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ORIGINAL ARTICLE

# Removal of crude oil from aqueous solutions by natural adsorbents

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The pollution caused by the oil industries, refineries and tanker's uploading are one of the biggest challenges. since the recent accident with spreading oil spill on the surface of sea water, the need for environmentally friendly oil sorbents has intensified. The investigation deal with the sorption of crude oil by Nano perlite and Nano zeolite, as a mineral sorbent. Results showed at the maximum level of initial oil concentration, the sorption capacity of Nano perlite and Nano zeolite are 3.11 and 2.89 g/g respectively. It woud be worth mention that the sorption capacity of Nano perlite is higher than Nano zeolite. Brunauer-Emmett-Teller (BET) surface area and X-Ray diffraction analyses were performed to characterize the properties of Nano adsorbents.

Keywords: Oil pollution; oil sorbent; nano perlite; nano zeolite

## Introduction

Crude oil released to the marine environment through accidental spillage or drainage from land causes serious damage to the environment when oil spill released in to the environment, An effective decontamination and cleanup is necessary for protection of environment and human health (Nwadiogbu et al., 2016). Adsorbent materials are attractive for some application because of the possibility of collection and complete removal of the oil from oil spill site some properties of good adsorbents include hydrophobicity and oleophilicity, high uptake capacity, high rate of uptake, retention time, oil recovery from adsorbent, re-usability and biodegradability of the adsorbents (Adebajo et al., 2003). According to the complex nature of oil spills, they do not behave the same in the environment. some constituents are noted for their tendency to vaporize while others prefer to bind to solids, some oil materials are extremely unreactive while other interacts with light, so they have different toxicological effects on aquatic life and hence on humans (Rafeah et al., 2013) various conventional and advanced technologies have been used to treat and remediate contaminated water by oil spills to reduce the costs of current treatments. The technologies that are generally applied during the removal of oil spills usually include biological treatments, membrane filtration, or adsorption on to the activated carbon. In an earlier work a selected clay mineral used in the cement industries in Egypt, in its thermally and chemicaly treated forms, was used to study the sorption of oil spills from oil contaminated water at different condition parameters. In 1991, Stowe used expanded perlite or vermiculite or sand coated with an oleophilic/hydrophobic layer. In 1999, Ivanov treated oil spills by a composite adsorbent-alominosilicate material that was exposed to water repellent treatment (Sayed and Zayed, 2005). Gönen and önalan investigate the application of chitosan for the removal of procion MX-R dye from synthetic reactive dye wastewater. The results showed that the chitosan/dye interaction was feasible and chitosan could be a promising alternative for eliminating dyes from industrial wastewaters (Gönen, 2015). In this study, we report the sorption capacity of Expanded Nanoperlite and Nanozeolite which in fact we compare this two types of Nano adsorbents and we figure out which one is best for oil spill adsorption.

## **Experimental selection**

Nano perlite and Expanded Nano zeolite used in this study were provided from physical treatment of Expanded perlite and natural zeolite. First of all, they were washed by distilled water to remove impurities, then dried in vacum at 75 °C for 24 h, then dried samples were powdered in ball mill to obtain grain size of 100-150 nm these materials which mainly composed aluminum silicate which are effective in oil spills cleanup. The crude oil applied in this work which provided from Hengam oil field in south of Iran.

In table 1, the result of BET analyses were found 48.26 and 39.22  $m^2g^{-1}$  for Nano perlite and Nano zeolite, respectively. As seen from this table Nano perlite with lower bulk density and higher surface area makes it more appropriate in adsorption of oil when it's compared with Nanozeolite. In Table 2, the maximum adsorption capacity of Nano perlite and Nanozeolite regarding their surface area is compared with other adsorbents reported in the literature.

As we can see in Table 2, The adsorbents with high surface area have a good oil sorption capacity.

Nano zeolite

100-160

39.22

Table 1. The Results of BET analyses for two type of different nano adsorbents.					
Adsorbent Type	Particle size(nm)	Bulk density(gcm <sup>-3</sup> )	BET surface area(m <sup>2</sup> g <sup>-1</sup> )		
Nano perlite	100-160	<0.12	48.26		

Table 2. Reported Surface area o	f natural adsorbents oil sorbent	previous study (Rafeah Wahi et al, 2013).

Sorbent	Surface area (m <sup>2</sup> g <sup>-1</sup> )	Sorption capacity (gg <sup>-1</sup> )
Salvinia cucullata	218	Vaseline:
		3.6-7.3
		Marlin Oil:
		4.8-11.6
Walnut Shell	0.17	Mineral Oil:0.6
		Canola Oil:0.6
		Cutting Oil:0.8
Rice Husk	240	Gasoline:3.7
		Diesol Oil:5.5
		Light Crude Oil:6
Nano perlite	48.26	Hengam oil:3.11
Nano zeolite	39.22	Hengam oil:2.89

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X-Ray Diffraction studies (Model PTS 3003, SELFERT, Germany, X-Ray diffraction) with scanning limited  $2\Theta$  =5-60 in Nano zeolite and  $2\Theta$  =5-90 in Expanded Nano perlite were carried out in order to assign the present significant phases whitin Nano adsorbents structure.

The broad powder X-Ray diffraction pattern of Nano perlite (Figure 1), revealed that the heating of perlite at over 800 °C could be convert less ordered structure in unexpanded perlite to a more highly ordered structure in expanded one and single crystalline peak at  $2\Theta = 27.642^{\circ}$  which shows presence of quartz in the sample (Jain et al., 2011) presence apeak at  $2\Theta = 22.23^{\circ}$  confirming amorphous nature of silica (Javeed et al., 2010; Amutha et al., 2010).

The result of XRD analyse of Nano zeolite (Figure 2) showed that the clinoptilolite is the main mineral in the sample and content of, mordenite and heulandite are low. In fact sharp diffraction that could be attributed to clinoptilolite (JCPDS files Card Nos.025-1349, 2000).



Figure 1. X-ray diffraction pattern of Expanded Nano perlite.



Figure 2. X-ray diffraction pattern of Nano zeolite.

Oil sorption capacity of Nano adsorbents were measured using method based on the ASTM standard F726-06 test method for the sorbent performance of adsorbents (ASTM Satandard F726-06, 2006).

At the begeining of this stage, adesired amount of each Nano adsorbents (1 g) and 40 g oil with 100 ml sea water of Persian gulf were placed in a glass beaker separately. The entire system of each Nano adsorbent was placed over a shaker table and was shaken at 75 rpm for 15 min to creat a dynamic environment. After that the oil-soaked sorbent in the mesh was removed and drained for a minimum of 2 minutes. After the removal of excess oil that had adhered to the surface of Nano adsorbents, the samples were transferred to a weighing balance and the samples weight was recorded. It is worth to mention that the weight of the adsorbed water was measured by Karl Fischer technique as well described in ASTM D1533 (ASTM, D1533-00, 2005). The crude oil sorbed was obtained as shown in eq1:

#### (1) qe= $C_e$ - $C_w$ - $C_a$

Where  $C_e$  is the total weight (g) of oil, water and adsorbent material at equilibrium condition which was measured gravimetrically.  $C_w$  is the water weight (g) as determined by the Karl Fischer Technique and  $C_A$  is weight of the nano adsorbents.

### **Results and discussion**

To investigate the effect of the amount of initial oil concentration on sorption capacity, amount of oil from 10 to 40 g with 1 g of each Nano adsorbent in 100 ml seawater was tested. Results showed that when the initial oil concentration increases, sorption capacity increases, In fact, at high oil concentration, oil occupies the sorbent surface thus saturation is reached much faster, as seen in Figure 3, the sorption capacity increases till reach maximum at 3.11 and 2.89 g/g for Nano perlite and Nano zeolite, respectively.



Figure 3. Effect of initial oil concentration on sorption capacity of nano perlite and nano zeolite.

## Conclusion

Results obtained in this present investigation approved that silica is a main constituent of Nano perlite. Also the result of XRD analyses of natural zeolite confirmed that natural clinoptilolite is the most important mineral in the sample. The amount of oil sorbed on to Nano adsorbents was measured using dynamic method. The result of oil sorbed on to the adsorbents could be changed by initial oil concentration. The BET surface area analyses showed that the Nano perlite has highly surface area in compared with Nano zeolite result in higher oil sorption.

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