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Comparison study of adsorption and nanofiltration methods for removal of total petroleum hydrocarbons from oil-field wastewater

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ABSTRACT

The aim of the current study was to investigate and study adsorption and nanofiltration systems for separation of total petroleum hydrocarbons (TPH) from wastewater generated by an oil refinery in Bushehr, Iran. In the first step (previous study), NaA zeolite nanoparticles (NaA-ZNPs) were synthesized and granulated using a hydrothermal method in which the silica source was extracted from oat bran and then modified by the surfactant hexadecyltrimethylammonium bromide (CTAB). In the second step, polyaniline membranes and nanocomposite membranes were synthesized containing polyaniline as the main polymer and clinoptilolite zeolite as the mineral. The NaA-ZNPs and membranes were characterized by gas chromatography-mass spectroscopy (GC-MS), Fourier transform infrared spectroscopy (FT-IR), scanning electron (SEM), and atomic force microscopy (AFM). In order to evaluate the absorption and nanofiltration process, adsorbents in both batch and continuous process in a fixed-bed column with upward flow were used to remove TPH. Time, pH, and adsorbent dosage were studied in the batch process, and flow rate and column height were evaluated for the continuous process. In the batch and continuous systems, when the parameters were optimized, the highest TPH removal efficiencies were 92.3% and 87.4% respectively. Furthermore, in the nanofiltration process, permeation flux and TPH rejection were 190.96 $kg\,h^{-1}\,m^{-2}$ and 99.77%. Finally, the removal efficiency for the hybrid absorption and nanofiltration process was 99.83%. In the hybrid system, due to the combination of two absorption and nanofiltration processes, as well as the effects of NaA-ZNPs functioning as adsorbents, membrane deflation was minimized and the efficiency of removing of TPH increased to 99.83%.

1. Introduction

In Iran, as in other oil-rich countries, more and more attention continues to be placed on ameliorating the detrimental effects of oil pollution. The purpose of this study was to investigate the removal of total petroleum hydrocarbons (TPH) using granulated NaA zeolite nanoparticles (NaA-ZNPs) modified with surfactants.

Synthetic zeolites have considerable advantages over their natural forms due to their high purity, formation of pure rather than mixed samples, easy access for synthesis at industrial levels, and above all, ability to fix and control pore size (Saeed et al., 2011; Xi et al., 2015). The fundamental changes each property of this material undergoes when the particle size is reduced from micrometers to nanometers make it very effective in catalyst applications and separation performances (Saremnia et al., 2015). Zeolite synthesis is accomplished by hydrothermal crystallization of an active solution of hydrated aluminosilicate or gel in an alkaline environment. Alkaline environments can be provided with alkali hydroxides, alkaline earth, and organic and combined bases (Esmaeili and Saremnia, 2016). The permanent negative charge on the surface of clay minerals and zeolites allows their modification with a cationic surfactant. This results in more effective absorption of other compounds (anionites and nonpolar organic materials) and prevents their movement. Surface-modified zeolites can be used as a low-cost, highly potent material for elimination of contaminants (Esmaeili and Beni, 2014a, 2015).

TPH refers to the varying mixtures of hydrocarbons found in crude oil. These oils include hydrocarbons that are not heavy enough to change from a gas into a solid state. As a byproduct of oil refinery operations engaged in by petrochemical companies, the presence of TPH in the environment amounts to a global disaster (Aguilar-Arteaga et al., 2010; Esmaeili and Sadeghi). Laboratories and industries throughout the world process crude oil to yield petroleum products, making TPH a major component of the output to be found in waste refineries (Ghogare and Gupta, 2012). TPH in these outputs represents a significant threat to human, animal, and plant life, as this pollution in refinery waste leads to increases in the level of toxicity of water and in

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the general environment (Akar et al., 2009; Esmaeili and Fazeli, 2012). In recent years a number of materials and tools useful for cleaning wastewater have been identified, and various methods for separation of TPH from wastewater have been reported (Esmaeili and Sadeghi). Similar to other oil-rich countries, Iran in recent years has been faced with the elevated destructive effects of oil pollution on its ecosystem, prompting increased efforts to discover different composites that may be effective in combating the effects of TPH pollution. Biological elimination of TPH or conversion to harmless or beneficial products has been studied (Esmaeili and Sadeghi), with promising results. The novelty of this study is based on the use of an adsorption process to achieve high removal efficiency of TPHs. For this study NaA-ZNPs granulated using an alginate granulation method and modified by a hexadecyltrimethylammonium bromide (CTAB) cationic surfactant were used to decontaminate wastewater sampled from an oil refinery.

2. Materials and methods

2.1. Materials

Wastewater containing TPH (pH = 3.6) was supplied by Bahregan Oil Refinery, Bushehr, Iran. Sodium hydroxide, sodium aluminate, hydrochloric acid, sodium alginate, barium chloride, CTAB, clay, and acetone acid were obtained from Merck. Deionized water was used exclusively throughout the investigation.

2.2. Preparation of NaA-ZNP granules

Two g of powdered zeolite nanoparticles (ZNPs) were suspended in 5 mL distilled water with the necessary amount of clay, to which sodium alginate gel 2 wt % was added. The slurry was mixed with a magnetic stirrer. In next step, sodium alginate (2 g, 2 wt %) to 98 g of water was added and then stirred while being heated to a temperature of 90 °C. The sample changed to thicken and bind the mixture. So a gel had produced, the gel was used in next section. After the preparation of the gel, the solution was left sitting for 1 h to allow any trapped air bubbles to be released. The masses of sodium alginate was separated by centrifugation at 2500 rpm for 15 min. The solution was mixed into a 1 L tank filled with a 400 mL barium chloride (0.1 M) which was being stirred. The resulting granules continued to be mixed mildly in the barium chloride solution for 2 h and were then repeatedly flushed with distilled water. The washed granules were immersed in acetone for 1 h and the mixture was left for the water to drain. The granules were then dried in open air for 24 h. The nanozeolite granules were then placed in a furnace for 5 h at 500 °C in order to burn off the alginate (Esmaeili and Saremnia, 2016; Saremnia et al., 2015; West and Harwell, 1992). Fig. 1 show a diagram of the various involved synthesies of NaA-ZNPs in the comparison study of adsorption and nanofiltration methods for removal of total petroleum hydrocarbons from oil-field wastewater.

2.3. Surface modification of absorbents

Surface modification of the granular NaA-ZNP absorbents was accomplished using a cationic surfactant at a concentration of 200 mmoL per kg of zeolite in the NaA-ZNP. Three g of sodium zeolite in the NaA-ZNP was combined with 100 mL of surfactant solution in a 125 mL polyethylene container. The container was slowly shaken at 120 rpm in a screen shaker for 48 h. The samples were filtered and washed with a large amount of distilled water until the outlet water no longer foamed. The adsorbent was then dried for 72 h in open air, placed in watertight polyethylene bottles, and refrigerated (Esmaeili and Saremnia, 2016).

The GC-MS of the concentrated wastewater generated by the oil refinery in Bushehr registered the debi entrance input of the system as 17.26 mL/min (Fig. 2a).

2.4. Batch experiments for adsorption process

2.4.1. Contact time

To study the effect of time in the batch process, 2 g adsorbent was added to 50 mL of wastewater at neutral pH and ambient temperature, and the reactor was placed on a shaker at 300 rpm. TPH values for different time for batch experiments for adsorption process show in Table 1. At time intervals ranging from 5 to 80 min, the wastewater samples were removed and the adsorbent separated out by centrifugation. The samples were then evaluated using GC-MS (Esmaeili and Loghmani, 2016; Esmaeili and Saremnia, 2016).

2.4.2. Adsorbent dosage

To study the effect of sorbent dosage in the batch process, samples of the adsorbent in increments of 0.25 g from 0.25 to 1.25 g were added to 10 mL of waste oil with neutral pH. The samples were shaken at 300 rpm for 20 min at room temperature and the adsorbent separated out by centrifugation. The samples were then evaluated using GC-MS (Esmaeili and Loghmani, 2016; Esmaeili and Saremnia, 2016).

2.4.3. pH assay

The pH of the solution is a critical aspect of absorption and can modify the chemical properties influencing the affinity of the sorbent materials. In the absorption process, the solution pH affects the degree of ionization and the formation of different types of contaminants and results in varied reaction kinetics and equilibrium properties. To measure the effects of pH on the removal of pollutants, hydrochloric acid solution 0.1 M and sodium hydroxide 0.1 M were used to adjust the pH of the wastewater to a range of 3–11 in increments of 2. For each 10 mL sample of effluent at a determined pH, 0.5 g of 10 mL adsorbent was added. The samples were placed on the shaker for 20 min at 300 rpm and 25 °C. The adsorbent was then removed from the wastewater via centrifugation (2500 rpm) and the samples were analyzed by GC-MS (Esmaeili and Loghmani, 2016; Esmaeili and Saremnia, 2016).

The statistical significance of values was assessed using one-way ANOVA, with data of significance level of 0.05 being considered in this study.

2.5. Continuous experiments for adsorption process

In order to evaluate the conditions and fully utilize the adsorbent to achieve minimal concentration of pollutants in the waste, a 3 cm × 20 cm Pyrex glass column was used as an ascending-flow fixed bed reactor. In this section used a pump for transfer waste water. The flow going up, according to Fig. 1. Fiberglass was used to plug both ends to block escape of NaA-ZNP granules. In order to study the behavior of the column, the height and input flow rate parameters were observed. Fig. 1 depicts the column (Esmaeili and Loghmani, 2016; Esmaeili and Saremnia, 2016). The length of the column was varied between 5, 15, and 20 cm by adding glass beads. For each column length, 5 g of adsorbent mass was placed in the column. In other research, we investigated of amount of adsorbent instead increase the length of the column. The efficiency was not high for removal of wastewater (Esmaeili and Beni, 2014a).

Distilled water was pumped into the column for 10 min. After thoroughly washing the column in this manner, an inlet hose was used to connect to the waste tank, and wastewater at pH = 3 was fed to the column at 17.26 mL/min, allowing the adsorbent particles to attract the contaminants and remove them from the solution. Samples were taken 20 min after the wastewater reached the bottom of the column bed and the adsorbent particles separated out by centrifugation at 2500 rpm for 10 min. GC-MS was used to evaluate the samples and determine the percentage of pollutants removed. In this study, a peristaltic pump was used to provide rates in terms of milliliters per minute. To determine the impact of flow rate on the behavior of the column, flow rates of 7.4, 12.3, and 17.26 mL/min were investigated (Esmaeili and Loghmani,