pollutants. The aim of the present study was to eliminate xylene from aqueous solutions using this hybrid adsorbent. The xylene solution was made in a synthetic manner with 10, 30, 70, and 100 mg/l concentrations. In different pHs (2–11), nanoparticles of magnetic iron were added to the solution in different concentrations namely 500, 1000, 1500, and 2000 g/l and the efficiency of xylene elimination was determined by SWCNT-MN nanoadsorbents after 2–20 min. Xylene was measured using GC/MS device. The data were analyzed by Design Expert software using Taghochi OA method. Also, ISOFIT software was used to investigate the isotherm of xylene adsorption. The optimal conditions of xylene elimination was obtained in primary concentration of 100 mg/l, adsorbent dosage of 2000 mg/l, contact time of 20 min, and pH 8. In the created optimal conditions, 99.2% of xylene was eliminated by SWCNT-MN nanoadsorbents and the adsorption capacity of SWCNT-MN nanoadsorbents for xylene was 50 mg/g. The effect of main factors and important interaction obtained by the software in designing the experiment showed that Prob □ F was less than 0.05 for all main factors and the interaction between xylene's primary concentration and SWCNT-MN nanoadsorbents dosage. Among the main factors, the contact time had the highest effect (34%). The investigation of isotherm of xylene adsorption by SWCNT-MN nanoadsorbents indicated that GLF isotherm is the most appropriate adsorption isotherm.



## 1. Introduction

BTEX which is a general name for benzene, toluene, ethylbenzene, and xylene isomers (para, meta, and ortho xylene) are typical volatile organic compounds (VOCs) that are harmful to the environment and human health (Wang et al., 2016). Xylene is a clear, colorless, ignitable, and hydrophobic liquid which has pungent smell. Xylene is extensively used as solvent in dye, caoutchouc, leather, insecticide, and covering industries (Saravanan and Rajamohan, 2009). It was reported that BTEX contamination indeed enhanced the toxicity of groundwater. USEPA has determined the criterion of xylene isomers to be 10 mg/l for drinking water (Shim et al., 2002). The removal of xylene from aqueous

* *Corresponding author at*: Environment Research Center, School of Health, Isfahan University of Medical Sciences, Hezar Jerib Ave., Isfahan, Iran. Fax: +98 311 669 5849.
E-mail address: mhashemi120@gmail.com (M. Hashemi).
http://dx.doi.org/10.1016/j.psep.2017.05.010
0957-5820/

solutions has been extensively studied and a variety of physical, chemical, and biological processes have been successfully used to eliminate this pollution. The processes which have been employed till date to refine waters polluted by xylene include biological technology (Shim et al., 2002), adsorption by different adsorbents (Lu et al., 2008), the use of membrane processes (Yahaya, 2008), wet air oxidation (WAO) and advanced oxidation process (Ramteke and Gogate, 2016). Adsorption process is regarded as the most common.

$Fe_3O_4$ nanoparticles have the nature of super paramagnetic which enables it to be easily separated under magnetic condition. Currently, one of the important problems in the application of nanoparticles is their separation after the given reactions (Ren et al., 2008). In order to eliminate pollutants and special compounds from sewage, the magnetic nanoparticles can be modified. The modification of these nanoparticles can be done by combining or modifying functional groups with inorganic compounds created in magnetic adsorbent (Chang et al., 2006).

Carbon nanotubes ($CNT_s$) are unique single-dimensional macromolecules with high thermal resistance and chemical stability (Shim et al., 2002). Carbon nanotubes have pin like structure which are created due to graphite molecules in the form of lamella individual tubes leading to single-wall carbon nanotubes (SWCNT), or a large number of individual tubes with different diameters which are formed around a pivot building multi-wall carbon nanotubes (MWCNT). Owing to their high specific surface area, small size and hollow and layered structures, it has been demonstrated that they possess great potential as superior adsorbents for the separation of various kinds of organic and inorganic materials (Fazelirad et al., 2015). Carbon nanotubes have more adsorption capacity than other carbon compounds (such as porous graphitic carbon (PGC)). Since carbon nanotubes are placed next to each other in the form of many graphitic lamella and generate weak intermolecular Van der Waals forces, hence the compounds are effectively absorbed by nanotubes (Valcárcel et al., 2008; Su et al., 2010). Combination of magnetic nanoparticles with carbon-based materials, have several unbeatable features including good thermal stability, a wide electrochemical (conductivity) spectrum, tunable miscibility, and good extraction capability, which make them a promising materials for numerous environmental applications such as water and wastewater treatment (Yu et al., 2016a). Yu et al. (2016a) conducted a research on toluene, ethylbenzene and xylene removal from aqueous solution by magnetic iron oxide nanoparticles functionalized multi-walled carbon nanotubes. Ghaedi et al. (2011) used multi-wall carbon nanotubes to eliminate organic compounds namely alizarin and murine (Ghaedi et al., 2011). (Su et al., 2010) used carbon nanotubes oxidized by NaOCl to eliminate BTEX compound. Ai et al. (2011) utilized magnetic multi-wall carbon nanotubes hybrid adsorbent to eliminate methylene blue which yielded favored results. The comparison between carbon nanotubes and other adsorbents such as active carbon showed that carbon nanotubes are better adsorbent for organic compounds (Lu et al., 2008; Yu et al., 2016b). Carbon nanomaterials (e.g. graphene and carbon nanotubes) are based on their large specific surface area (SSA), premier for making adsorbent very efficient for adsorbing contaminants, nanostructured porous layer, and the ease factor (Yu et al., 2016a), while magnetic materials for the separation of pollutants from the adsorbent due to the inherent advantages of magnetic separation process "green process" is simple, efficient and economical. In this study, during the construction of the magnetic nanoparticle we have added CNT, as a result carbon nanotubes were used to control the size of the building $Fe_3O_4$ within the tube. In addition to controlling the size of $Fe_3O_4$, the magnetic CNTs will also facilitate separation. Furthermore, the aim of this study was to investigate the efficiency of xylene elimination by SWCNT-MN hybrid adsorbent. The adsorption isotherms were also studied in this research.

## 2. Materials and methods

### 2.1. Stock sample preparation

Ultrapure xylene was purchased from Merck Company. To prepare the samples, first, a 100 mg/l xylene stock solution was made and the required concentrations were provided. To dissolve these compounds in water, the mentioned solution was placed in ultrasonic bath for 60 min and then mixed at 25 °C for 24 h. After 24 h, the ultrasonic bath was repeated for 30 min. In this way, the solution was in single phase and homogenous. Milli Q Water was used to prepare and dilute the samples, and the samples were prepared daily.

### 2.2. The preparation of SWCNT-MN nanoadsorbent

The adsorbent SWCNT was purchased from the Iranian Research Institute of the Petroleum Industry. The distilled water was added to 1 g of SWCNT by the use syringe in drops so as to prepare pasty matter without extra water. Considering that at this point, we had to make a nanoadsorbent comprising 20% MN with SWCNT base, for 18 g nanoadsorbent, and 3.6 g magnetic nanoparticles that entered the pores of 15 g SWCNT. To make 18 g nanoadsorbent, nitrogen was injected to 30.6 ml of distilled water at 85 °C for 10 min. Then, $FeCl_2{\cdot}4H_2O$ (2.6 g) and $FeCl_3{\cdot}6H_2O$ (1 g) were added to the above mentioned water at the same temperature and mixed for 5 min. After 5 min, 15 g of SWCNT was added and mixed by a manual mixer. Then, 25% Ammonia (1.5 ml) was added and mixed. The mentioned solution was placed under hood at room temperature for 30 min to become homogenous. Water was removed from the sample by

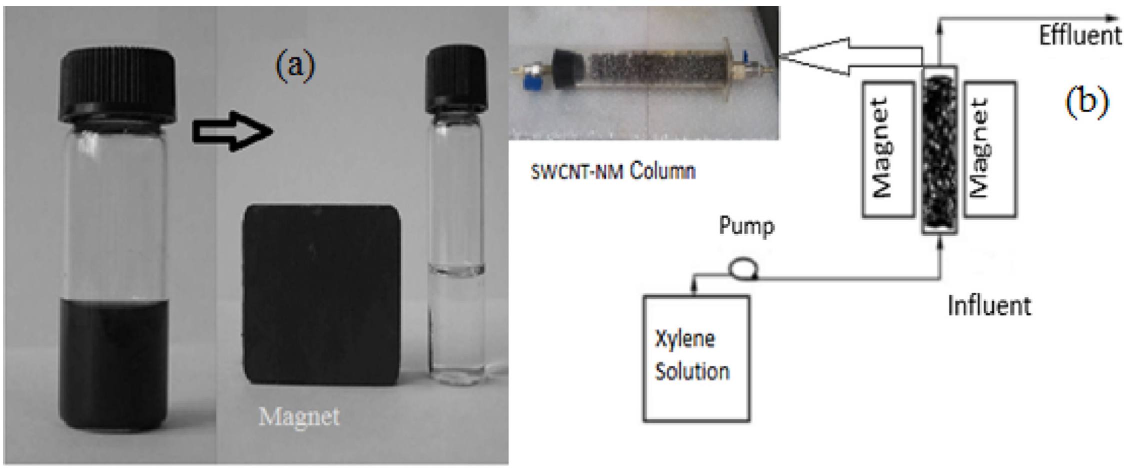


**Fig. 1 – (a) Magnetic properties of nanotubes containing iron nanoparticles (b) up flow magnetic column used for continues experiments.**